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Net SOA formation from BB plumes in field studies, comparison to lab studies, and implications for global models

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DOE ASR/ARM PI Meeting
Tysons, VA 21-March-2018

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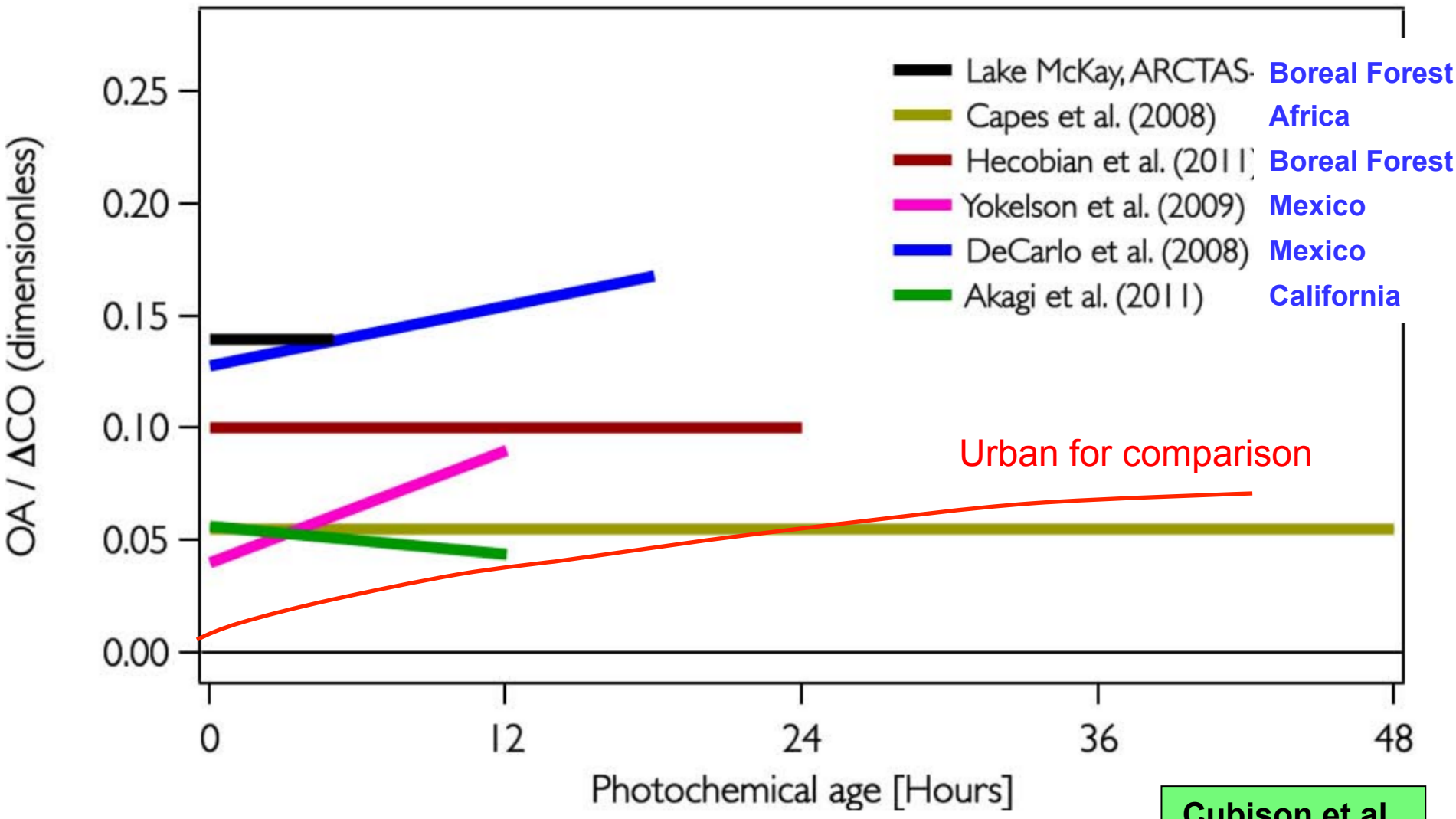
"Net SOA": added mass w/ BB aging is small



Cubison et al., ACP 2011

Chemical aging always observed!

