Improved treatment of atmospheric organic particulate matter concentrations from biomass combustion emissions

NCER STAR GRANT R833747

Research Team

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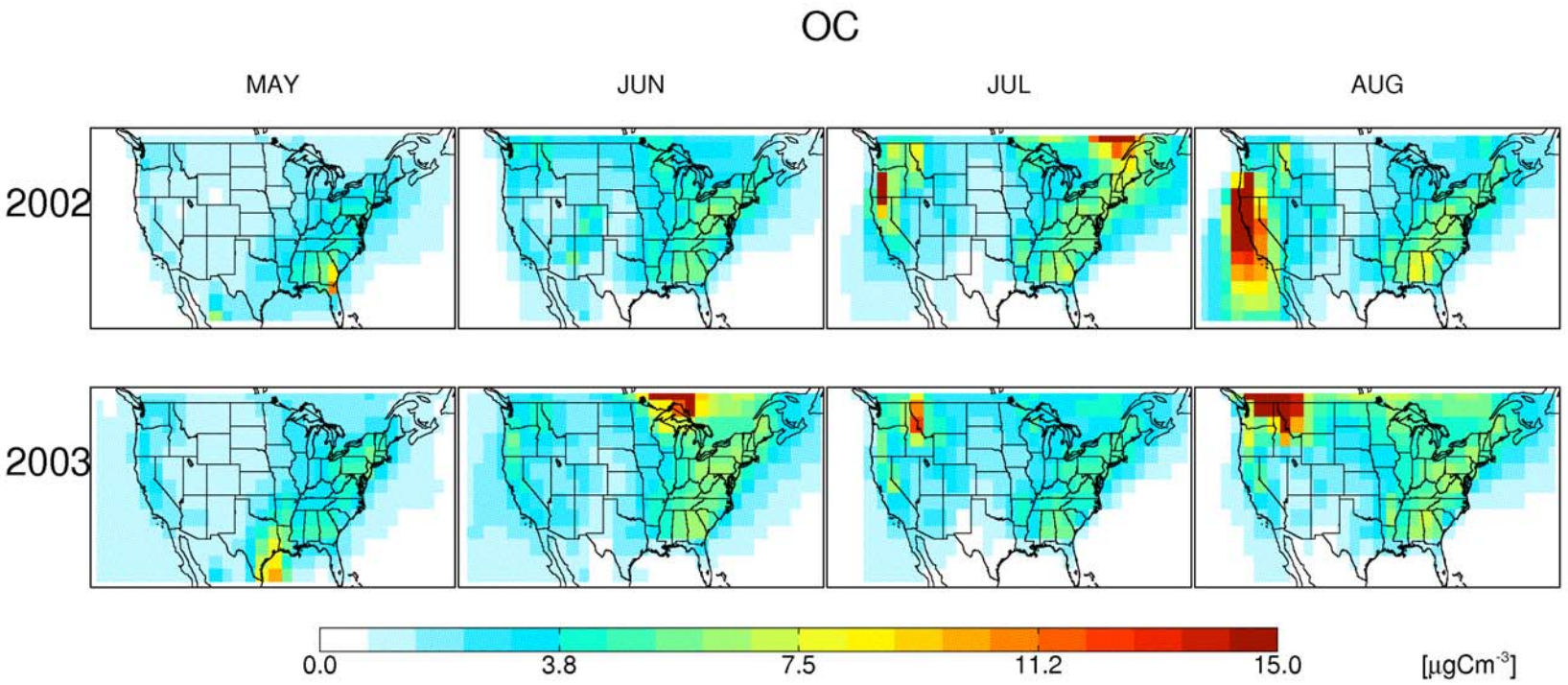
Premise

- Biomass burning (BB) in the U.S., from both wild and prescribed fires, is an important yet poorly-characterized source of organic aerosols
- Biomass burning emissions, like SOA, have constituents spanning a wide range of volatilities
  - Prior studies that developed source profiles are specific to the total aerosol mass concentrations used in those studies
    - In particular, aerosol "yields" at low mass concentrations may be biased, leading to model errors
    - Temperature dependence of volatilities have not been characterized
  - Models do not have emissions estimates for semivolatile species that may undergo oxidation in the atmosphere
    - May be one source of "missing carbon"
  - Stabilities of commonly-used BB tracers, like levoglucosan, against dilution / transport have not been unequivocally demonstrated

GEOS-Chem (current inventories)



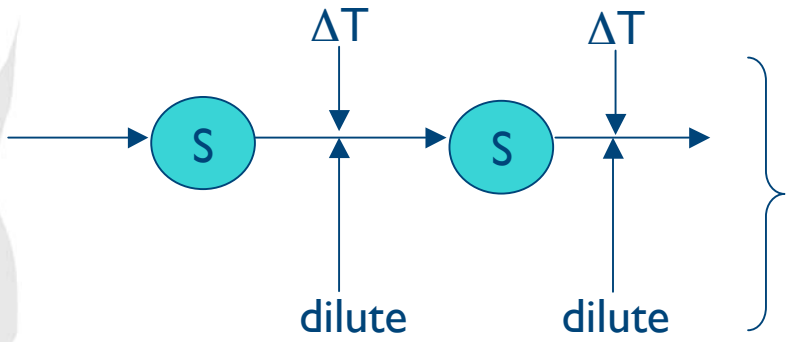
Fire is an important contribution to particulate organic carbon across the US

Project Objectives

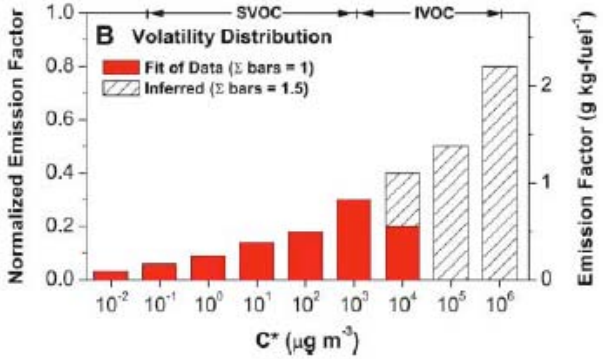
Study the role of biomass-burning emissions in U.S. air quality:

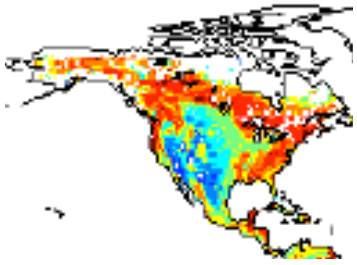
- Measure volatility distributions, as functions of both dilution and temperature, of open biomass burning emissions
  - test a variety of fuel types relevant to U.S. air quality
- Interpret data using semivolatile partitioning models
- Implement and test new biomass-burning emissions maps and partitioning models in large-scale model runs

