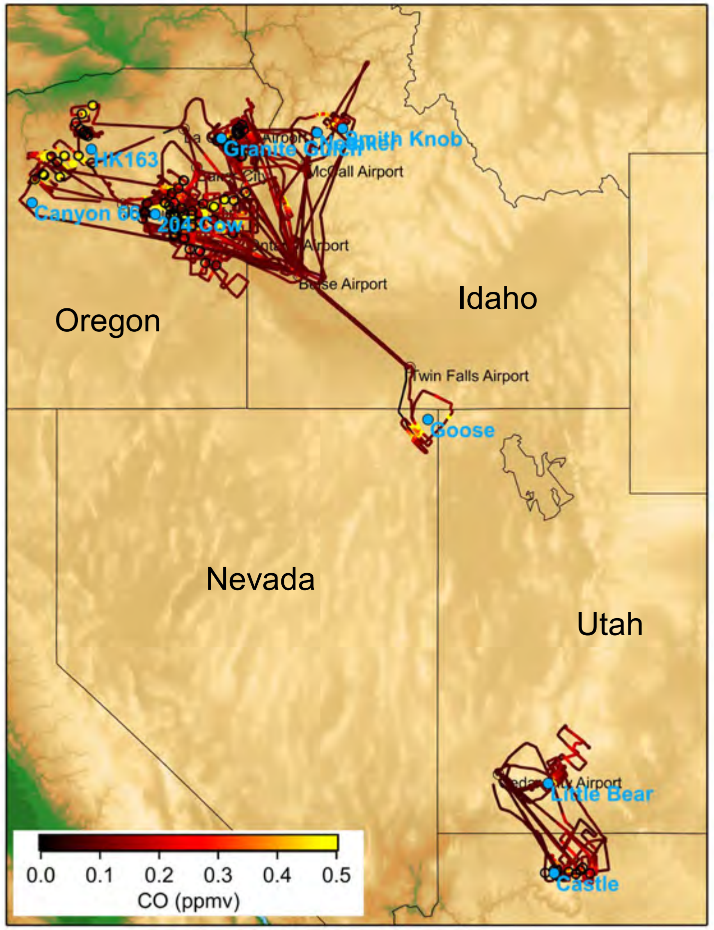
FIREX-AQ Chem Otter Science Meeting

Agenda:
1. Updates and logistics
2. Comparison of three plume age methods
3. Science summaries



13 July 2020

Data and flight information

Flight summary:
https://docs.google.com/spreadsheets/d/1puikxaDmMgNoscQ_-I1vTQKAr1jNtZUYtiOtpqJTifU/edit?usp=sharing

Flight Num	Flight Leg	Start Time (UTC)	Stop Time (UTC)	Duration	Time of Day	Start Airport	Start Code	Stop Airport	Stop Code	Fire Name	Fuel Type	Flight Scientist	Instrument Scientist	Other Details
1	20190729	07/29/2019 17:20:00	07/29/2019 19:25:00	2:05:00	Afternoon	Rocky Mountain Metro	BJC	Rocky Mountain Metr	BJC	Small ag fire	Grass fire	Mike Robinson	Ale Franchin	
2	20190803	08/03/2019 22:08:00	08/04/2019 00:28:00	2:20:00	Afternoon	Boise Airport	BOI	Boise Airport	BOI	None	N/A	Mike Robinson	Rebecca Washenfelde	Calibration of met probes
3	20190805_L1	08/05/2019 21:49:00	08/05/2019 22:33:00	0:44:00	Afternoon	Boise Airport	BOI	Twin Falls Airport	TWF	None (Transit)	N/A	Ale Franchin	Geoff Tyndall	
4	20190805_L2	08/05/2019 23:43:00	08/06/2019 01:44:00	2:01:00	Afternoon	Twin Falls Airport	TWF	Twin Falls Airport	TWF	Goose		Ale Franchin	Geoff Tyndall	
5	20190805_L3	08/06/2019 02:37:00	08/06/2019 03:22:00	0:45:00	Sunset	Twin Falls Airport	TWF	Boise Airport	BOI	None (Transit)	N/A	Ale Franchin	Geoff Tyndall	

R0 data for all instruments:
https://esrl.noaa.gov/csd/groups/csd7/measurements/2019firex-aq/TwinOtter/DataDownload/
Username: firexaq; Password: sm0k3y!

Fire characteristics by Amber Soja and Emily Gargulinski:
https://docs.google.com/spreadsheets/d/1_Jfc3GP9taF8IvP82VHplX1IgsUvvQV7P9CTxrJNRV8/edit#gid=971771572

Spreadsheet for planned manuscripts:
https://docs.google.com/spreadsheets/d/1YpAdNyDaXEe_QeKSimau5-vHDAtV4UVhxLF0TLhYEPQ/edit?usp=sharing

Available soon: Metadata for each plume transect

Metadata is organized as an ICARTT file with one row per plume transect. Available very soon!

Category	**Metadata**
Transect time	Transect_Start_Time, Transect_Stop_Time, Transect_Start_Row, Transect_Stop_Row
Flight information	Transect_Flight_Name, Transect_Flight_Leg, Transect_Plume_Number, Transect_Type
Aircraft location and altitude	Transect_Lat_Midpoint, Transect_Lon_Midpoint, Transect_Alt_Avg, Transect_Alt_Range
Wind speed and direction	Transect_WindSpd_Avg, Transect_WindDir_Avg
Reanalysis plume age	Transect_Reanalysis_Plume_Age, Transect_Reanalysis_Plume_Age_Unc
Average CO and CO_2	Transect_CO_Avg, Transect_CO2_Avg
Fire information	Fire_ID, Fire_Lat, Fire_Lon, Fire_Type
Background times	Background1_Start_Time, Background1_Stop_Time, Background2_Start_Time, Background2_Stop_Time, Background1_Start_Row, Background1_Stop_Row, Background2_Start_Row
Background CO and CO_2	Background1_CO_Avg, Background1_CO2_Avg, Background2_CO_Avg, Background2_CO2_Avg
MCE	MCE_by_Integration, MCE_by_ODR, MCE_by_ODR_r2

Thanks to Chris Holmes for reanalysis plume ages, Zach Decker for checking the transect times, and Kat Ball for making the ICARTT file.

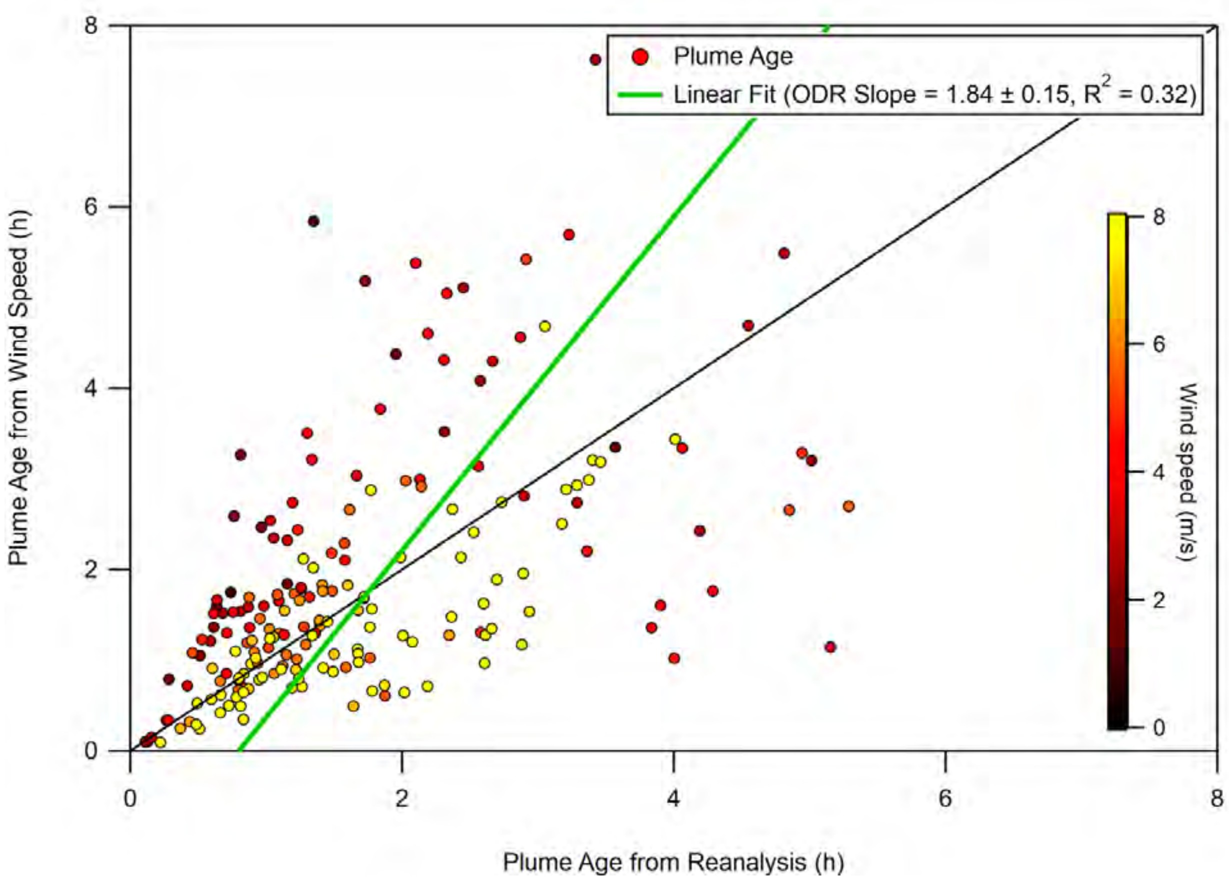
Comparison of Three Methods to Represent Plume Age:

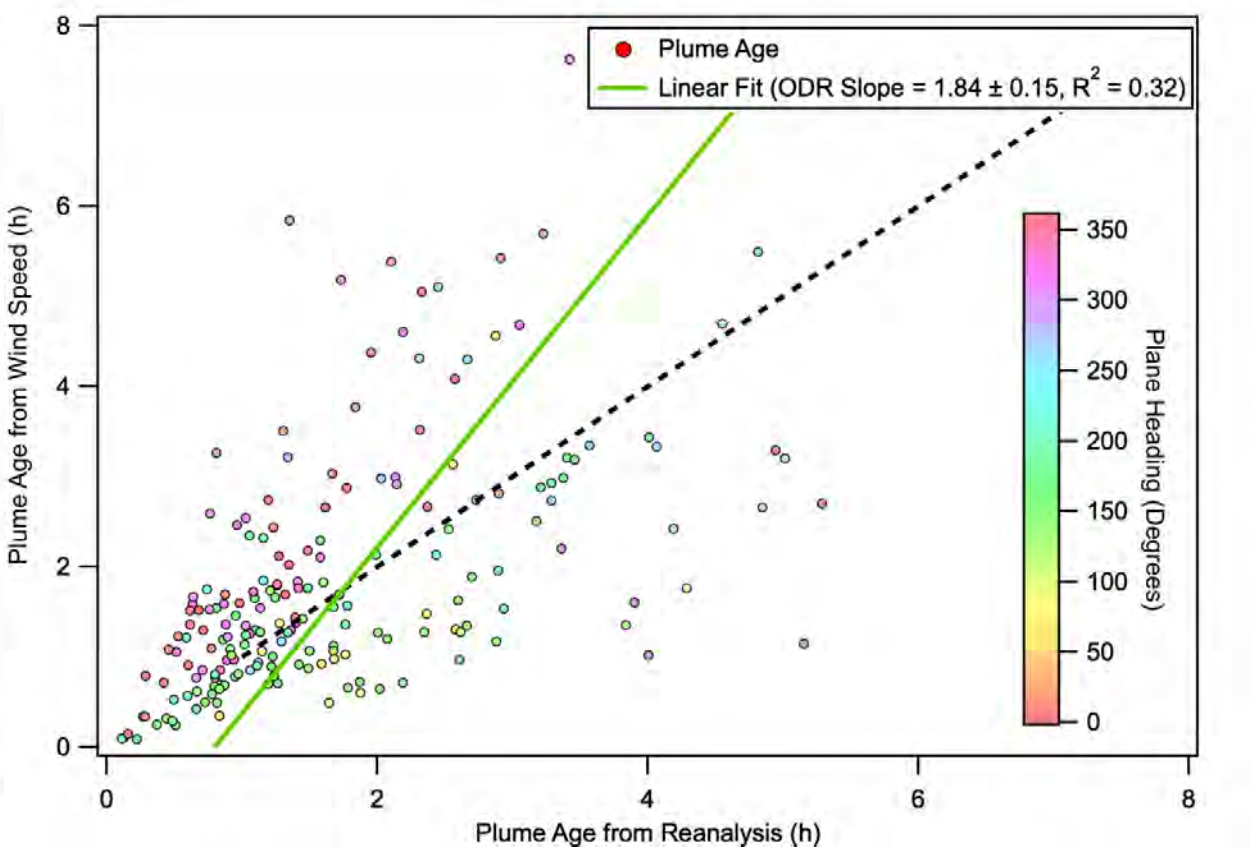
1) Physical age from wind speed and direction
2) Physical age from back trajectory analysis
3) Chemical age from “chemical clocks”