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Net BB SOA in Field: 2017 update

- Out of the 17 published aircraft studies on aging of BB emissions, **10 reported no detectable net addition of OA mass with photochemical evolution, while 4 reported an increase and 3 reported a decrease** [*Hobbs et al.*, 2003; *Cubison et al.*, 2011; *Akagi et al.*, 2012; *Jolleys et al.*, 2012; *Forrister et al.*, 2015; *Jolleys et al.*, 2015; *May et al.*, 2015; *X. Liu et al.*, 2016].
- One mountain site saw **no change** [*Zhou et al.*, 2017]

Shrivastava et al., Rev. Geo. 2017

- Global modeling efforts should be consistent w/ field evidence!

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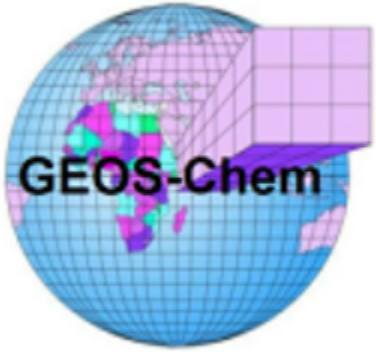
What to do in models? E.g. SIMPLE param. in GEOS-Chem v11



Page Discussion

Secondary organic aerosols

- Starting in GEOS-Chem v11-02, we will have an option for "simple" SOA that forms irreversibly. This option will allow GEOS-Chem users to get approximate the "correct" amount of global SOA without detailed chemistry.
- This scheme introduces two SOA-related tracers: SOAP (SOA precursor) and SOAS ("simple" SOA in the particle phase). The emission of SOAP is tied directly to emissions of […] biomass burning CO
- SOAP forms SOAS on a fixed timescale of 1 day.
- Biomass burning and biofuel: 0.013 g SOAP/(g CO emitted). As implemented in GEOS-Chem by Kim et al., 2015, based on field results from Cubison et al., 2011.