Approach



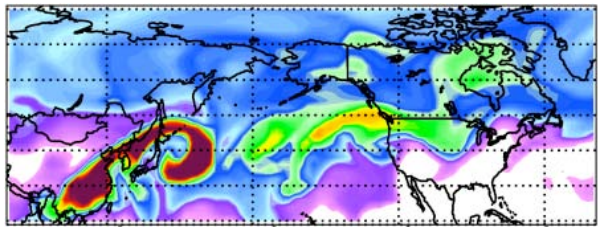
Robinson et al., *Science*, 2007





Van der Werf et al. (2006) BB emissions map

GEOS-Chem

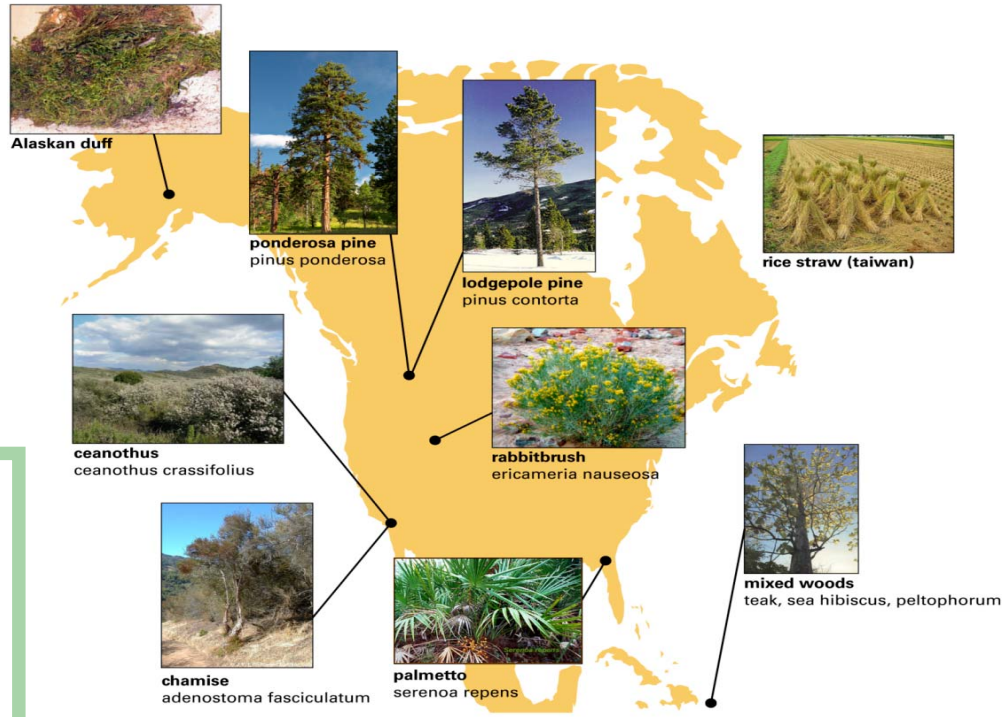


Fire Lab at Missoula Experiments (FLAME I & II, 2006 / 2007)

Year 1 activities focused on analysis of (limited) dilution and volatility data, to help plan Year 2 experiments at same facility

USFS / USDA Fire Sciences Lab
Missoula, MT
http://www.firelab.org/

- **Joint Fire Science Program**
- Physical, optical and chemical properties of open biomass burning emissions
- EFs, source profiles for FLMs
- Focus on W and SE US fuels



plant images courtesy santa monica mountains trails council, bay area hicker, alberta parks and recreation, daniel kirk, food and agriculture organization of the United Nations

FSL Burn Chamber

stack burns

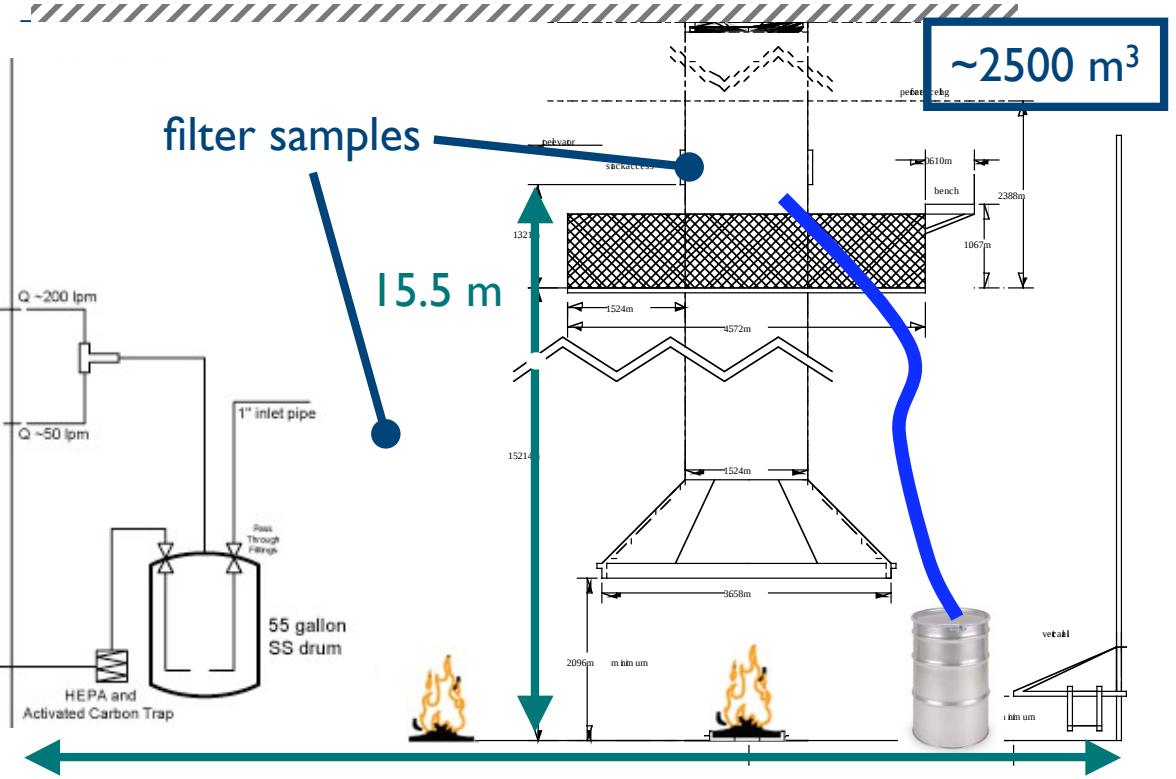
chamber burns

optical properties,
hygroscopicity,
CCN activity,
continuous OC/EC, ….