Katherine Ball, Rebecca Washenfelder

July 13, 2020

Physical age of smoke plumes





- ➢ **The measured wind speed varied with aircraft heading during the second half of the field campaign**
- ➢ **Reanalysis plume ages should be used. They have been calculated by Chris Holmes and will be available soon**

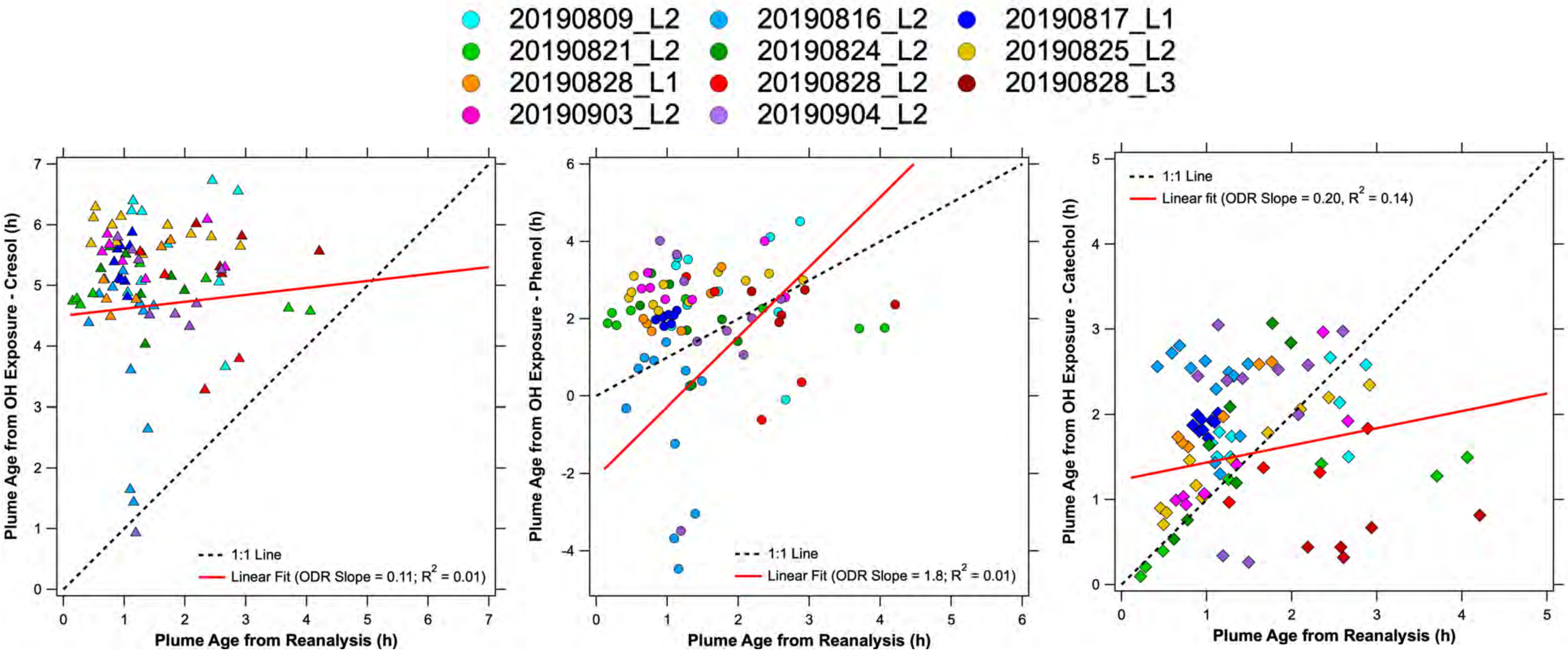
Chemical Age of Smoke Plumes

- Plume ages calculated based on VOC exposure to OH and NO_3 radicals (Eqn. 1 based on Roberts et al., 1984)

$$x\ Exposure = \frac{\ln(VOC/CO)_{aged} - \ln(VOC/CO)_{fresh}}{k_{x-CO} - k_{x-VOC}} \qquad \text{(Eq. 1)}$$

- x denotes oxidant species (either OH or NO_3)
- $\ln(VOC/CO)_{fresh}$ values from Koss et al., 2018 ACP
    - Ratios determined from FireLab experiments
    - Multiplied by 1.5 to account to difference in burning efficiency between field measurements and laboratory experiments
- k_{x-VOC} values from Coggon et al., 2018 ACP supplemental material
- k_{OH-CO} estimated as 1.5e-13 $cm^3\ molec^{-1}\ s^{-1}$ from NIST chemical reference
- k_{NO_3-CO} estimated as 4E-19 $cm^3\ molec^{-1}\ s^{-1}$ from NASA JPL Publication No. 15-10
- Calculations performed on flights with A or A/B rating

Three daytime chemical clocks from I^- CIMS

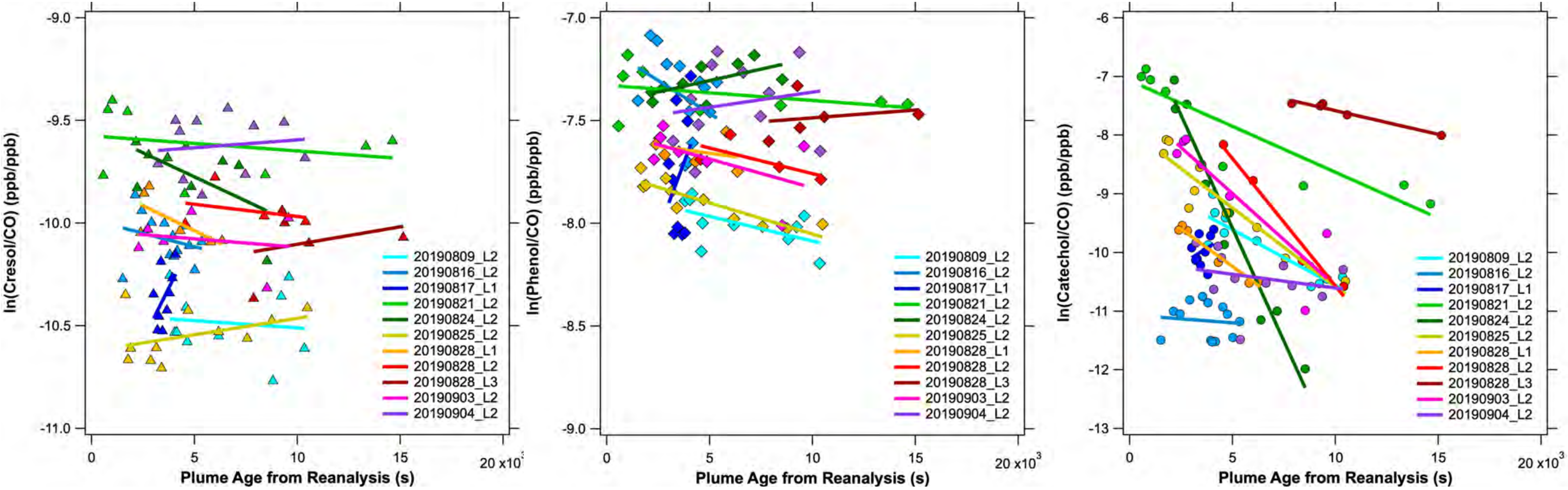


- ➢ Catechol OH exposure clock has best correlation with reanalysis plume age out of three VOC species
  - ➢ Unexpected due to significance of secondary formation of catechol from OH + phenol

Cresol, Phenol, and Catechol Clocks (OH Exposure)

Linearized form of Eqn. 1:

$$\ln\left(\frac{VOC}{CO}\right)_{aged} = [OH](k_{OH-CO} - k_{OH-VOC})\times PlumeAge + \ln\left(\frac{VOC}{CO}\right)_{fresh}$$



➢ **Avg [OH] derived from Catechol OH exposure clock has best correlation with literature [OH] concentration Ratio (Avg/Lit $R^2 > 0.2$) = 0.78**

Future Work

- Calculate chemical clocks from VOC cartridge measurements
  - Furan, 2-methylfuran, 2,5-dimethylfuran, and furfural
- Investigate correlation of various plume ages with BrC lifetime
- Adjust reanalysis plume ages with updates from Chris Holmes
  - Fire location origin based on fire start-point and will be updated with satellite observations
  - On days where individual fire was ambiguous, back trajectory will be performed on both possible fires
- Compare observations with modeled predictions

Short summary presentations with analysis and plans

- **Topic of your analysis and likely coauthors**
- **Key results**
- **Remaining work**
- **Any missing information that you need from others?**

Gas	Mike Robinson	Rapidly changing ozone-NO_x-VOC chemistry in western wildfire plumes: A comparison of afternoon and evening photochemistry
Gas	Zach Decker	BBVOC profile and evolution as seen on the Chem-Twin Otter and DC-8 by Positive Matrix Factorization analysis
Gas	Carley Frederickson	HONO enhancement ratios in daytime and nighttime wildfire plumes and their evolution in time
Gas	Zach Decker	Observations and box modeling of "nighttime" smoke as seen on the Chemistry Twin Otter and DC-8

Aerosol	Rebecca Washenfelder	Brown carbon lifetimes in wildfire plumes
Aerosol	Felipe Rivera-Adorno	Analysis of impaction samples
Aerosol	Lisa Azzarello	Characterization of smoke aerosols sampled in western USA using ion chromatography and size exclusion chromatography with ultraviolet–visible detection

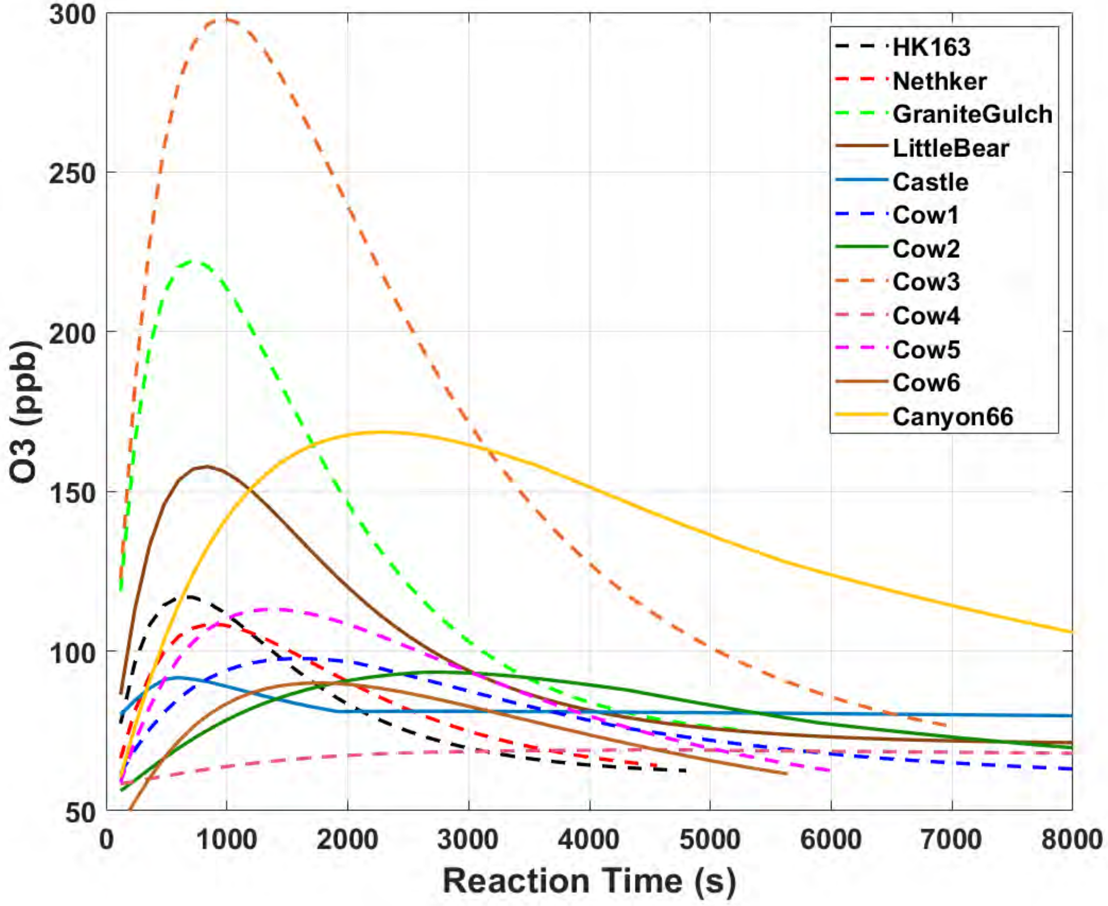
Mod	Megan Bela	Emissions effects on prediction of air quality impacts from fires