http://wiki.seas.harvard.edu/geos-chem/index.php/Secondary_organic_aerosols

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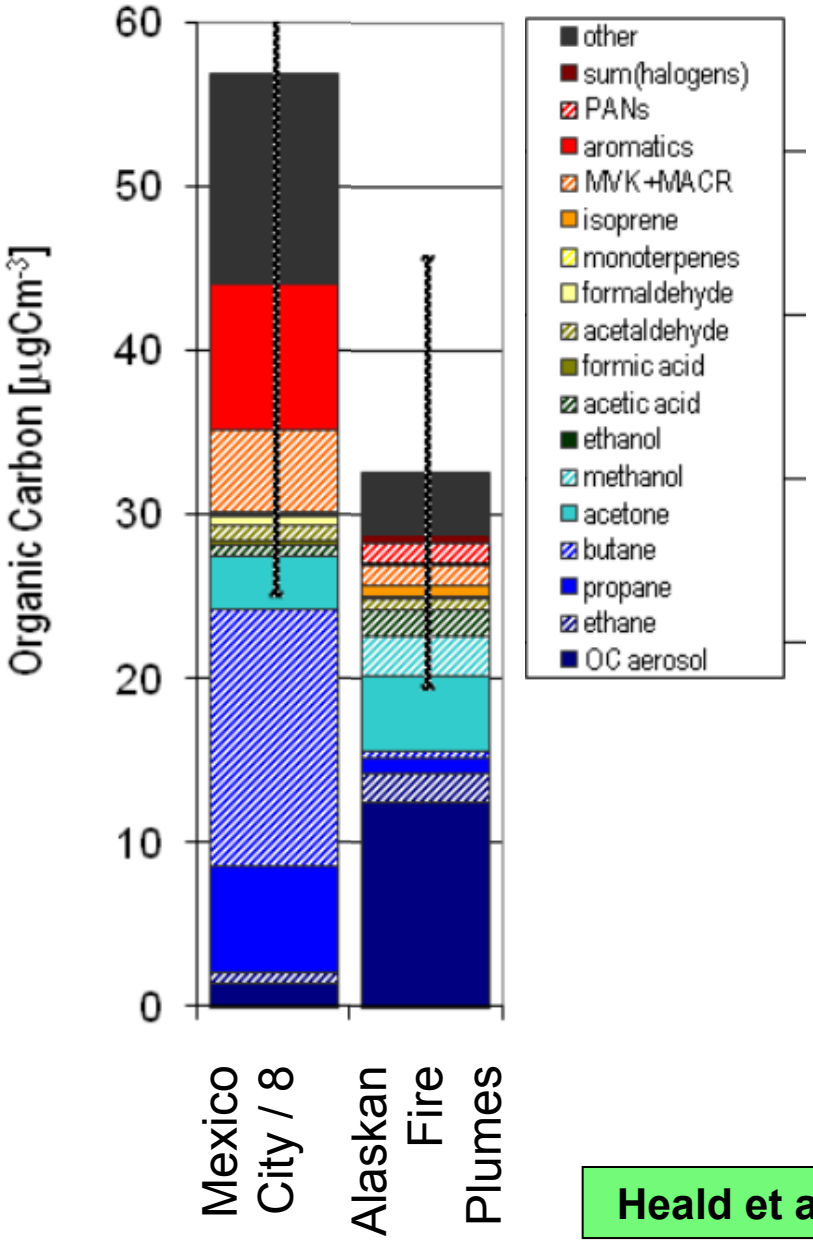
Why are fires different from urban?



- Gas/Particle OC
  - ~ 3 in BB smoke vs. ~50 in urban pollution
- Easy for SOA to compete with POA in urban, very difficult for fires
- Lots of room for POA evaporation in BB
- Makes sense that net SOA / POA is far larger in pollution than BB

Heald et al., ACP 2008

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Why can added SOA help models?

It cancels error of BB POA being too low!

Table 2. Summary of scaling factors applied in previous modelling studies to biomass burning emissions or modelled concentrations of biomass burning aerosol to match observations. Region abbreviations used in the table are defined in van der Werf et al. (2006): Northern Hemisphere South America (NHSA), Southern Hemisphere South America (SHSA), Northern Hemisphere Africa (NHAF), Southern Hemisphere Africa (SHAF), Southeast Asia including the Philippines (SEAS) and equatorial Asia (EQAS). See van der Werf et al. (2006, 2010) for discussion of differences between GFED versions 1, 2 and 3; on average GFED3 is 13 % lower than GFED2 van der Werf et al. (2010), with total GFED2 emissions lower than GFED1 in Central and South America and southern Africa (van der Werf et al., 2006).