Particle and Gas Phase Instruments in Control Room

Particle and Gas Phase Instruments in Viewing Room

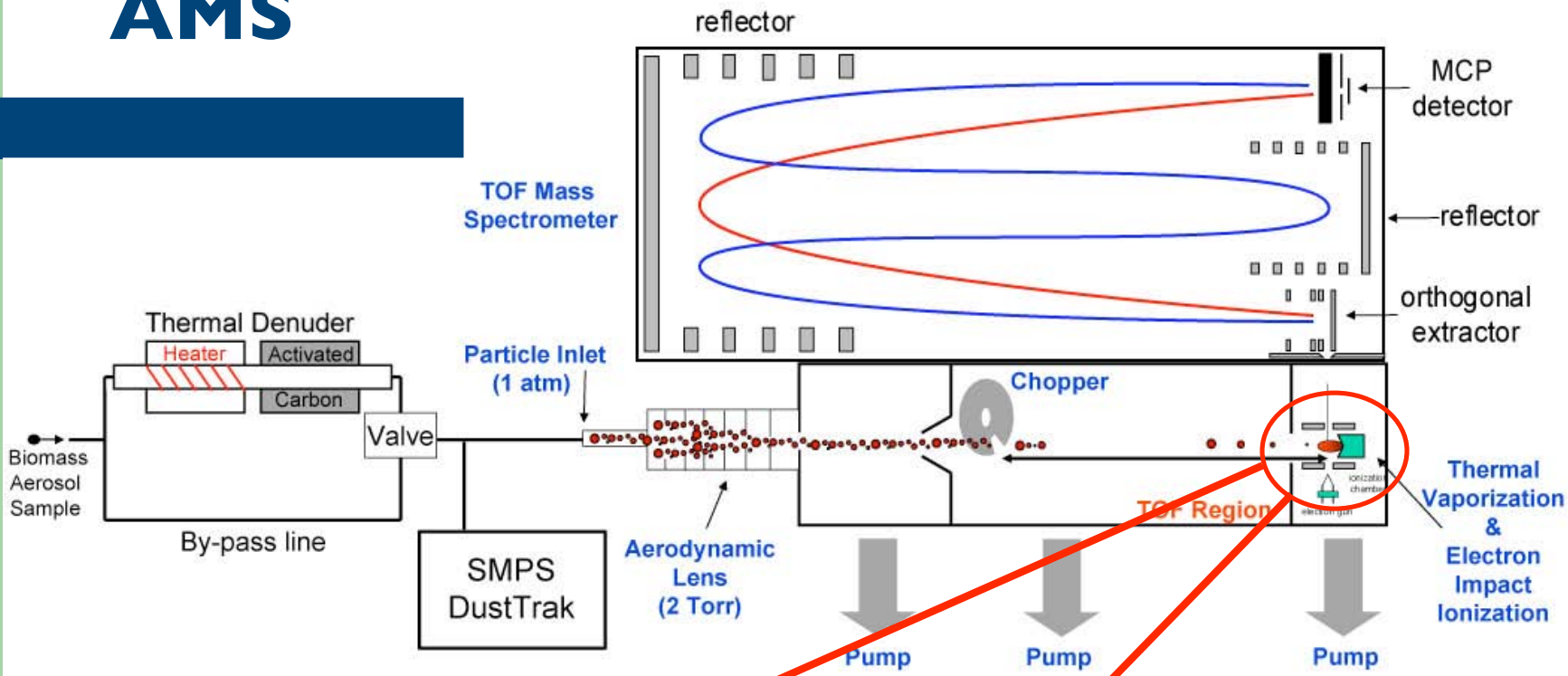
Multiple AMS, volatility AMS



256 burns
35 unique plant species
44 total fuels

Schematic of the FSL burn chamber, showing elevated platform and line and drum used for sampling during stack burns.

AMS



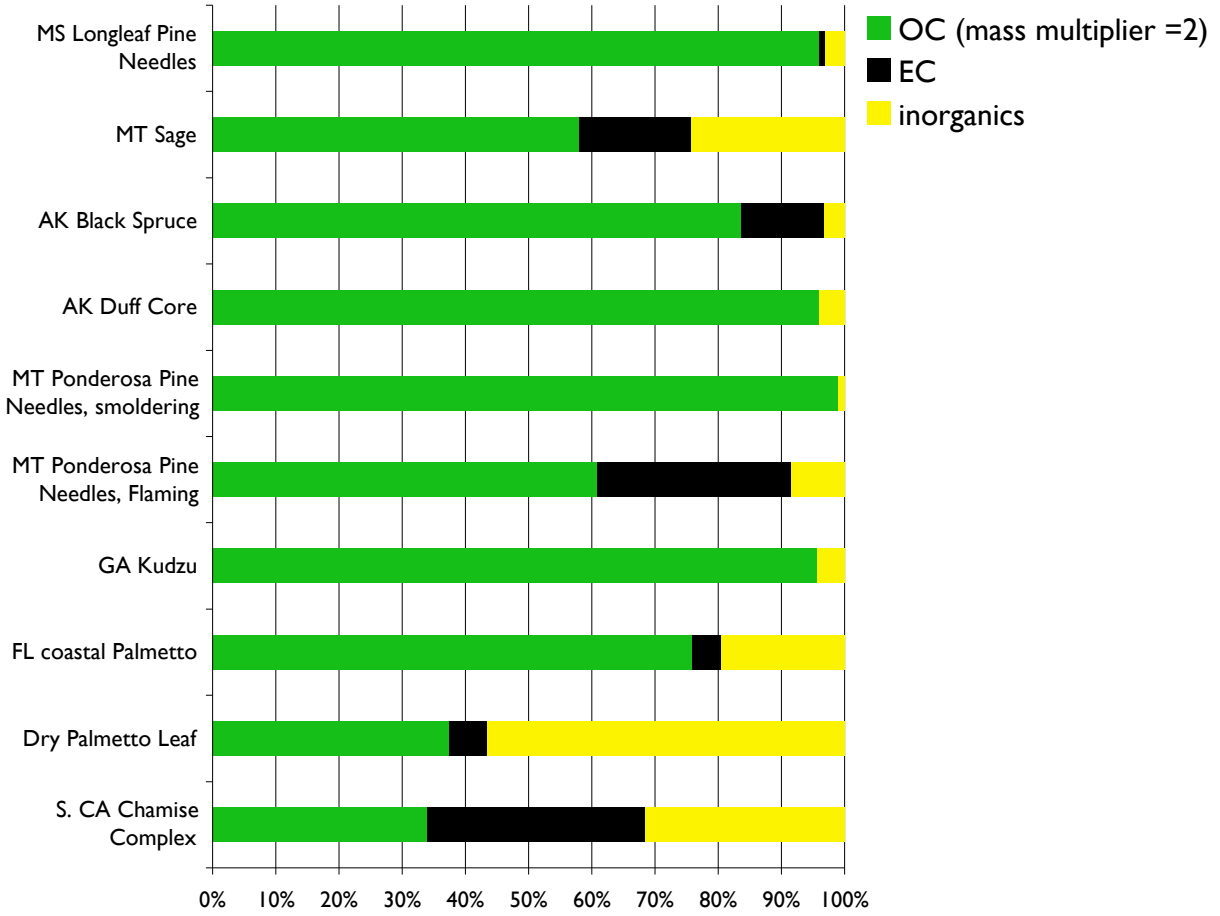
Thermal Vaporization & 70eV Electron Impact