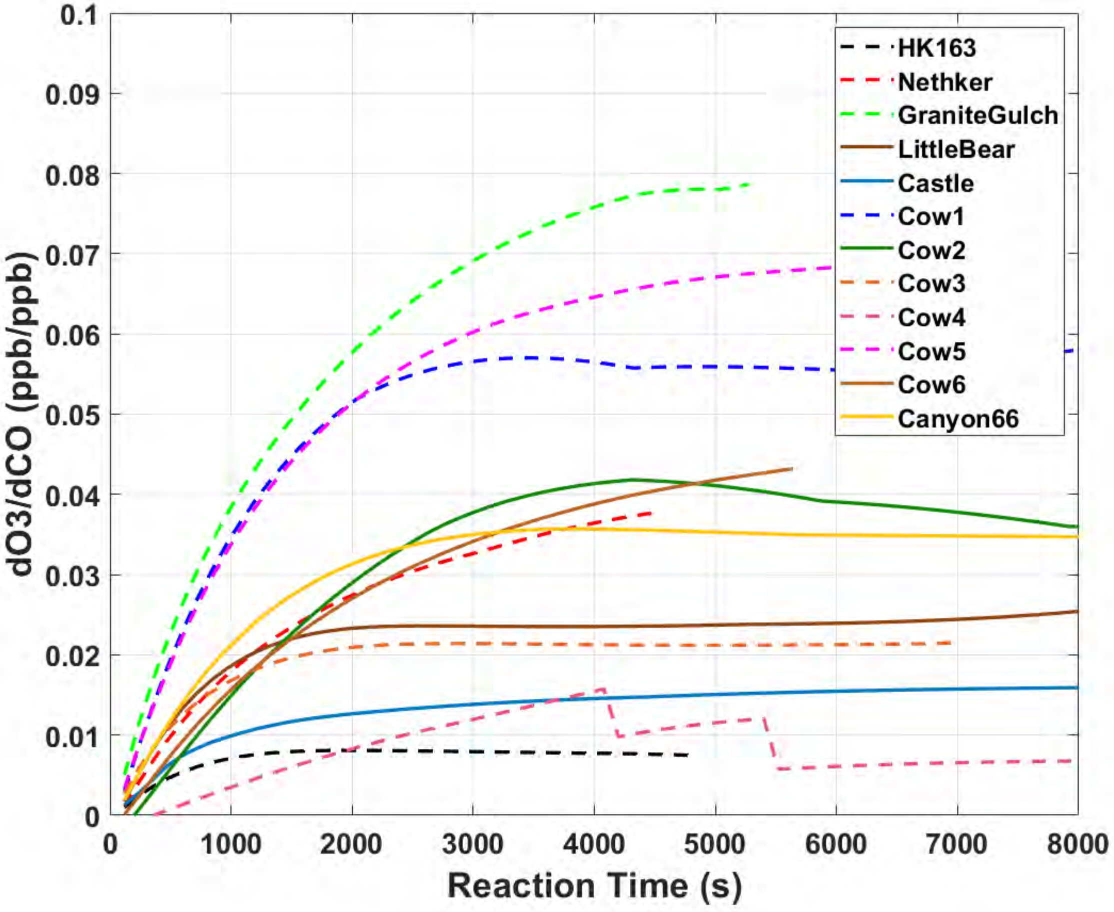
Paul Van Rooy and Ale Franchin are also working on analyses.

Rapidly changing Ozone-NOx-VOC chemistry in western wildfire plumes: A comparison of afternoon and evening photochemistry

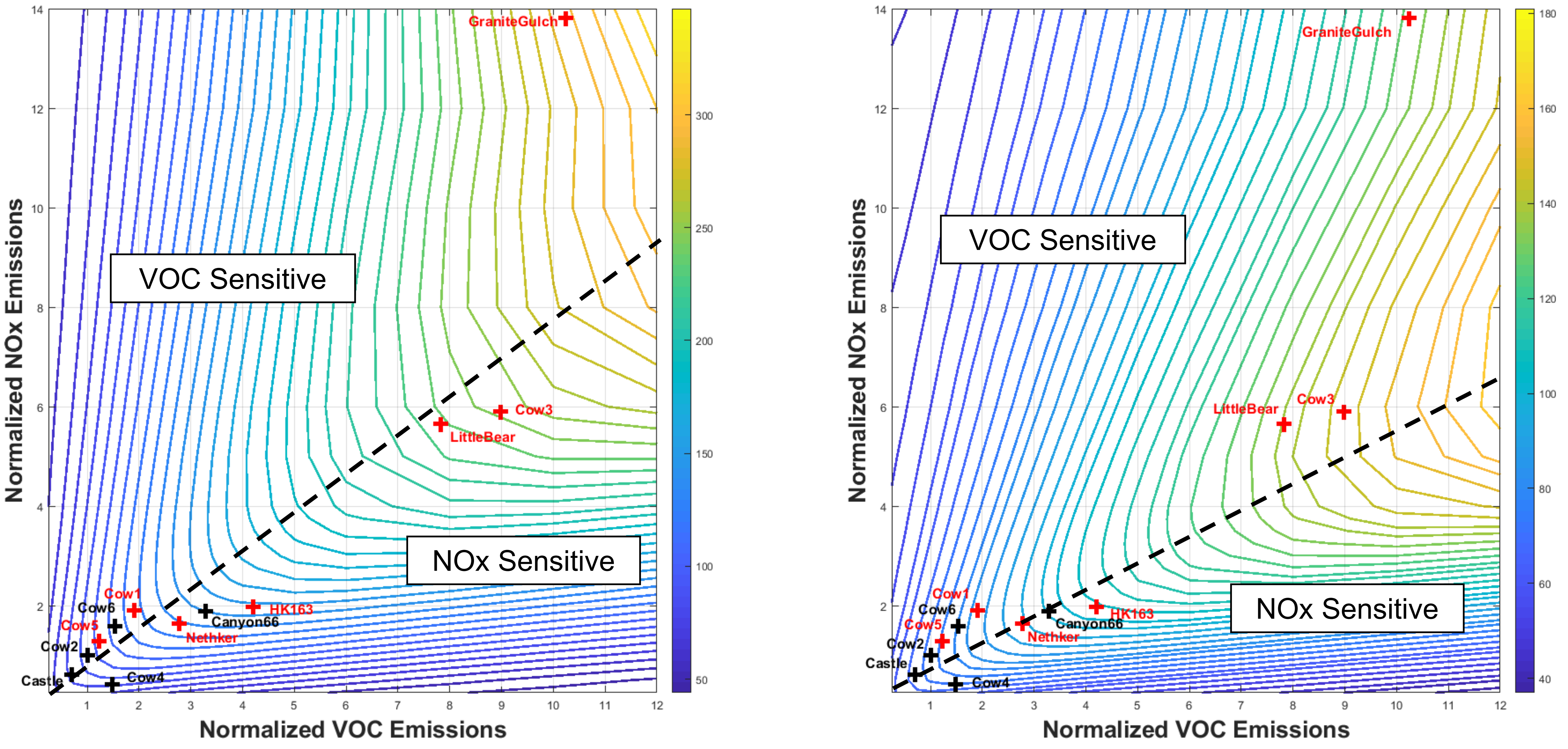
Michael A. Robinson, Zachary Decker, Kelley C. Barsanti,
Matthew M. Coggon, Frank Flocke, Carly Fredrickson, Avi Lavi,
Denise Monksta, Brett B. Palm, Joel A. Thornton, Geoff Tyndall,
Paul Van Rooy, Rebecca H. Schwantes, Andrew Wenhiemer, and
Steven S. Brown

Ozone photochemistry is fast.

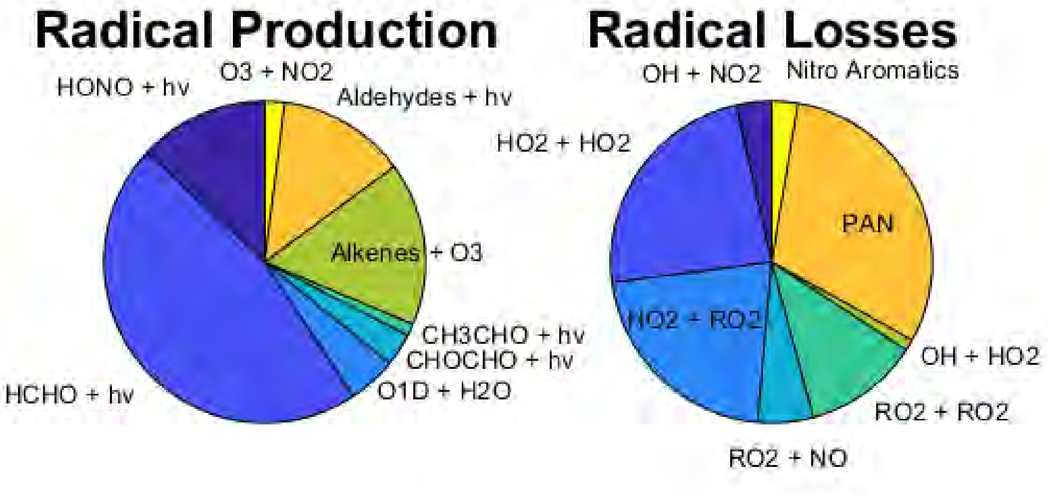


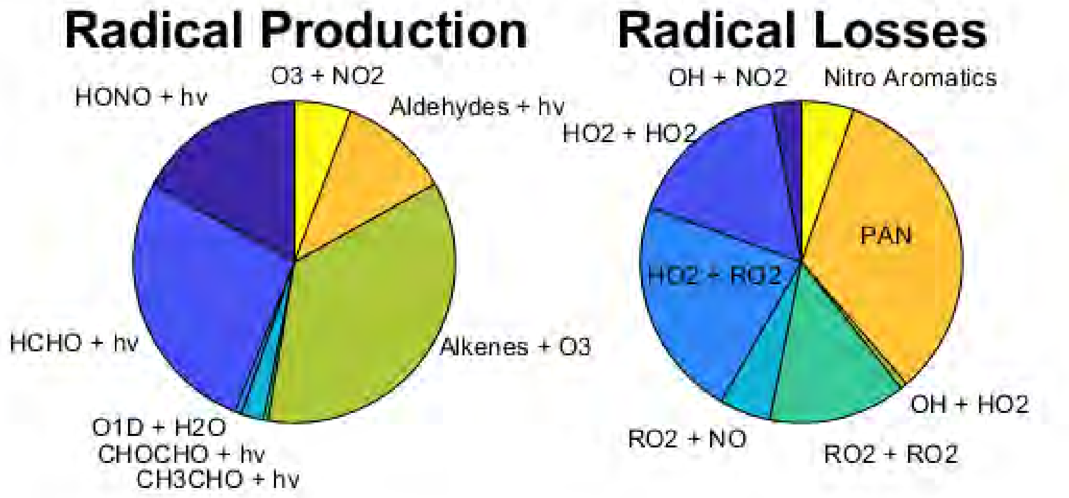


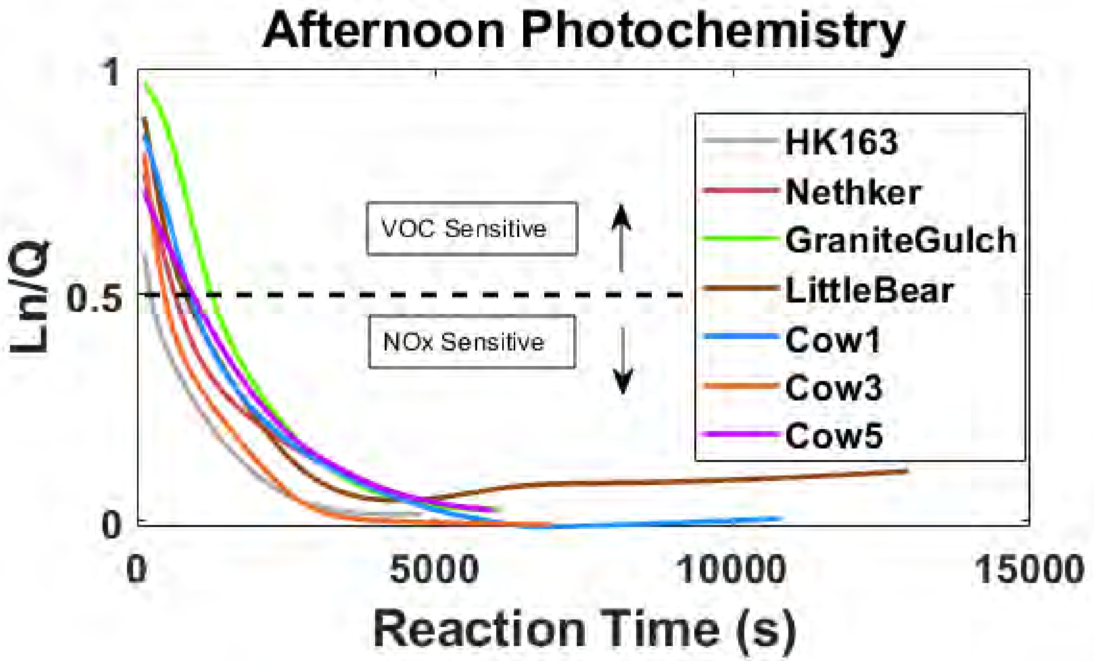
Ozone isopleth doesn’t tell the full story