Reference	Biomass burning emission inventory	Region of focus	Details of scaling applied	
Matichuk et al. (2008)	GFED2 (van der Werf et al., 2006)	South America	Smoke source function was scaled up by a factor of 6.	x6
Johnson et al. (2008)	Biomass burning emissions following Dentener et al. (2006): GFED1 (van der Werf et al., 2004) for year 2000 or a 5-year (1997–2001) average (not specified).	West Africa	Increased mass concentration of biomass burning AOD by a factor of 2.4.	x2.4
Chin et al. (2009)	Calculated using dry mass burned dataset from GFED2 (van der Werf et al., 2006).	Global	No scaling applied, but used emission factors of BC (1 g kg^{-1}) and OC (8 g kg^{-1}) that are 40–100 % higher than commonly used values (Andreae and Merlet, 2001).	x2
Sakaeda et al. (2011)	Aerosol fields taken from MATCH chemical transport model	Southern Africa.	OC and BC masses were increased by a factor of 2 over 10° N–30° S and 20° W–50° E.	x2
Johnston et al. (2012)	GFED2 (van der Werf et al., 2006)	Global	Scalar adjustments made for 14 continental-scale regions: NHSA (2.48–2.7), SHSA (1.9–3.3), NHAF (1.02–1.08), SHSA (1.68–2.01), SEAS (2.43–3.08), EQAS (2.3–2.72). Scaling factors were applied to modelled surface fire $PM_{2.5}$ to match satellite observations of AOD (non-fire aerosol was also scaled).	x3.3
Kaiser et al. (2012)	GFED3 and GFASv1.0	Global	Model was biased low in South America and Africa by factors of 4.1 and 3.0. Recommended a global enhancement of 3.4 for PM emissions from fires.	x3.4
Ward et al. (2012)	Calculated from Kloster et al. (2010, 2012) CLM3 simulations of global fire area burned, using emission factors from Andreae and Merlet (2001) and updates from Hoelzemann et al. (2004). Compared against GFED2.	Global	Scalar adjustments were made for continental-scale regions following Johnston et al. (2012) with slight modifications: SHSA (2.0), NHAF (1.0), SHAF (3.0), SEAS (1.5), EQAS (3.0). Scaling factor directly applied to model fire emissions.	x3
Tosca et al. (2013)	GFED3	Global	Biomass burning BC and OC emissions scaled by factor of 2 globally with additional regional scaling factors applied: South America (2.4), Africa (2.1), Southeast Asia (1.67).	x2
Marlier et al. (2013)	GFED3	Southeast Asia	Total aerosol burden scaled by 1.02–1.96 (depending on model), with additional scaling factors of 1.36–2.26 applied to fire aerosol.	x2.3
				x3.4

Reddington et al (2016)

Reddington et al., ACP 2016

- Fire inventories generally biased low

- Uncertainties in BB POA are far larger than those on net BB SOA production factor!

- Also large uncertainties with e.g. injection height etc.

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Recommendations

- Models:
  - It is great to try new BB SOA schemes, but… not ok to implement a scheme that makes a lot of net BB SOA and treat it as a real global option (given current understanding)
  - Always test the net SOA formation under typical conditions in your model
  - Compare with the SIMPLE scheme (Cubison et al., 2011, or as implemented in GEOS-Chem v11)
- Experimentalists:
  - Understand why SOA formation in the lab is higher (walls + tubing?)
- Everyone:
  - Better understand the fundamentals so that we can implement more realistic param. in models (incl. aging)

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Lab Studies: high net SOA

Atmos. Chem. Phys., 9, 1263–1277, 2009
www.atmos-chem-phys.net/9/1263/2009/

Atmospheric Chemistry and Physics

Laboratory investigation of photochemical oxidation of organic aerosol from wood fires 1: measurement and simulation of organic aerosol evolution

A. P. Grieshop, J. M. Logue, N. M. Donahue, and A. L. Robinson
Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, Pennsylvania, USA

- Photochemical oxidation produced **substantial new OA, increasing concentrations by a factor of 1.5 to 2.8** after several hours of exposure to typical summertime hydroxyl radical (OH) concentrations
- This and multiple subsequent lab studies generally have seen more net SOA than field studies
  - Including our own work (Ortega et al., 2013)
  - Why?