Standard HR-AMS

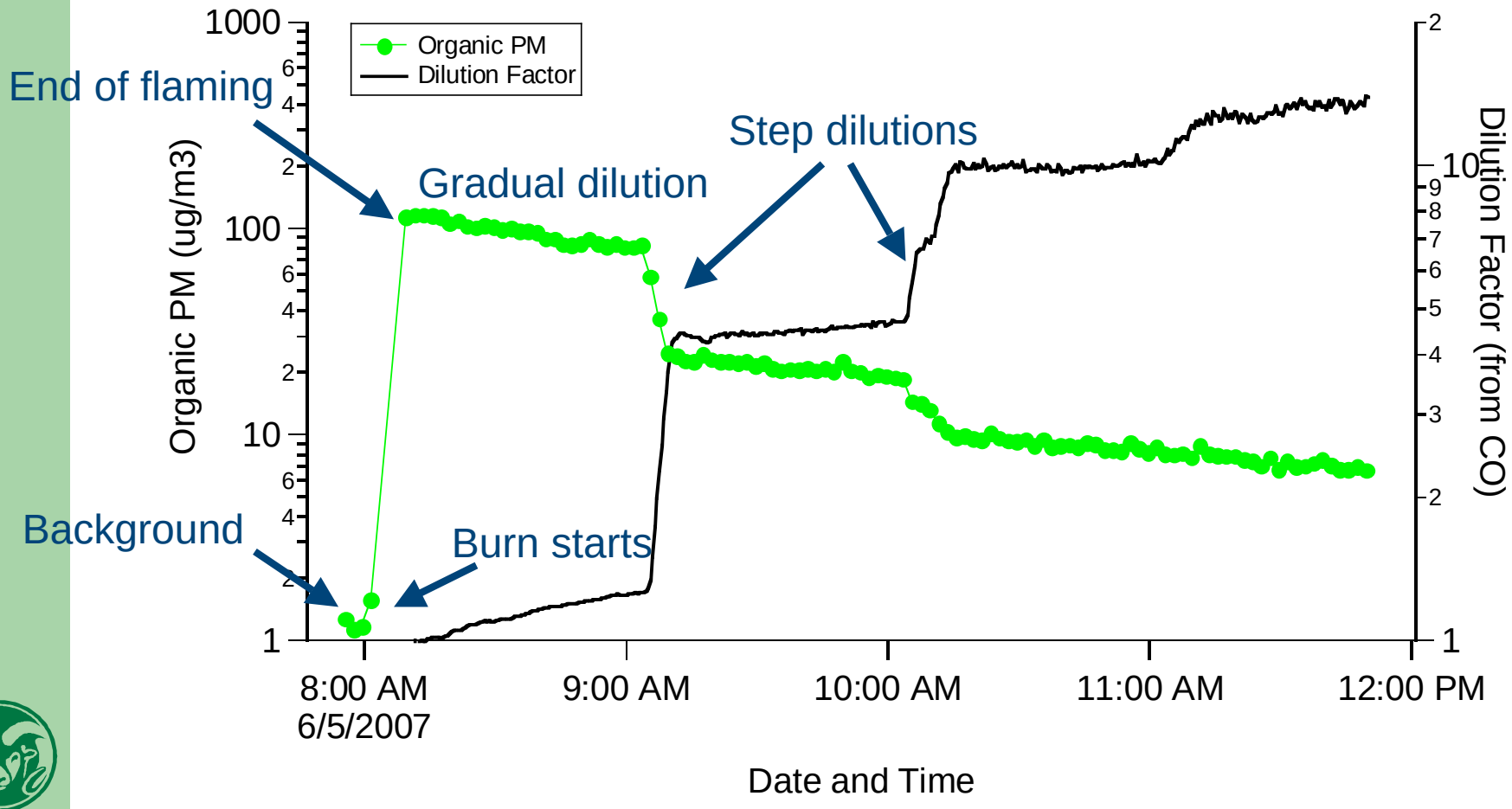
Thermal Vaporization & Li-ion Attachment