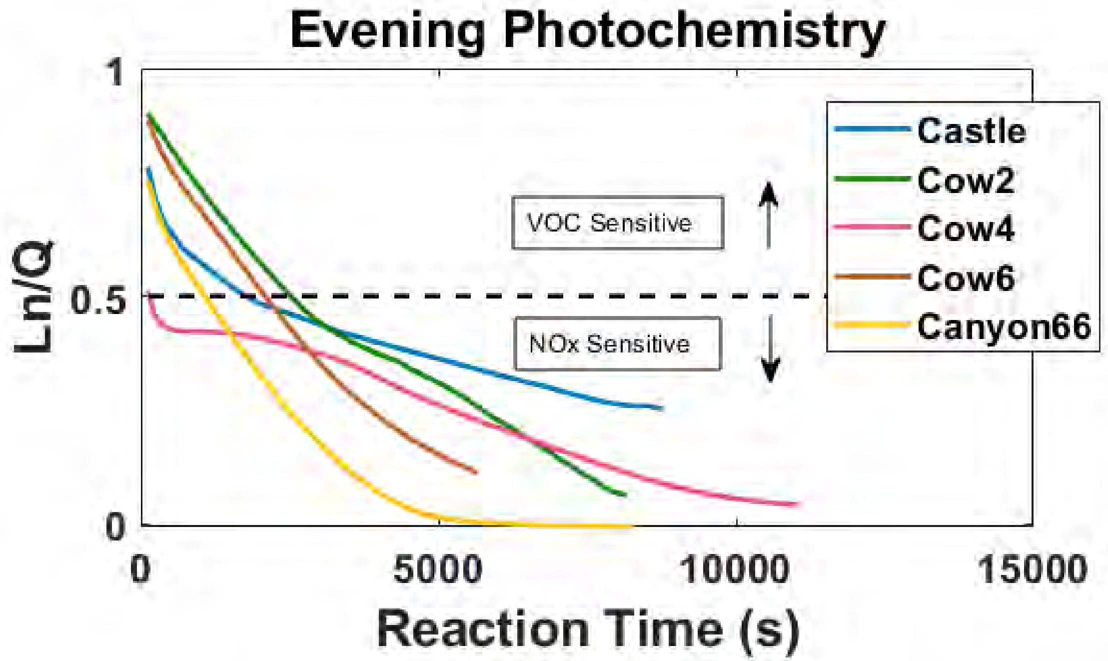


Fast transition to NOx sensitive chemistry



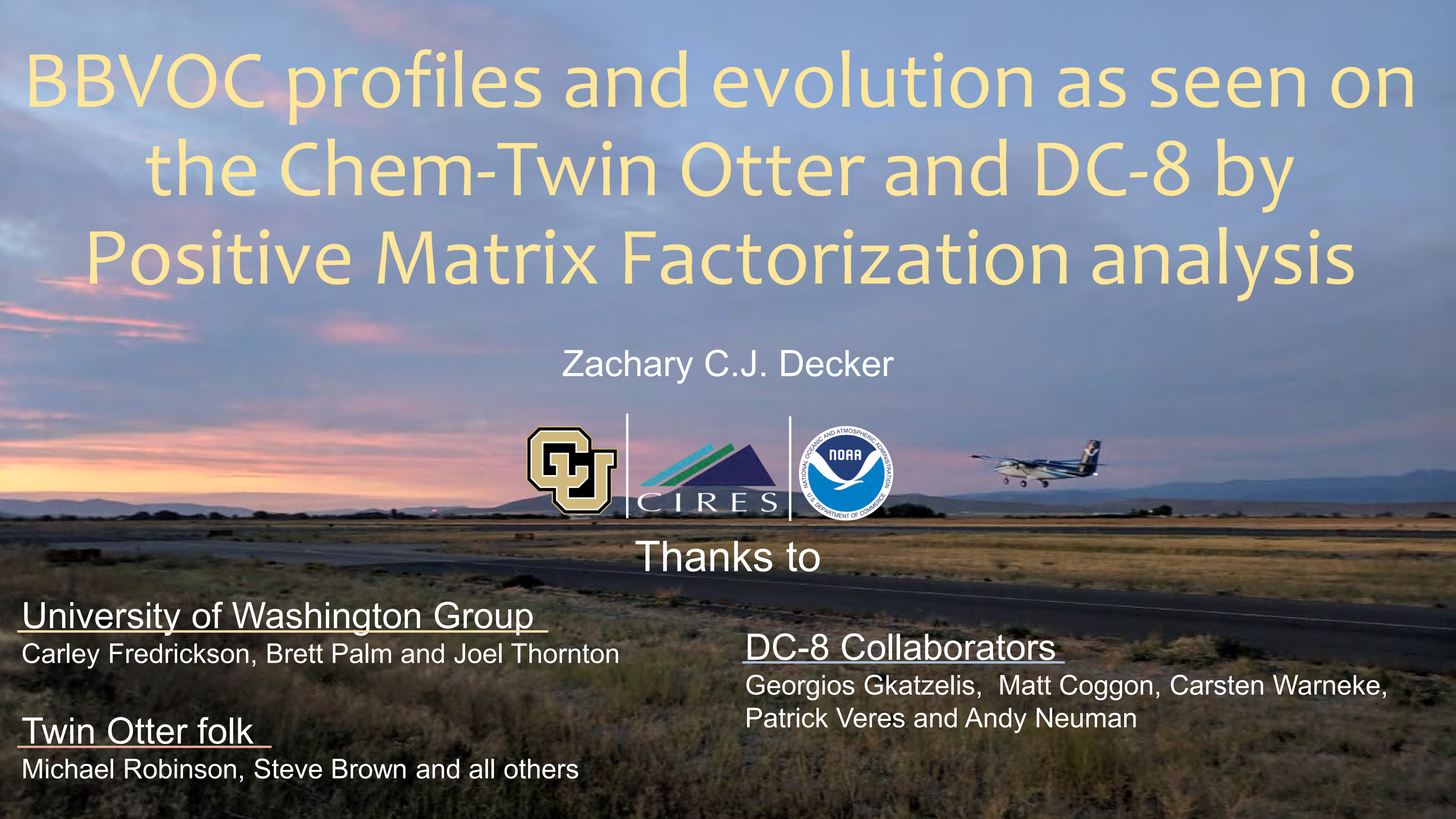






What's left to do?

- HONO sensitivity test
- Finish remaining model runs
  - Updated transport times
  - isopleth



BBVOC profiles and evolution as seen on the Chem-Twin Otter and DC-8 by Positive Matrix Factorization analysis

Zachary C.J. Decker

Thanks to

University of Washington Group
Carley Fredrickson, Brett Palm and Joel Thornton

Twin Otter folk
Michael Robinson, Steve Brown and all others

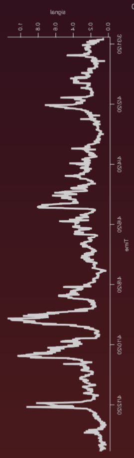
DC-8 Collaborators
Georgios Gkatzelis, Matt Coggon, Carsten Warneke, Patrick Veres and Andy Neuman

Can we find trends in BBVOC evolution from dark plumes?

Positive Matrix Factorization (PMF) is the right tool

- We correlate the time series of ~1500 I^- CIMS masses with CO
- Select the top correlations ($r^2 > 0.2$) → ~150 masses (excluding reagent ions)
- Provide our PMF program with campaign wide time series for all 150 masses

Masses



Time series

PMF calculates W and H

V (n x m) = W (n x p) • H (p x m)