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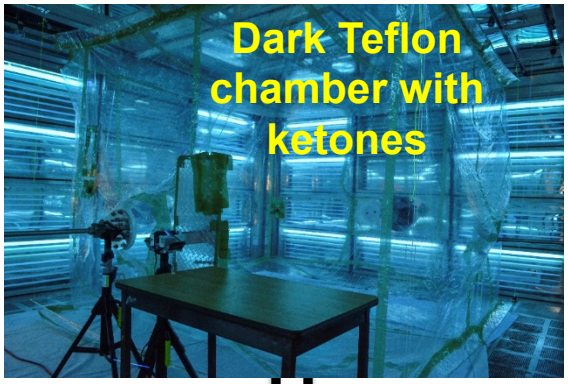
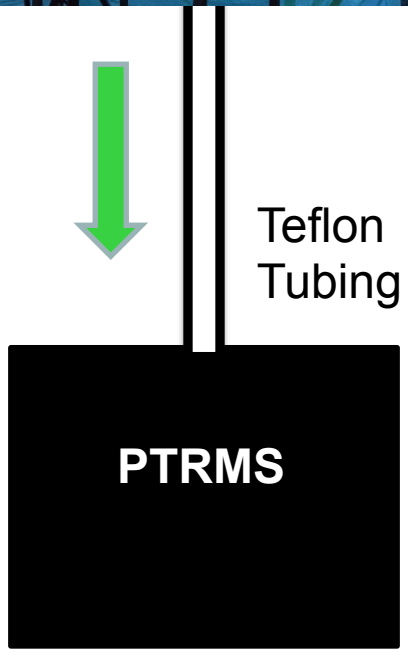
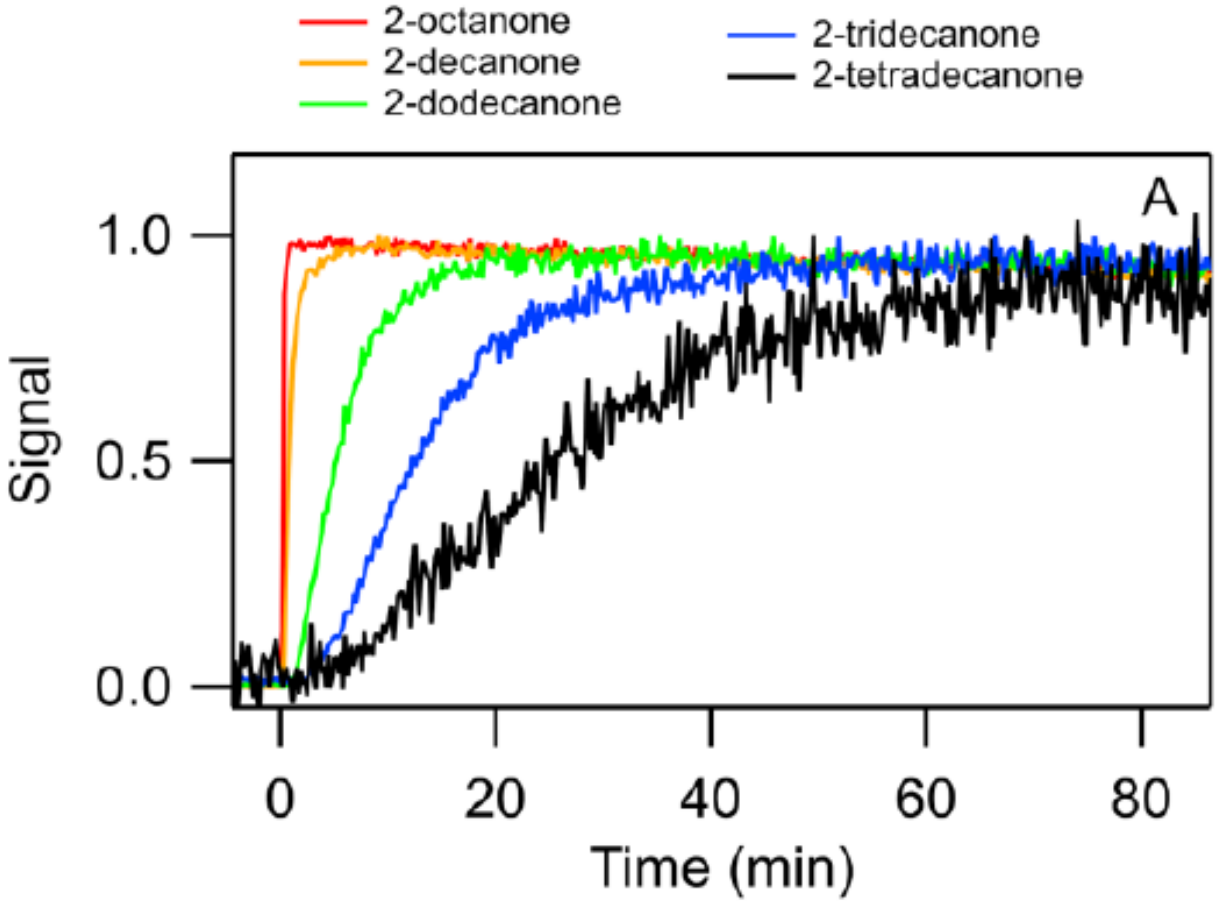
Gas Tubing Delays vs. C*