Soft-Ionization AMS

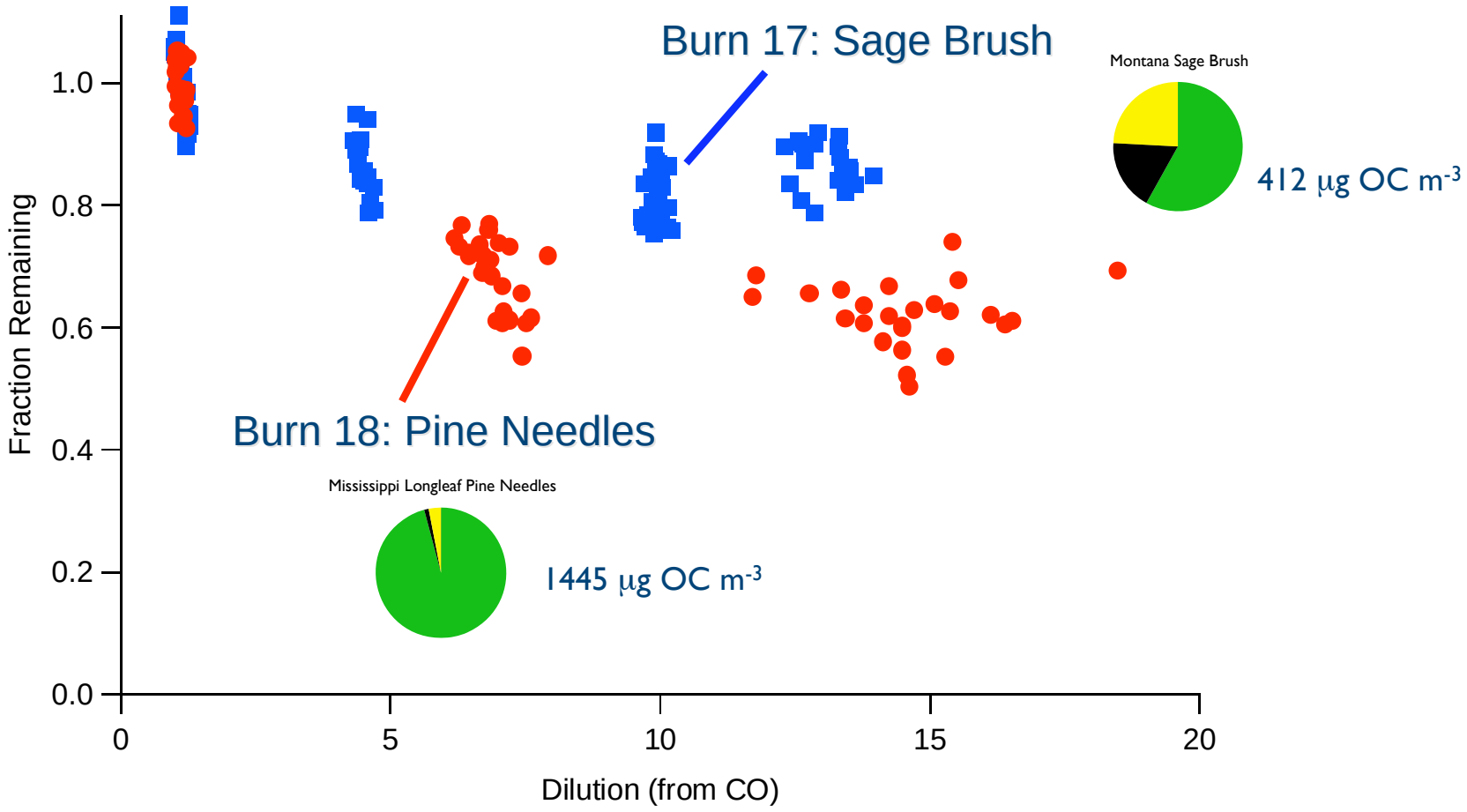
Example smoke compositions



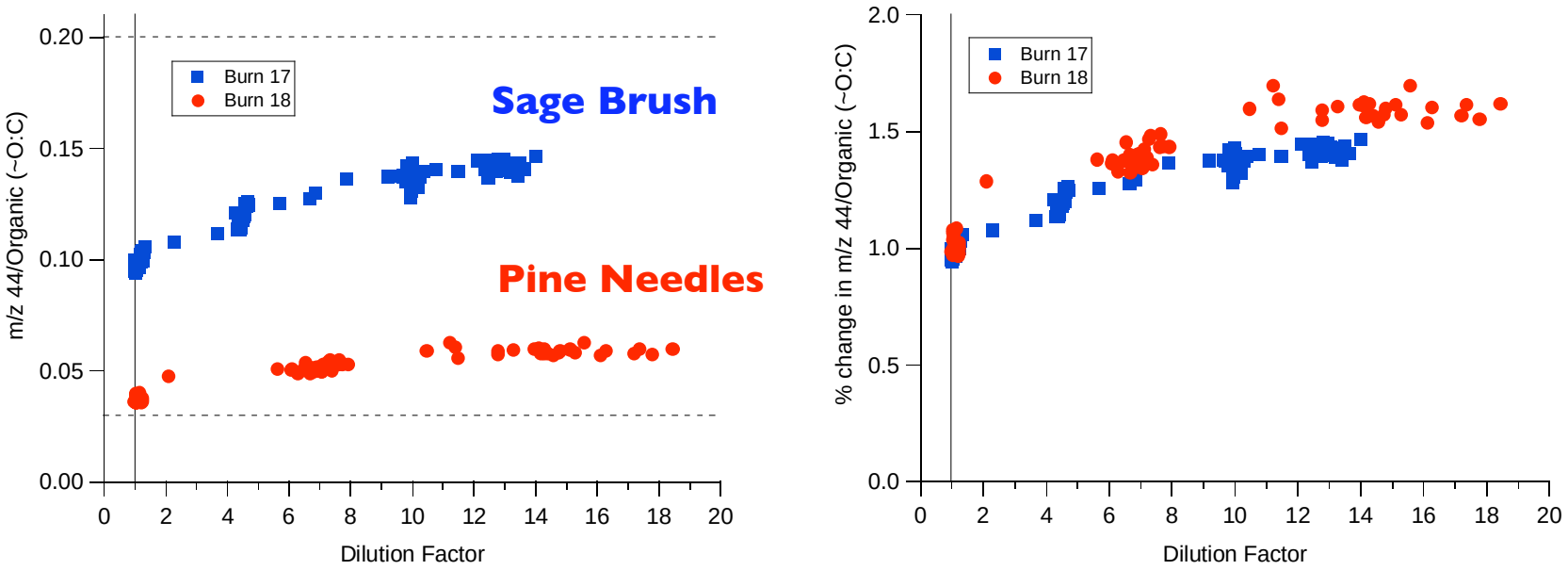
Dilution Experiments



Dilution Effects on Organic Fraction

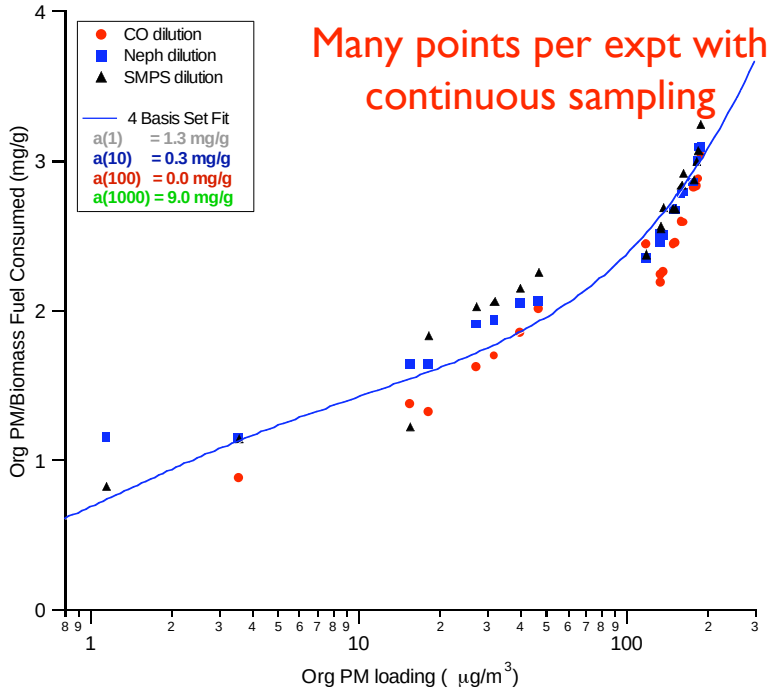


mz44/Org (~O:C) with dilution



Particles become more oxygenated with dilution

Example fit to basis functions



- Calculations used CO, nephelometer, and SMPS as corrections for particle losses due to dilution and deposition