An input matrix

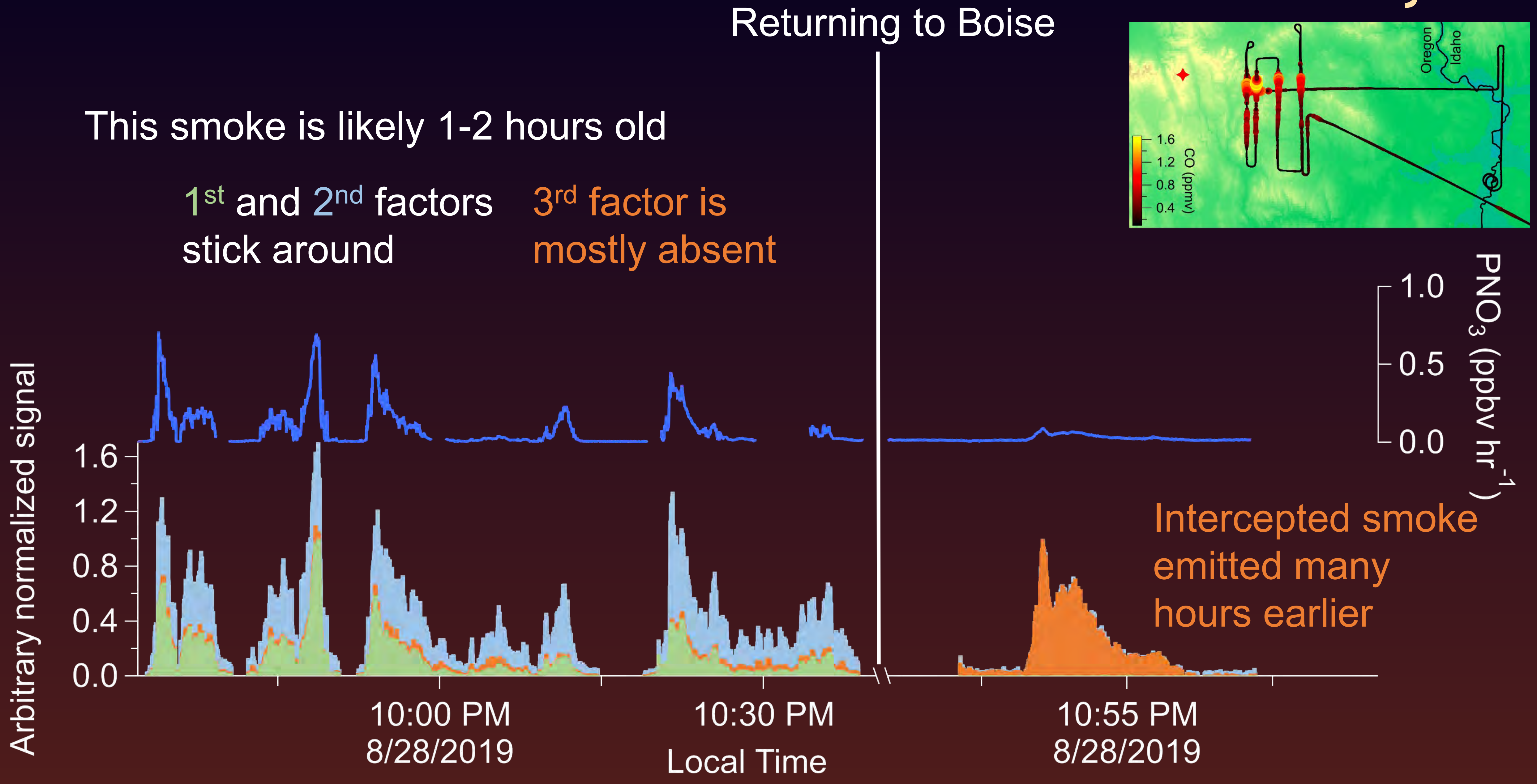
A matrix of trends

A matrix of weights for each trend

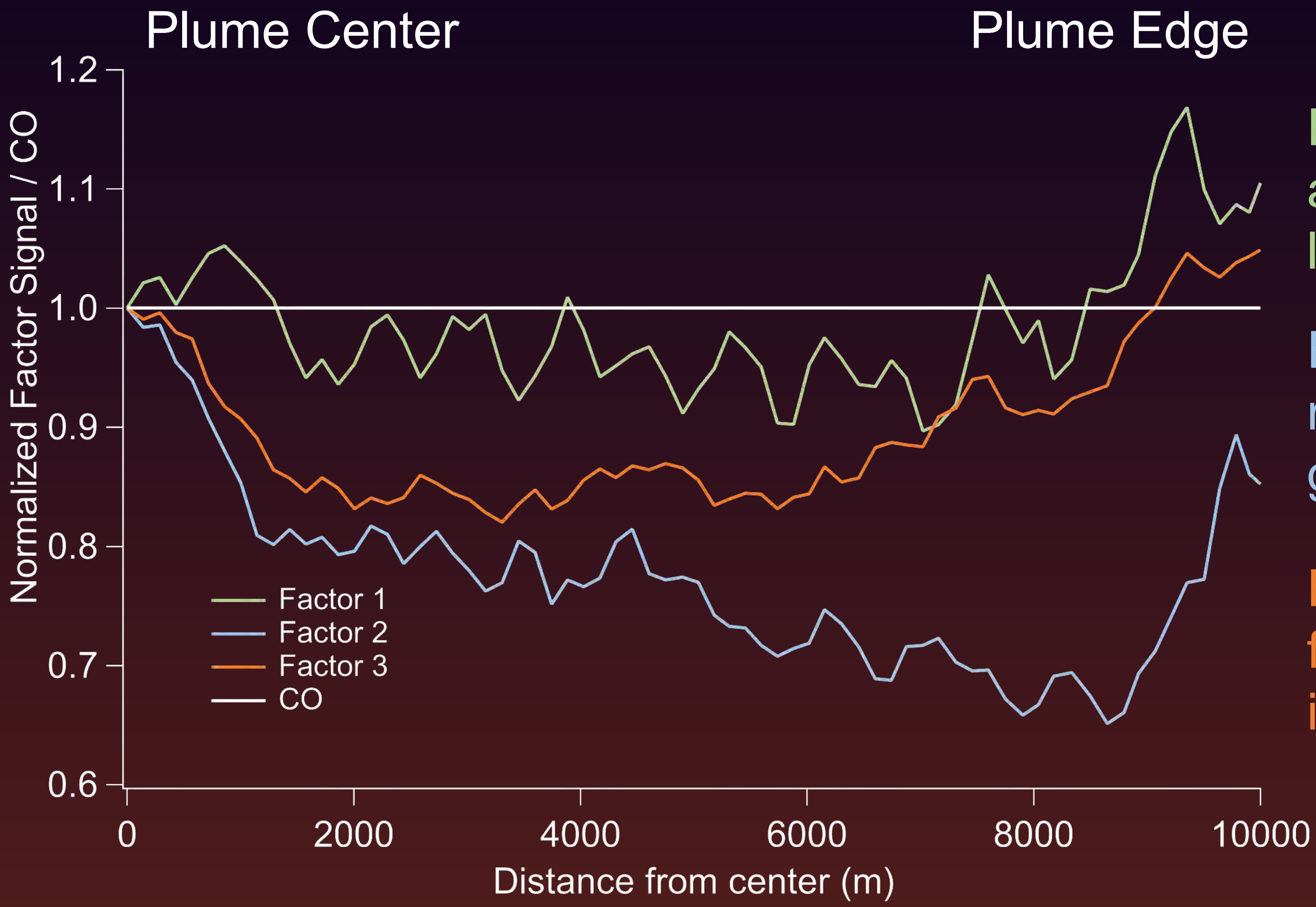
PMF outputs “Factors” or groups of compounds that correlate in time

1. Ranked “tracers” for each factor
2. Factor time series

Smoke Emitted After Sunset Reacts Slowly



What can we Learn About Center and Edge Effects?



Factor 1 reactivity doesn't appear to depend on location.*

Factor 2 is skinny. The reactivity of these tracers is greatest near the edge.

Factor 3 is wide. The formation of these tracers is greatest at the edge.

*May be due to a lack of Factor 1 signal in most plumes.

To Do

- Understand how these factors relate to jNO_2
- BrC correlation to these factors? – Coming soon.

- The DC-8 has lots of data too
  - PMF has been run with the NOAA CIMS dataset. Analysis is underway.

Planned Analysis: HONO enhancement ratios in daytime and nighttime wildfire plumes and their evolution in time

Carley Fredrickson
Advisor: Joel Thornton
University of Washington

Research Questions:

1. How much reactive nitrogen is emitted in wildfire plumes and what is the speciation of that emitted reactive nitrogen (HONO vs. NOx)?
2. How do the emissions and speciation of reactive nitrogen vary with fire characteristics?
3. What is the lifetime of HONO and NOx in wildfire plumes and what controls that lifetime?

Observations and box modeling of "nighttime" smoke as seen on the Chemistry Twin Otter and DC-8

Zachary C.J. Decker

Thanks to

University of Washington Group

Carley Fredrickson, Brett Palm and Joel Thornton

Twin Otter folk

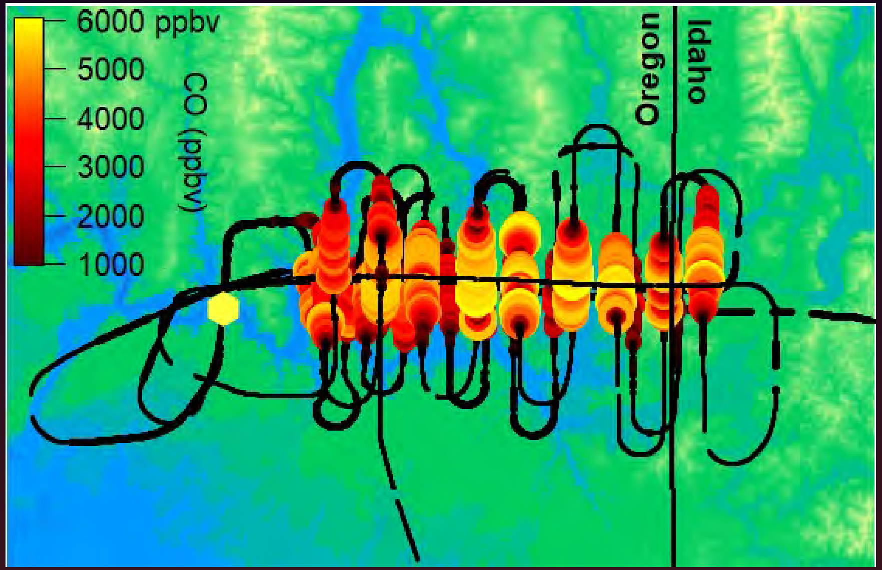
Michael Robinson, Steve Brown, Paul Vanrooy, Kelley Barsanti…and all others

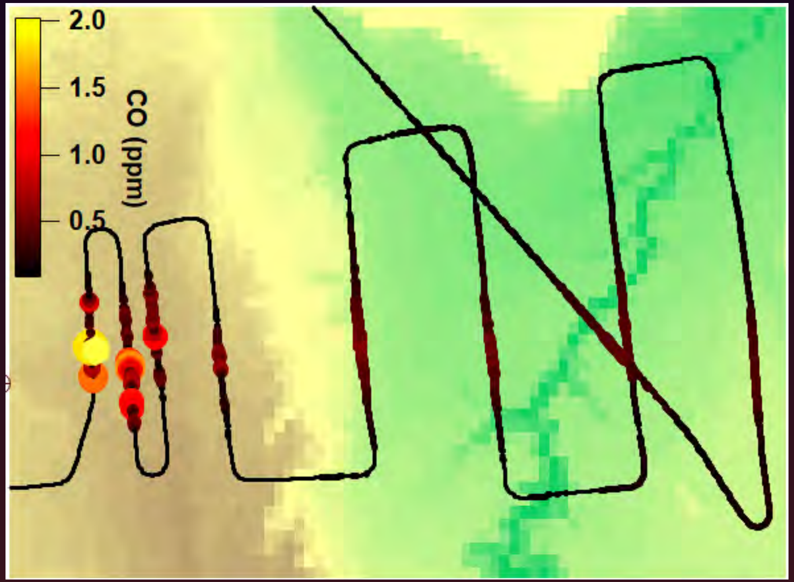
DC-8 Collaborators

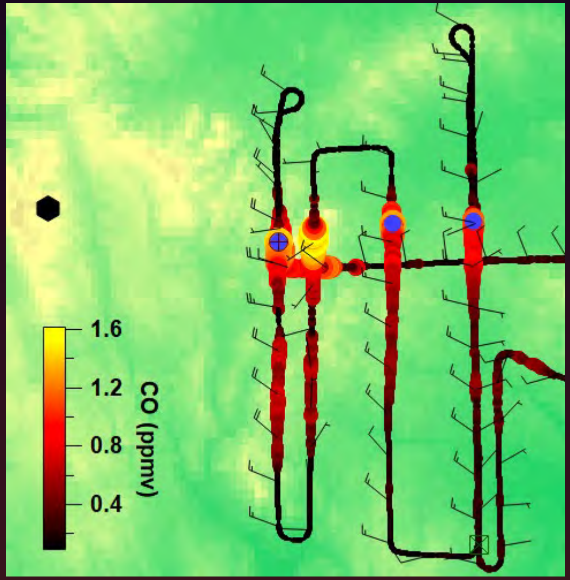
Patrick Veres, Andy Neuman, Aaron lamplugh, Jessica Gilman

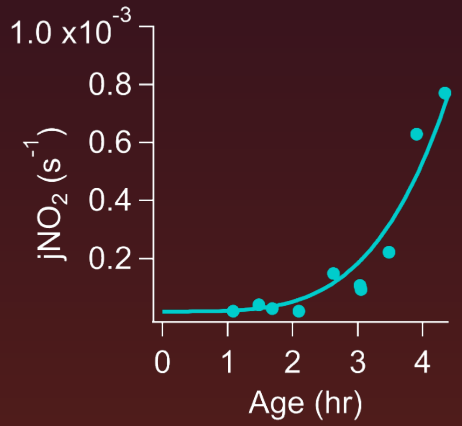
Focusing on four “dark” plumes

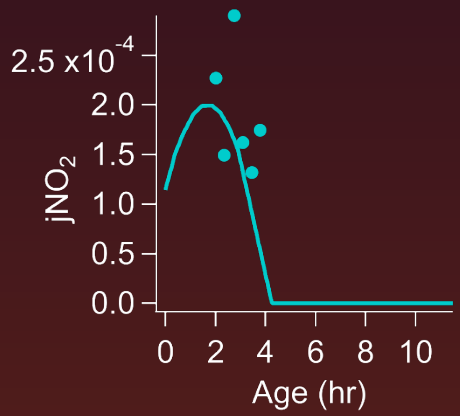
William Flats (DC-8) Aug 07 (mid-day and sunset)	Castle (C-TO) Aug 21 (Sunset)	Cow (C-TO) Aug 28 (Night)