- Some delay from PTRMS, but major delay from tubing, increases x10 for each decade in c*

Pagonis et al., AMT 2017

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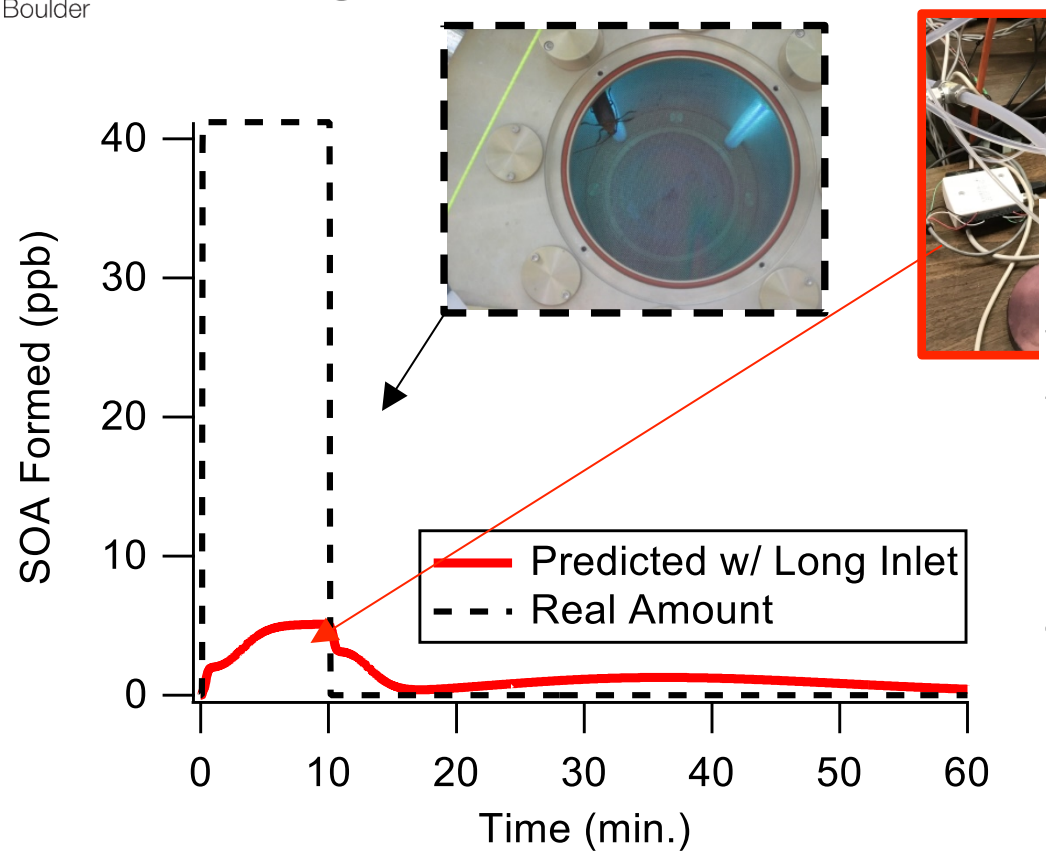
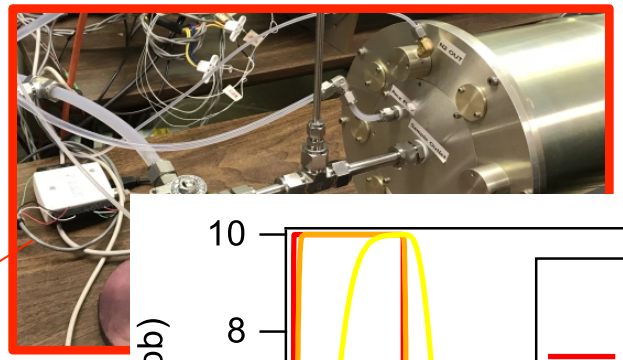
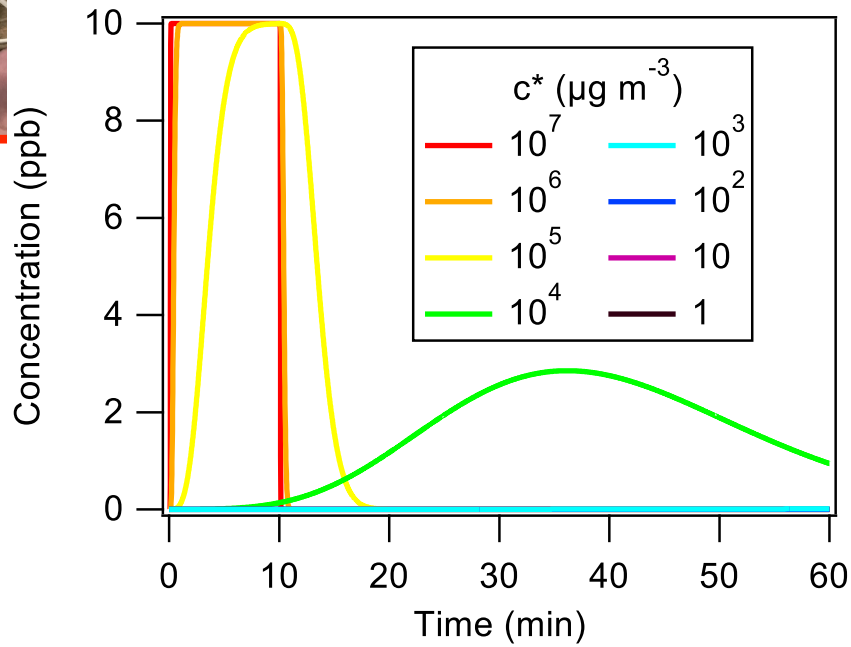
Large perturbation of exp. by surfaces