- Data are fit with a four-parameter basis set:

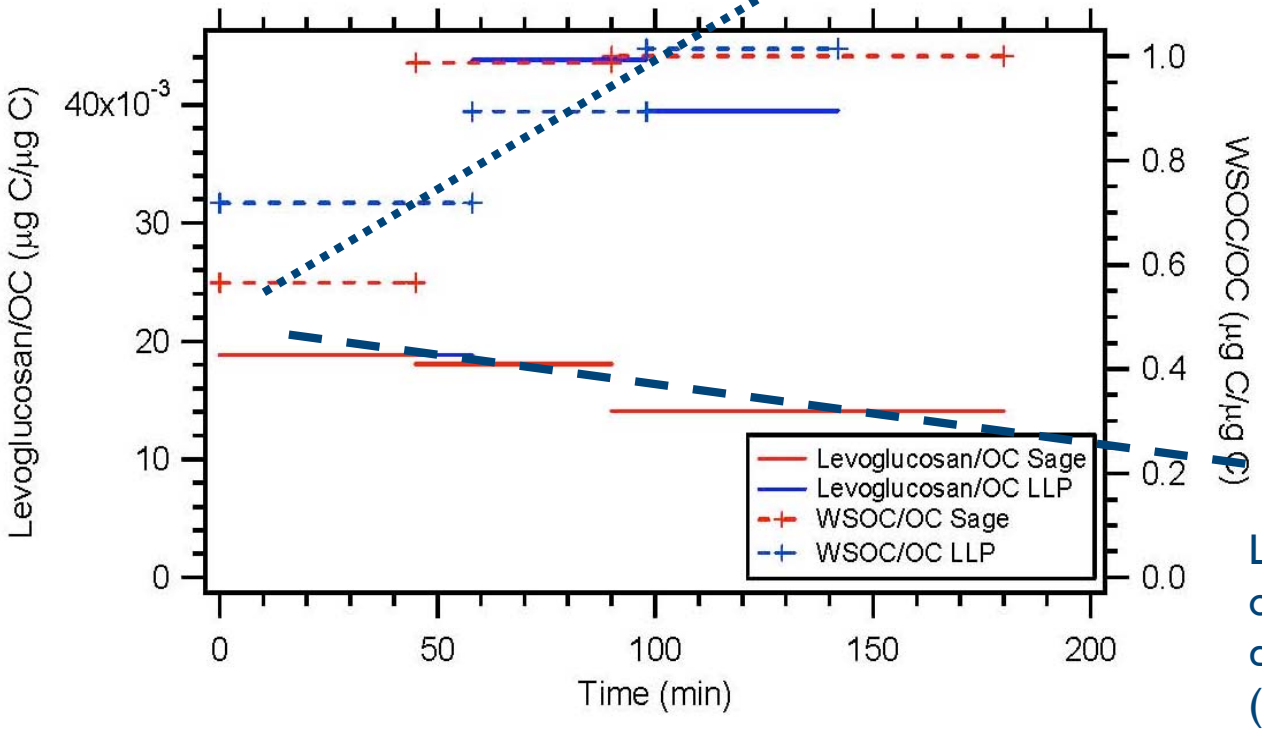
$$\xi = \sum_{i=1}^{n} C_i \xi_i$$

$$\xi_i = \left(1 + \frac{C_i^*}{C_{OA}}\right)^{-1}$$

- The resulting partitioning coefficients are shown in the legend.

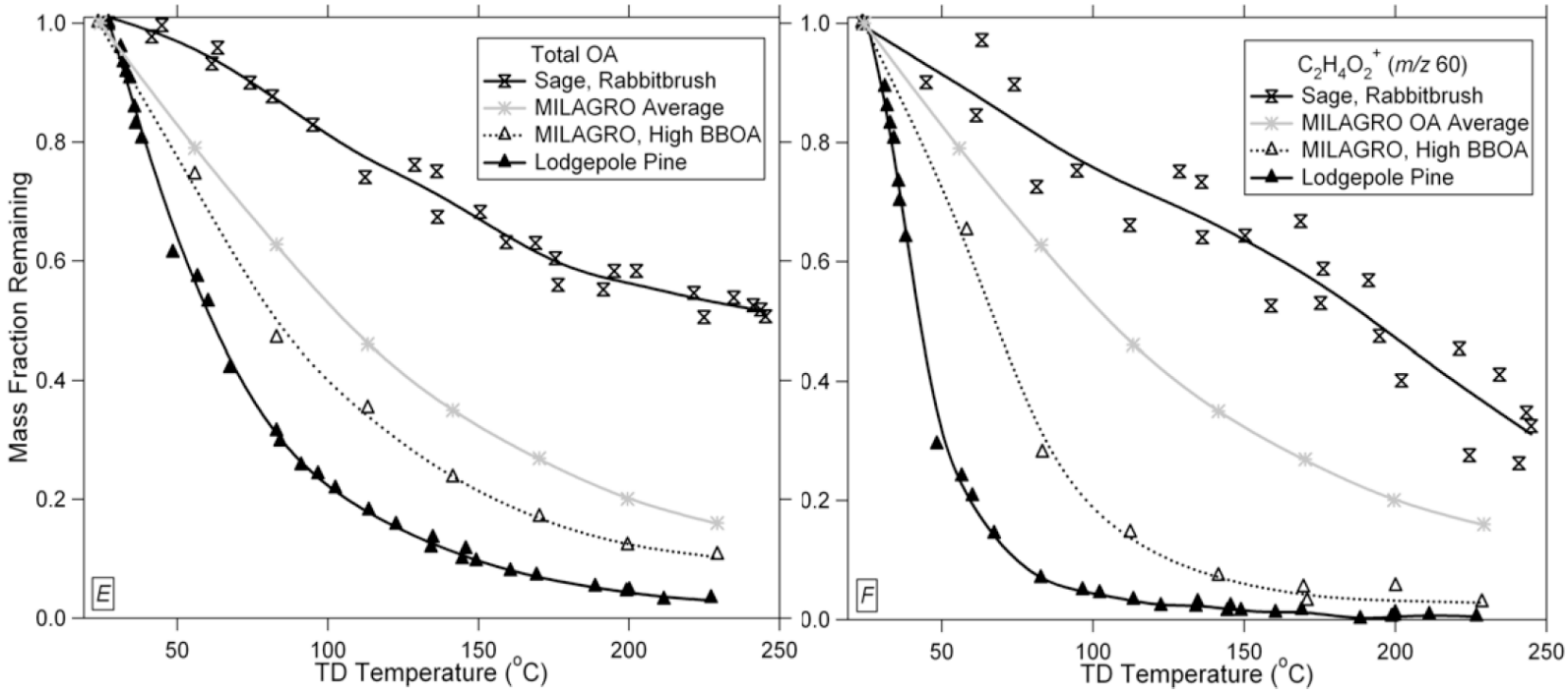
Molecular markers in sage and longleaf pine dilution experiments