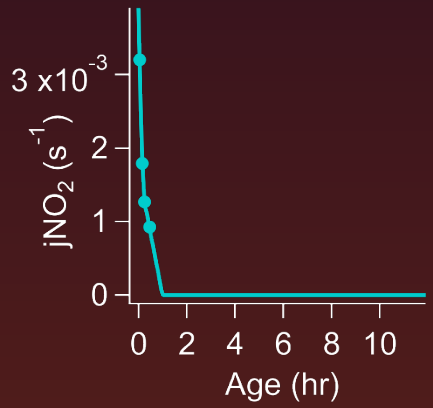


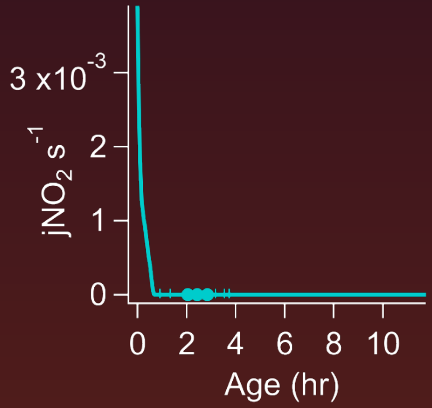




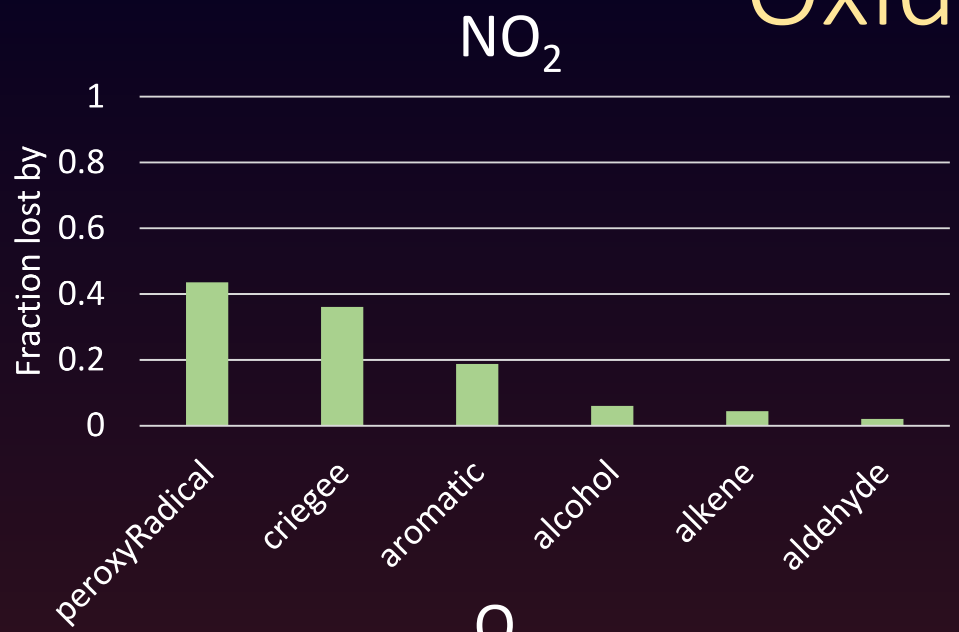


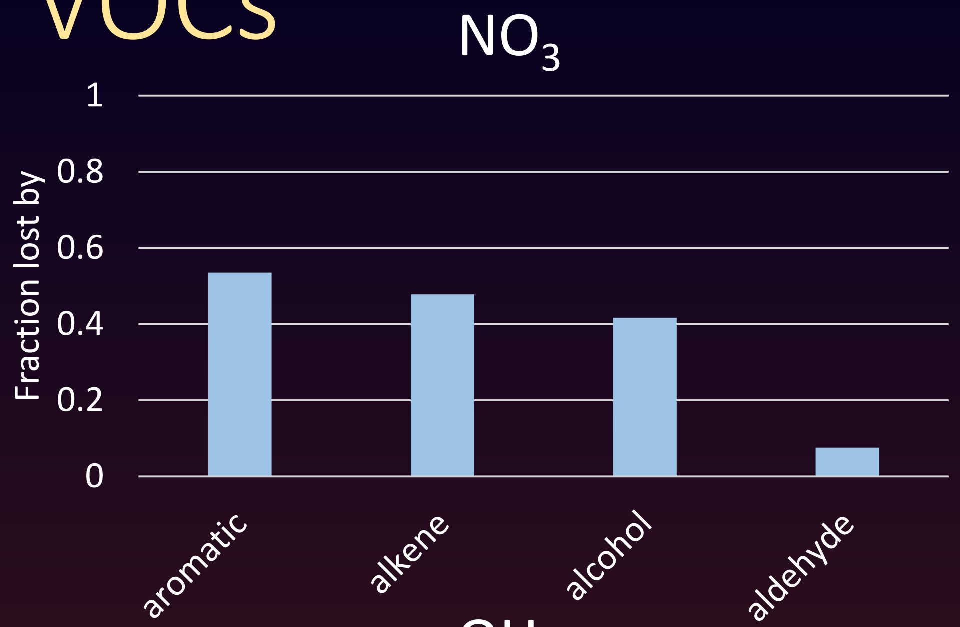


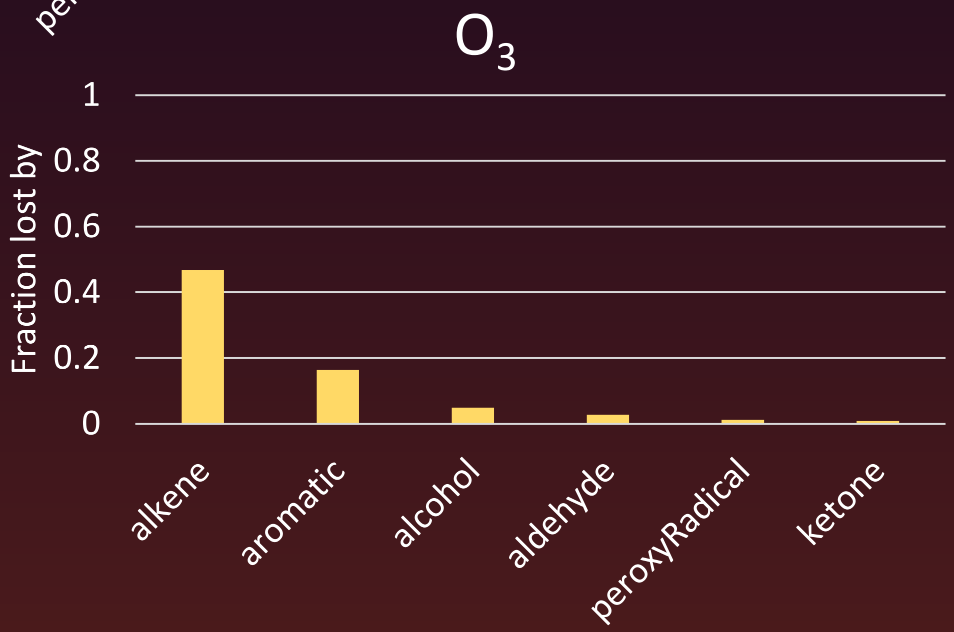


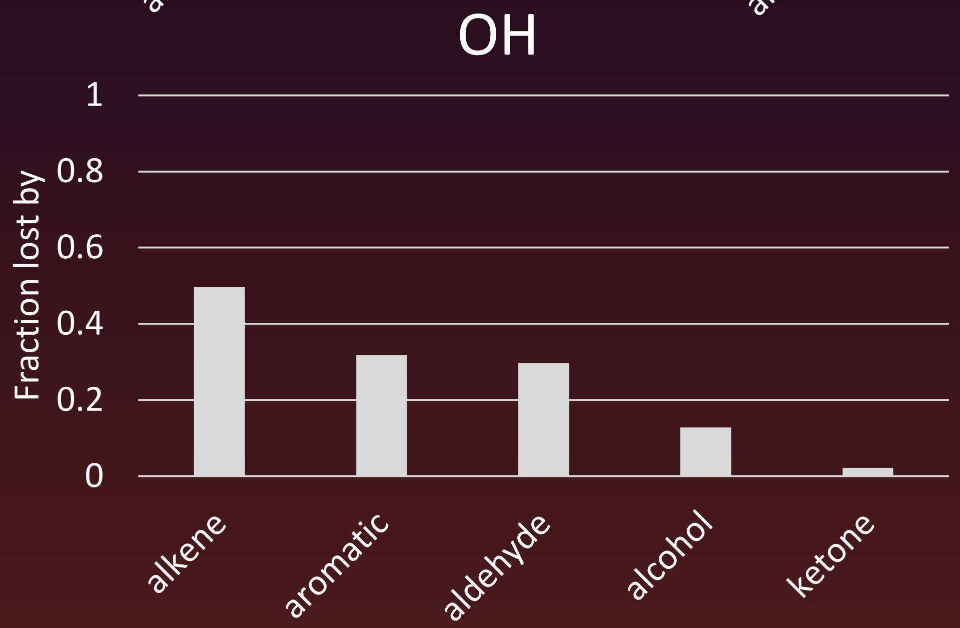


Oxidant loss to VOCs







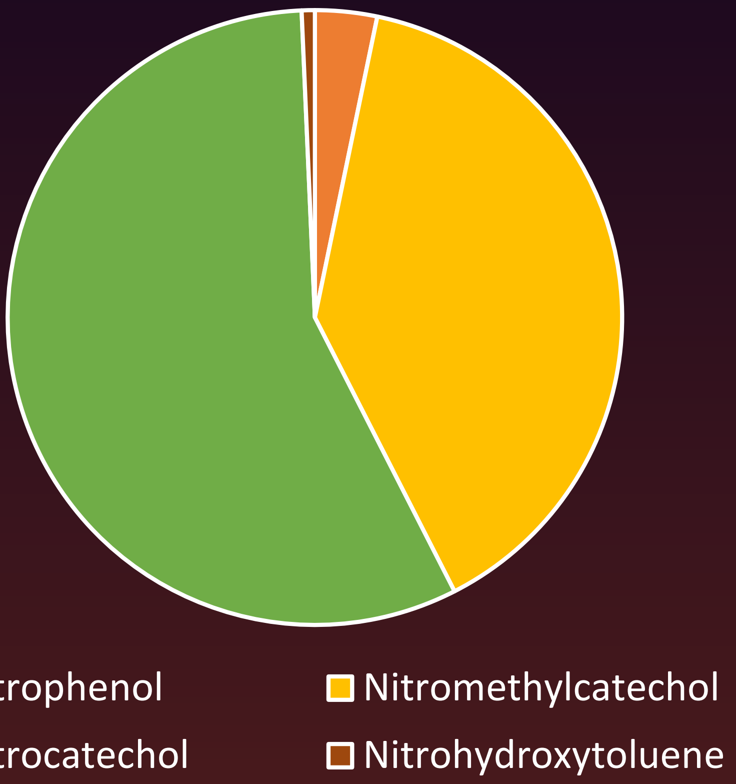


Castle fire 08/21 L2

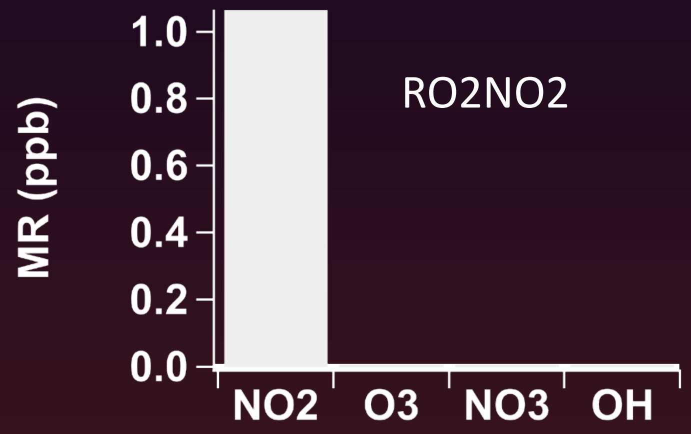
* Groups sum to greater than one because functionalities overlap (Catechol = alcohol + aromatic)

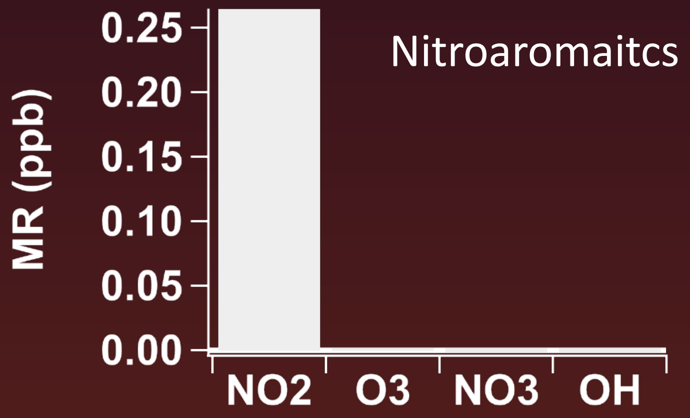
Functional group production

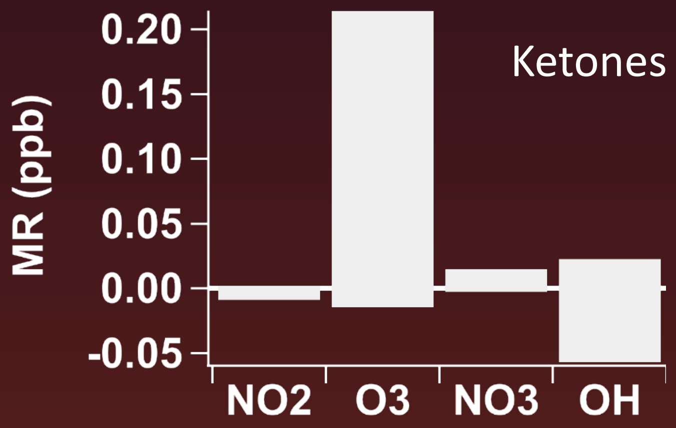
Fraction of produced nitroaromatic



Formation of functional group







Castle fire 08/21 L2

To Do

- Still some more data to pull from the model
- Working on reducing the model outputs into more concise “big picture” figures.
- Any input or ideas are welcome!!!