- 10 m inlet, 2 lpm (similar to recent study)
- Really short inlet: 1 ft, 2 lpm: Still lose ~1/2 of SOA potential!
- Heating can help a little but does not solve the problem
- *The point is that experimental system can majorly perturb results*

Palm et al. ACP 2018 (SI)

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BACKUP SLIDES

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Net BB-SOA in the Global OA Budget

- Global OA budget ~ 180 Tg OA yr^{-1}
  - Hallquist et al. 2009; Heald et al., 2010; Spracklen et al., 2011

$\Delta OA^{aging}_{global} = \left(\frac{\Delta OA_{aging}}{POA} \right) x\, POA_{global}$	$\Delta OA^{aging}_{global} = \left(\frac{\Delta OA_{aging}}{\Delta CO} \right) x\, CO_{global}$
= **8 ± 7** (field)	= **7 ± 6 Tg OA yr^{-1}** (field)
= **10**	(CU PAM lab – Ortega et al., in prep.)
= **23**	(CMU teflon bag – Hennigan et al., 2011)

- **Potentially Important global source, 5% of global OA**
  - **But substantial error bars**
  - **Future:**
    - **Stratify for important types of BB**
    - **Refine with additional data**

Cubison et al. ACP 2011

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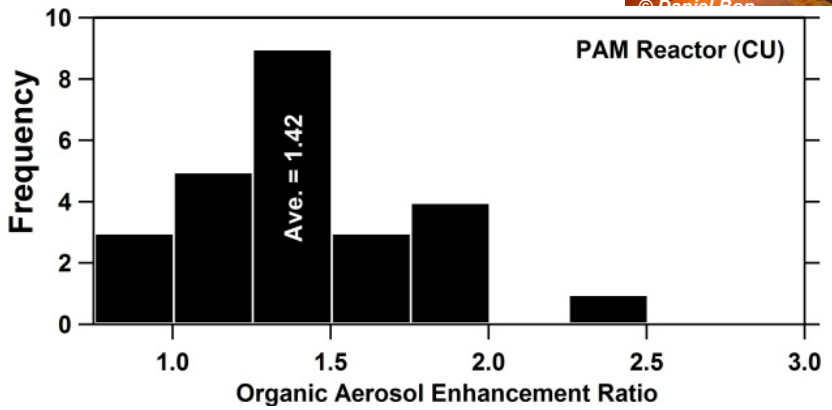
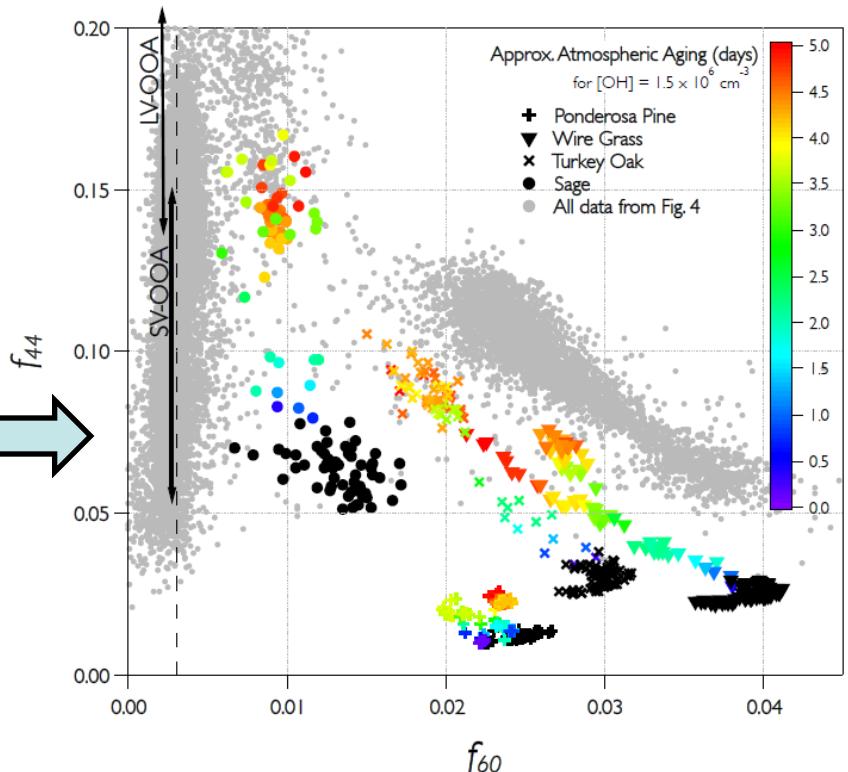
SOA Formation Potential: BB Smoke

- High variability
- Avg: aging smoke can produce net SOA mass
  - Laboratory avg. 42% of POA
  - Aircraft field avg. 20% (Cubison et al., 2011)
  - High diversity in both field and lab observations
- Using field data: aging of smoke contributes ~5% of total global OA source (~8 Tg yr^{-1})
- Chemical aerosol aging observed in all cases (lab & field)





Urban Air and Biomass Smoke