WSOC as fraction of OC INCREASES with dilution (goes to 1 quickly in sage smoke)



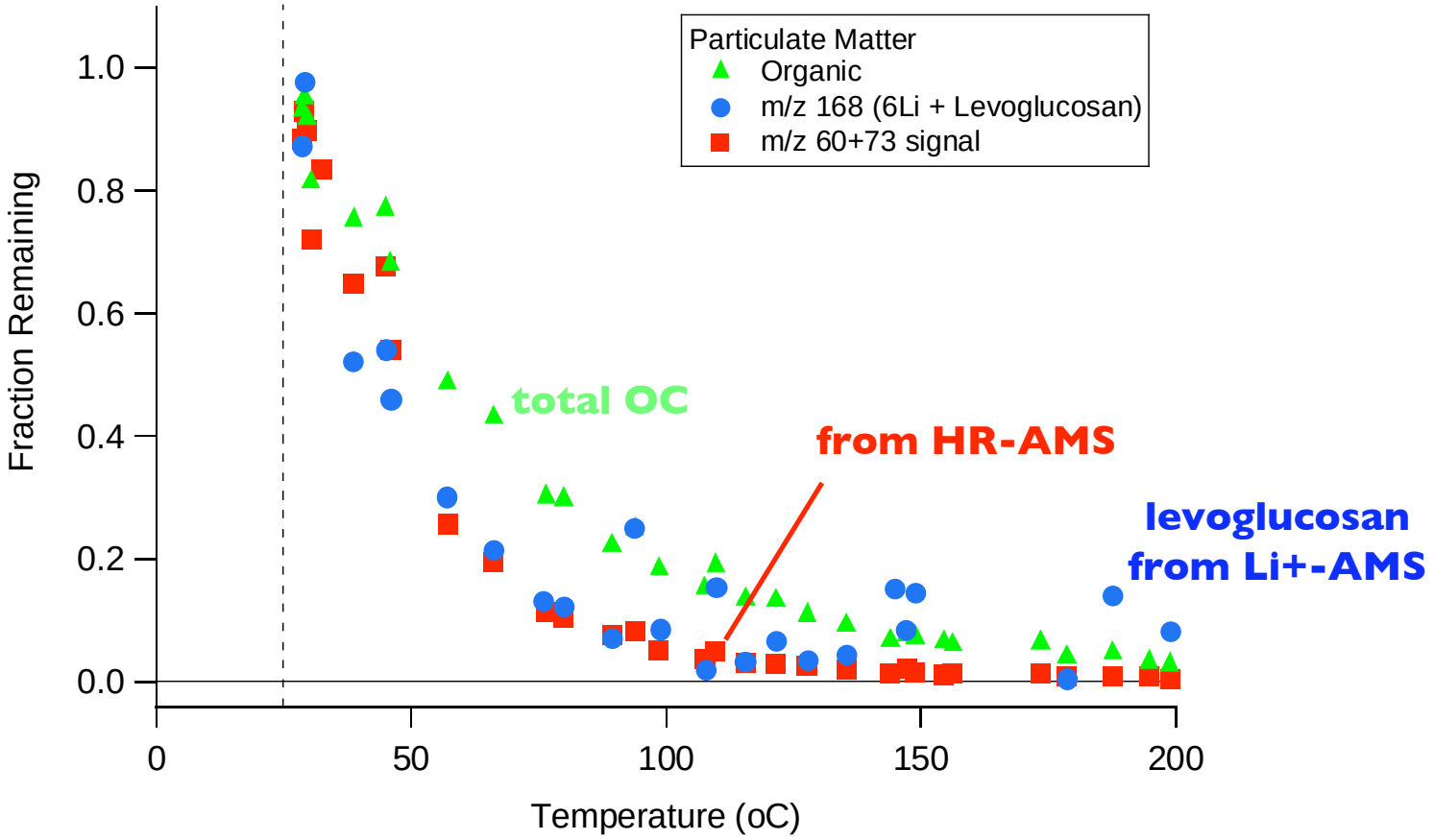
Levoglucosan as fraction of OC DECREASES with dilution in sage smoke (pine more complex?)

Volatility of BBOA and BBOA Tracer (*m/z* 60) during FLAME-1



[Huffman, Jimenez, et al., *ES&T*, under review]

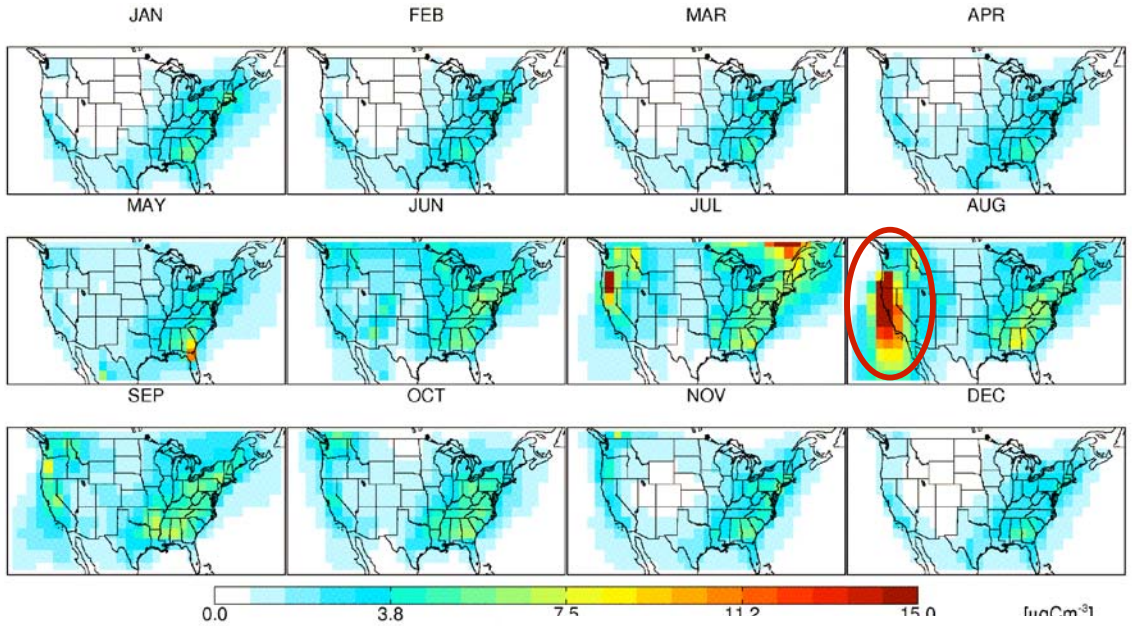
Volatility of molecular markers?



Plans for 2009 experiments

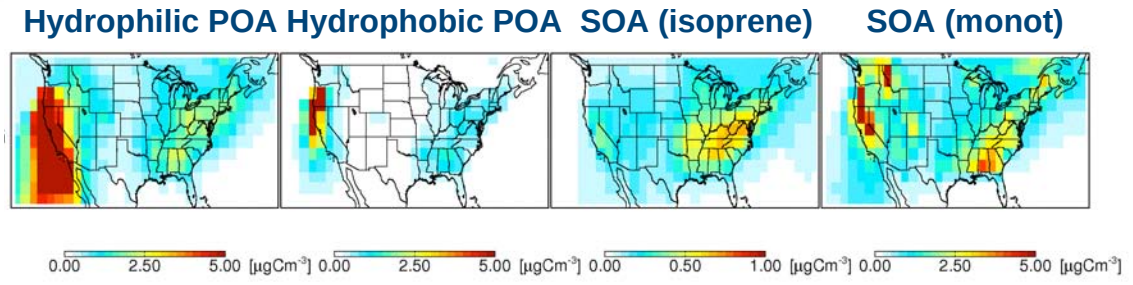
- Choose fuels that produce variable inorganic / organic and variable levoglucosan levels
- Stack burns for EFs
- Add measurement of total emitted SVOC + nonvolatile mass
  - Two channel system:
    - Ch 1, charcoal impregnated filter (CIF) to measure the total nonvolatile and semivolatile organic carbon
    - Ch 2, particulate OC only: carbon-coated diffusion denuder / quartz fiber filter / charcoal impregnated filter
- Design partitioning experiments to
  - cover several orders of magnitude in [OC], including to very low concentrations (~0.1 $\mu g\ m^{-3}$)
  - Cover range of temperatures, using thermal denuder
- Time-resolved measurements key new feature in chamber burns

Large fire events can dominate US OC surface concentrations



The fires in summer 2002 in Oregon led to the largest OC enhancements throughout the US that year (GEOS-Chem simulation)

Simulated OC type for Aug 2002:



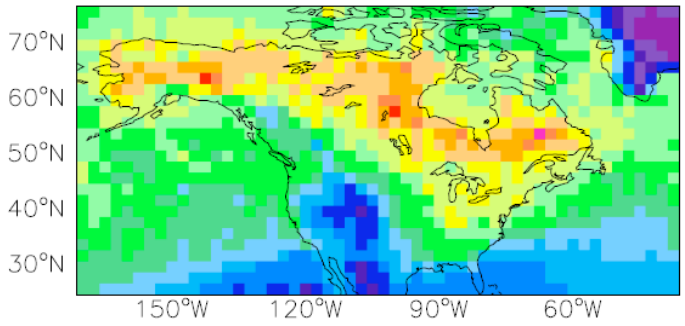
Plans for model development

- GEOS-Chem global 3-D CTM at 2°x2.5° resolution
  - Apply observed emission factors to GFED v2 (van der Werf, 2005) year-specific 8-day resolved biomass burning emission inventory for primary organics (lump to 3 fuel classes with similar partitioning characteristics) and SOA precursors (terpenes, aromatics)
  - Implement partitioning coefficients for each emission category following 2-product model SOA scheme in GEOS-Chem (3 new semi-volatile POA tracers, 2-3 new SOA from BB source tracers)
  - Implement oxidation rates and loss rates (wet/dry deposition) for new organics

Model applications

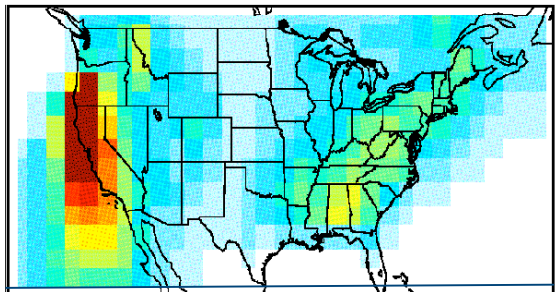
- Investigate OC aerosol loading over North America during wildfire (summer) season and compare to IMPROVE observations over the US (comparison to "standard" simulation) for 2002-2004
- Investigate particularly large events:

MOPITT CO – summer 2004



Alaskan fires of 2004: transported to East Coast and across Atlantic
[Turquety et al., 2007; Lewis et al., 2007]

Surface Simulated OC: Aug 2002



Larges fires in Oregon responsible for haze throughout California, Washington and Oregon in summer 2002 [McMeeking et al., 2006]

Summary

- FLAME data for smoke from a wide variety of U.S-relevant fuels present a unique opportunity for a first look at gas-aerosol partitioning of biomass burning emissions
  - Some of first available real-time data for BB [OC], some speciation (degree of oxygenation), and even molecular markers
    - Lots of filter data to back up newer techniques
  - Results will help guide design of new EPA-supported experiments
- Modules to be developed and tested in GEOS-Chem will provide insights into implications of our findings for regional air quality
  - Heavily-used and validated model for the US: good test bed
  - Coordinate with other EPA STAR studies on SOA formation from BB emissions