- Waiting on plume ages from Chris Holmes for the DC-8 models.

Title: “Brown carbon lifetimes in wildfire plumes"

Possible Authors:

BrC-PILS: Rebecca Washenfelder, Lisa Azzarello

CO: Mike Robinson

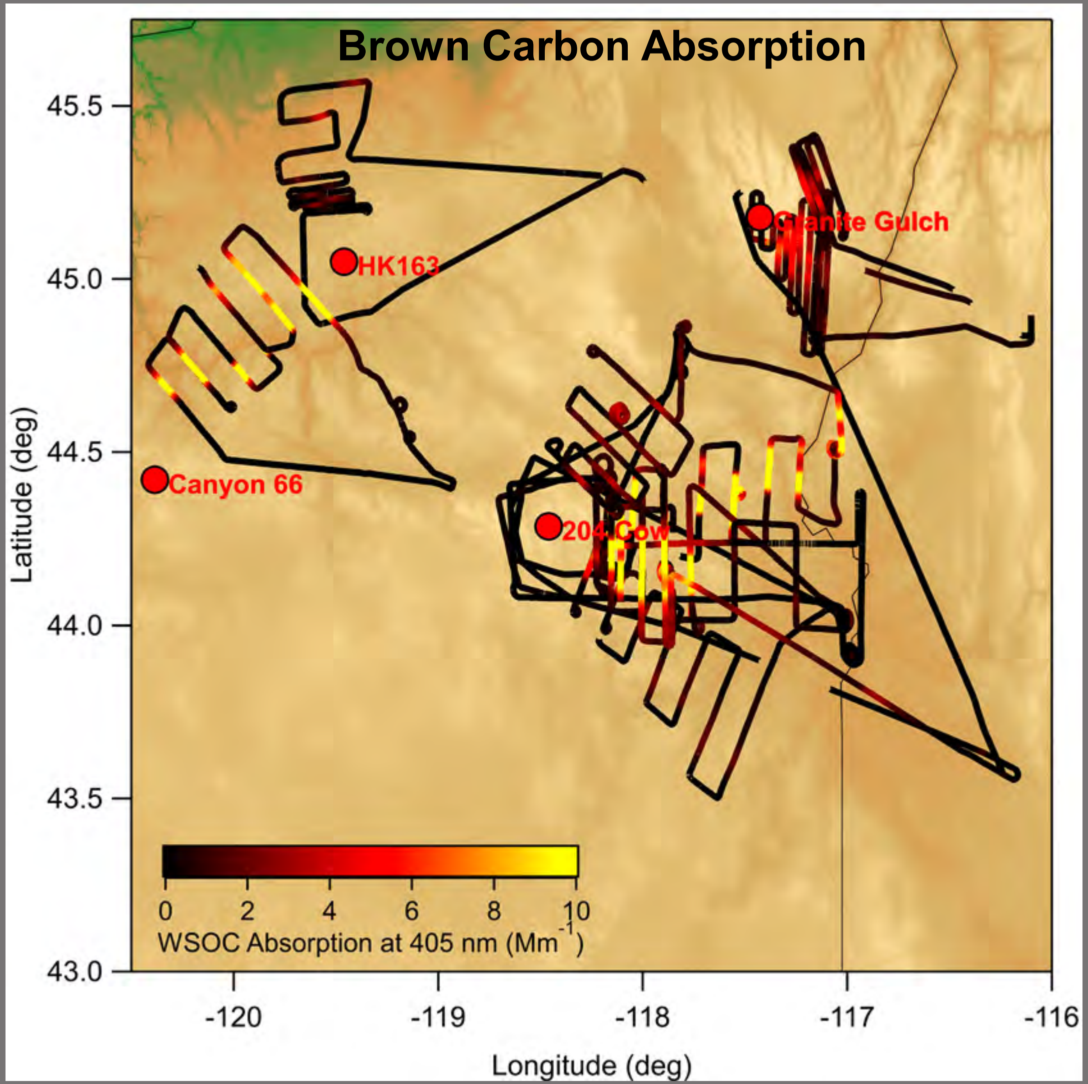
I^- CIMS: Carley Frederickson, Zach Decker, Brett Palm

GCxGC TOF-MS: Paul Van Rooy, …

AMS: Ann Middlebrook and Ale Franchin (AMS)

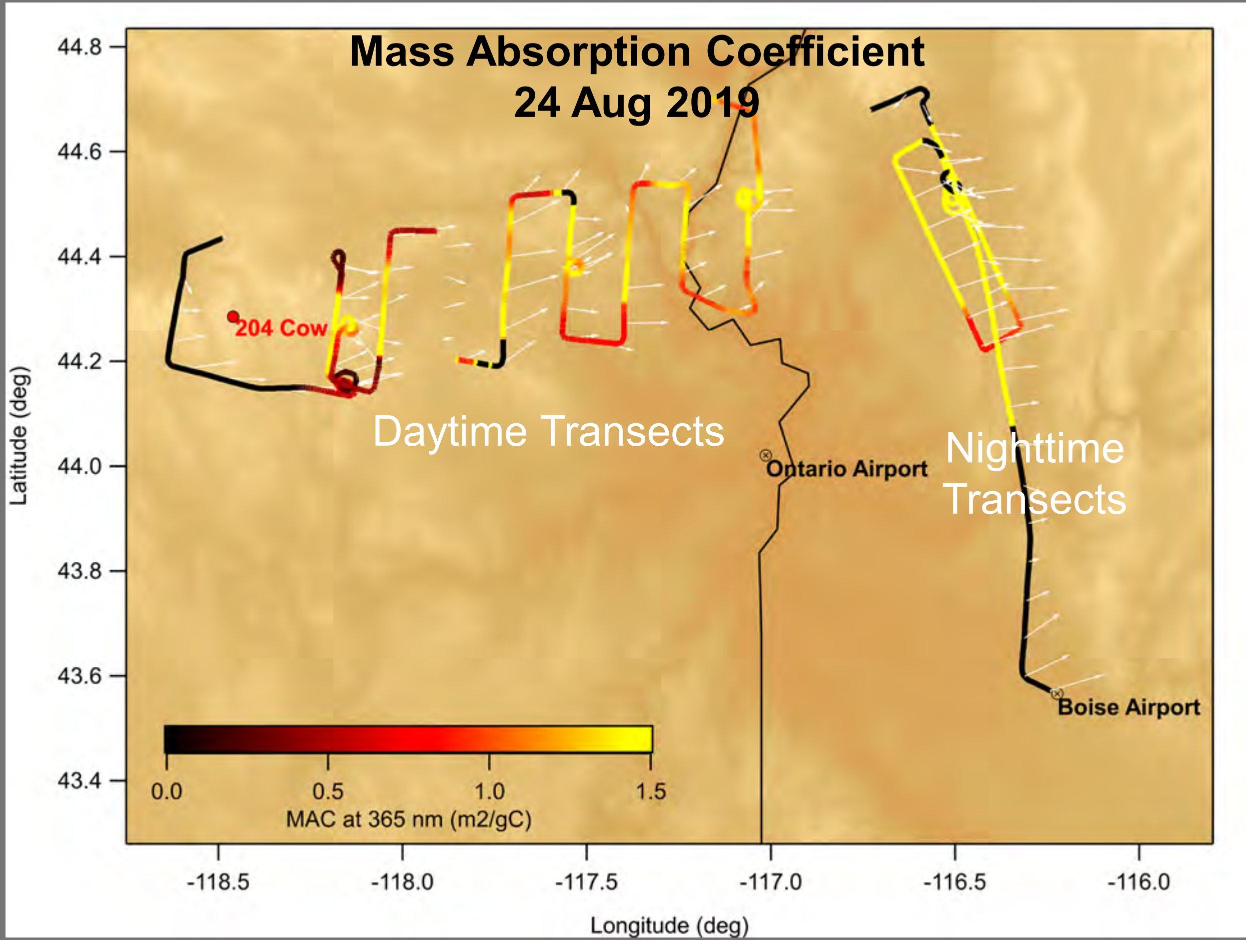
Measured Brown Carbon and MAC

A subset of the 40 flights had consistent wind directions and well-organized plumes. Selected flights are shown here:

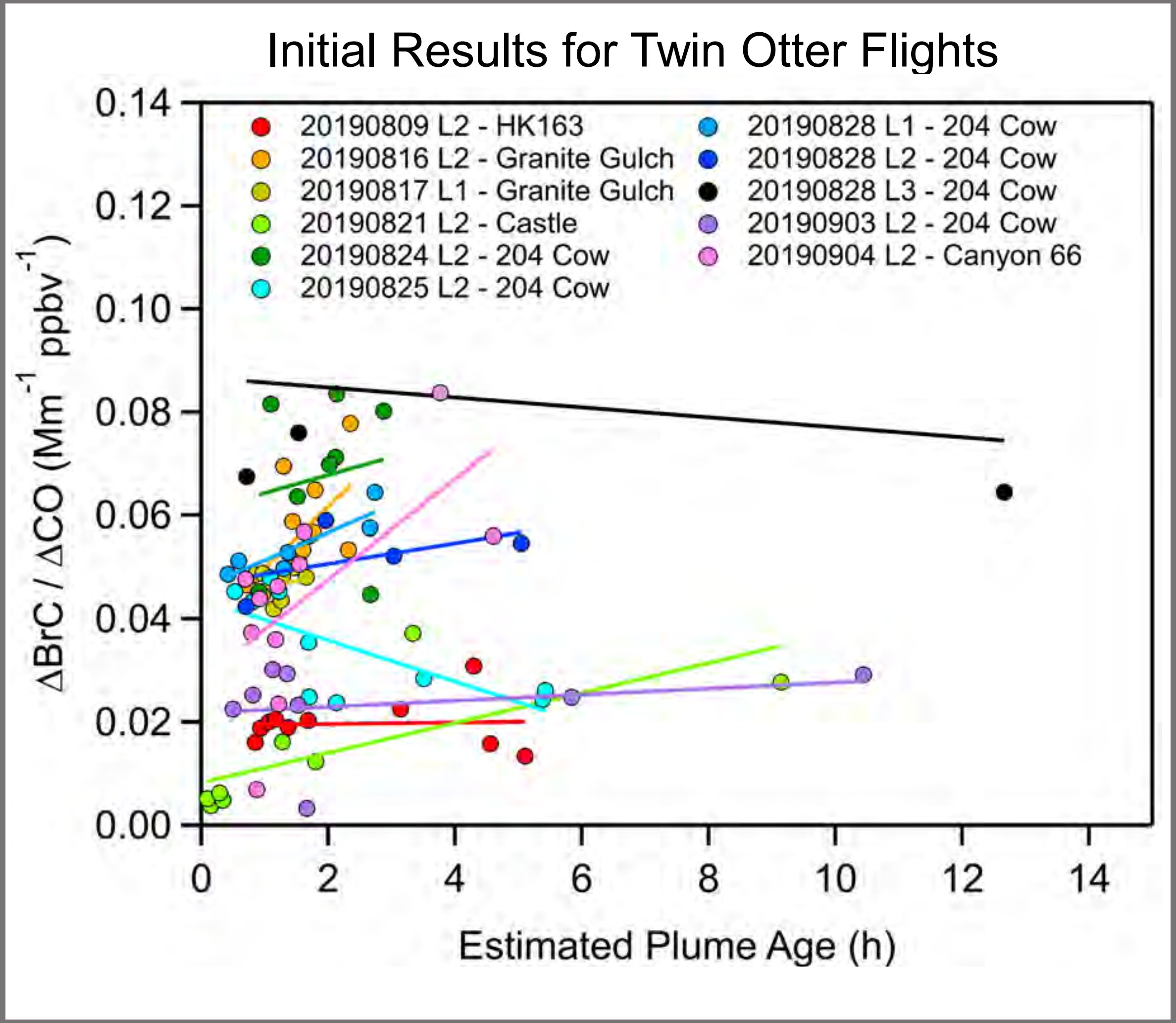


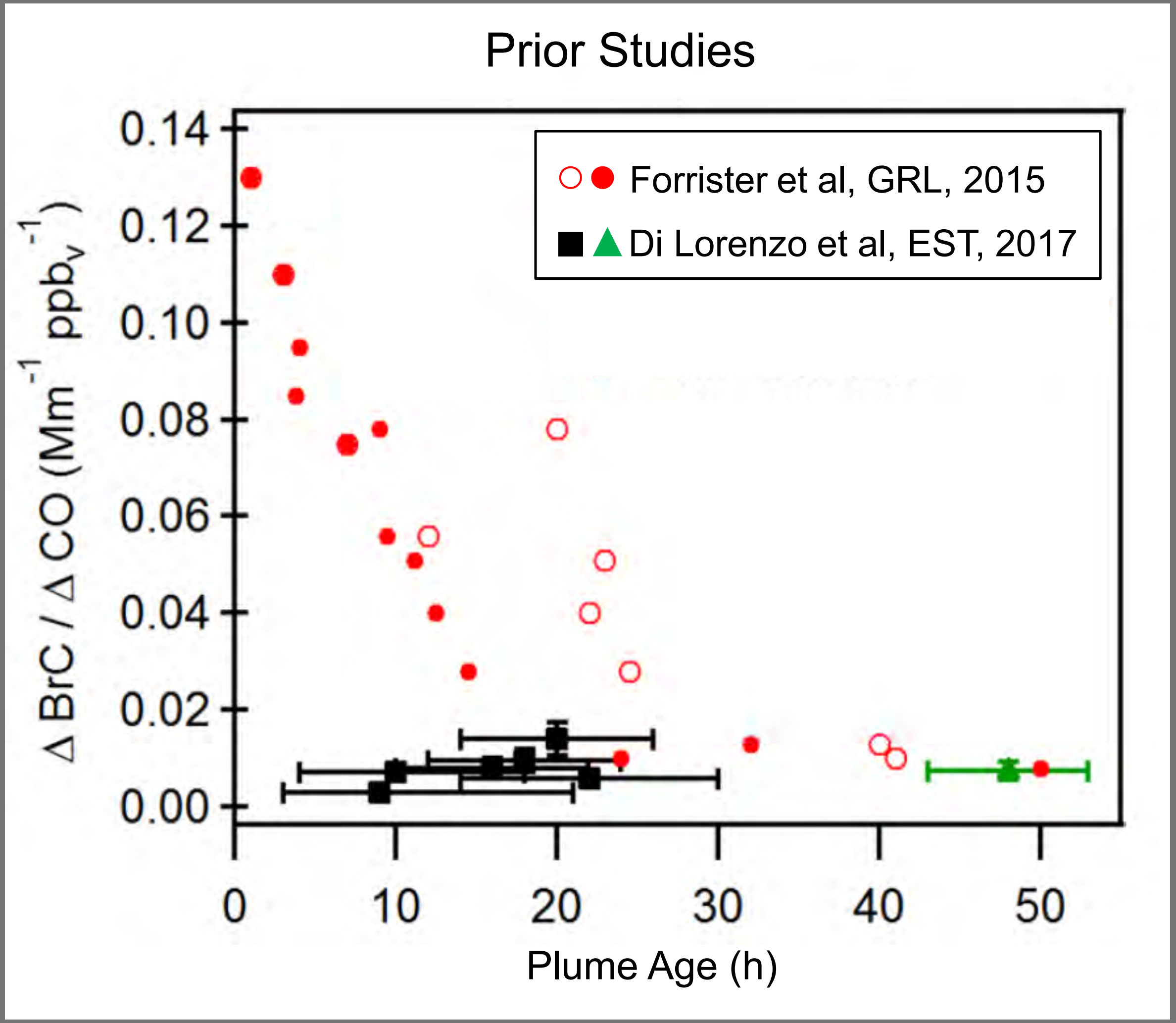
Downwind changes in brown carbon absorption can be determined by normalizing to aerosol WSOC or CO to account for dilution.

BrC absorption / WSOC concentration = Mass Absorption Coefficient



ΔBrC / ΔCO often increased with plume age

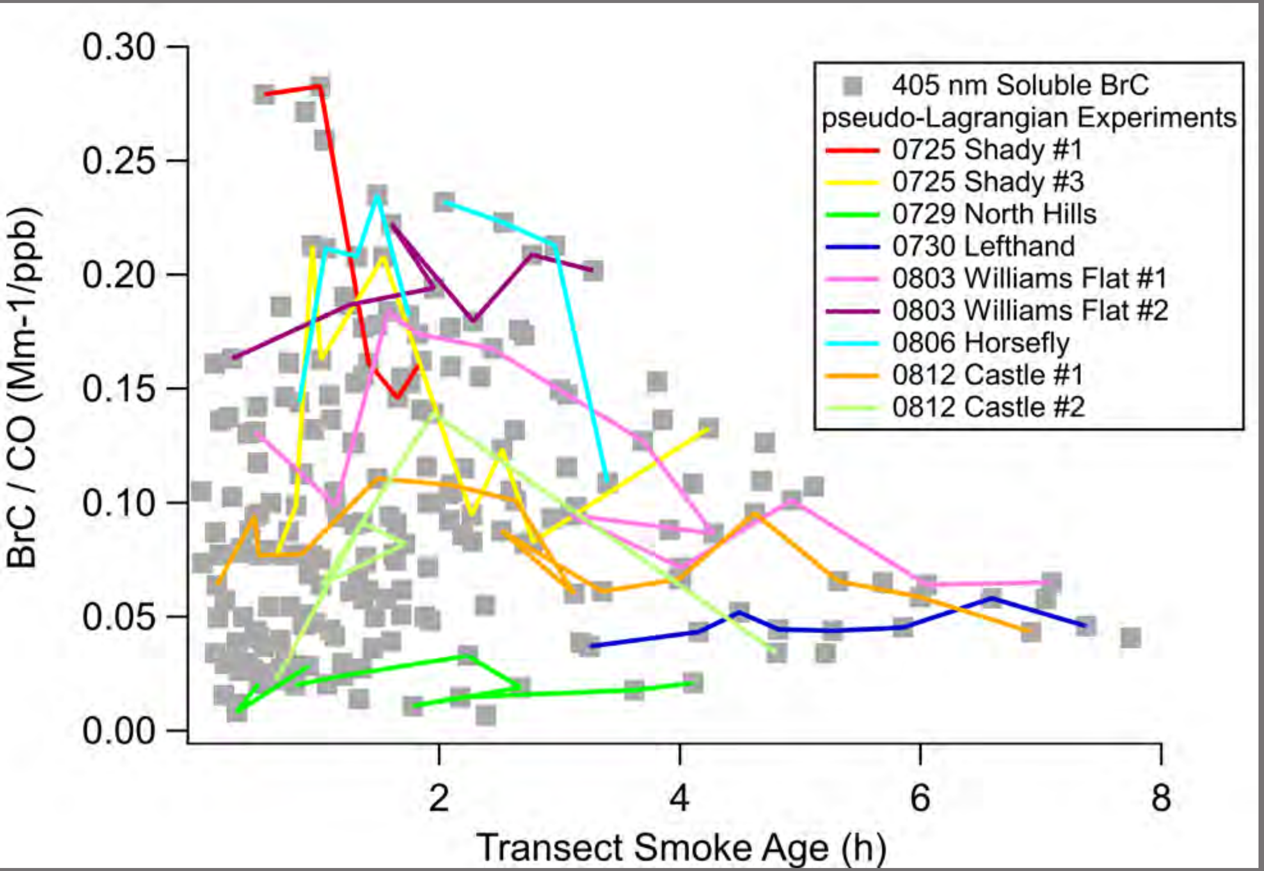




BrC absorption at 365 nm shown for all data sets.

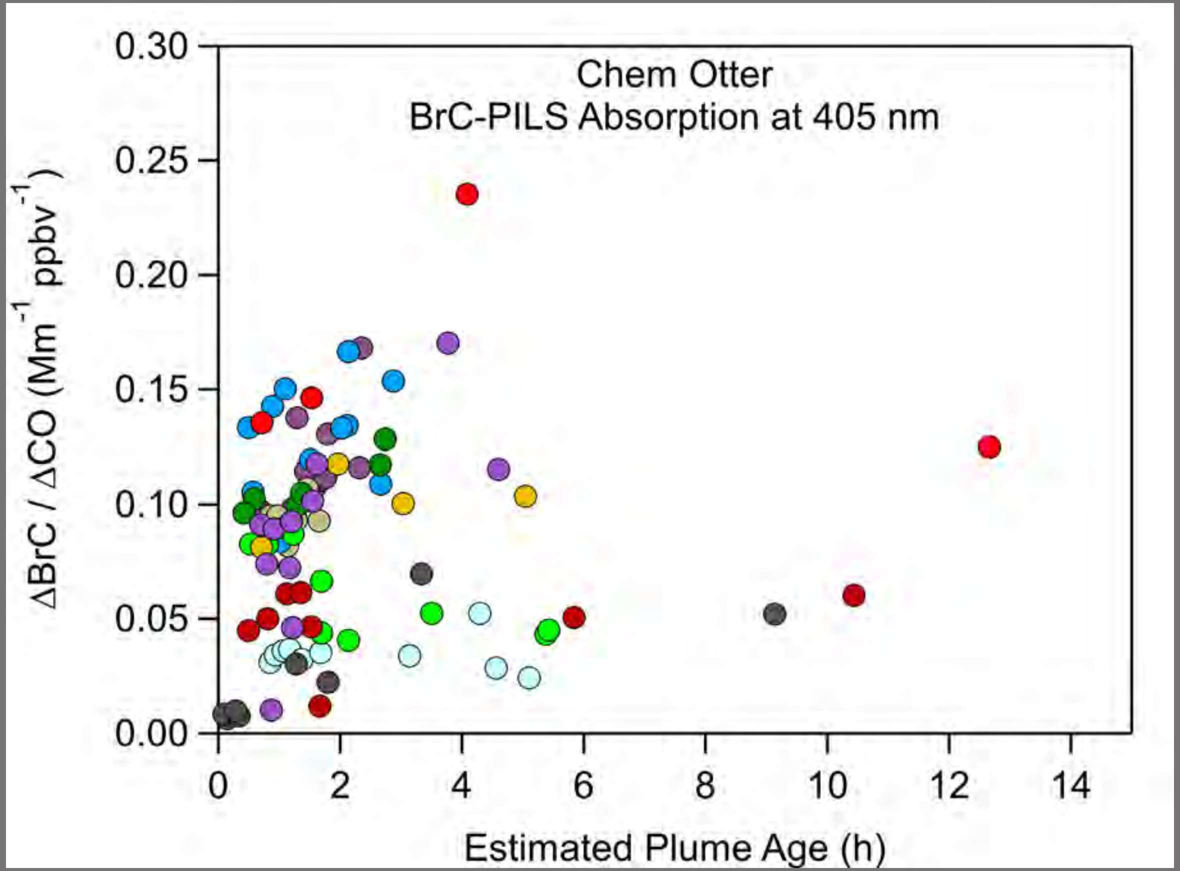
Trends for ΔBrC / ΔCO are similar for the DC-8

DC-8 Water-Soluble BrC



Plot from Nick Wagner using Georgia Tech measurements

Twin Otter Water-Soluble BrC



BrC absorption at 405 nm shown for all data sets.

Key results and future work

Key results:

1. Brown carbon / CO often increased during the first 10 hours of plume aging in the Twin Otter measurements.
2. This is consistent with initial results from the DC-8, but inconsistent with two published studies.

Improvements and future work:

- Analyze brown carbon lifetimes as a function of “chemical clocks”
- Consider sunset and night flights
- Consider other aerosol parameters (AAE, AMS O/C) as a function of plume age
- Calculate the range of radiative forcing impacts of different brown carbon refractive indices and lifetimes

Need from others:

- Final reanalysis plume ages from Chris Holmes

Chemical Imaging of Atmospheric Biomass Burning Particles from North American Wildfires: Daytime vs Nighttime Samples

F. A. Rivera-Adorno,[1] J. M. Tomlin,[1] R. Washenfelder,[2] A. Middlebrook[2], S. China,[3] D. Knopf,[4] R. Moffett,[5] L. Azzarello,[6] A. Franchin,[7] A. Laskin[1]….

[1]Department of Chemistry, Purdue University, West Lafayette, IN, USA

[2]National Oceanic and Atmospheric Administration, Chemical Science Division, Boulder, CO, USA

[3]Pacific Northwestern National Laboratory, Environmental Molecular Sciences Laboratory, Richland, WA, USA

[4]School of Marine and Atmospheric Sciences Department, Stony Brook University, Stony Brook, NY, USA

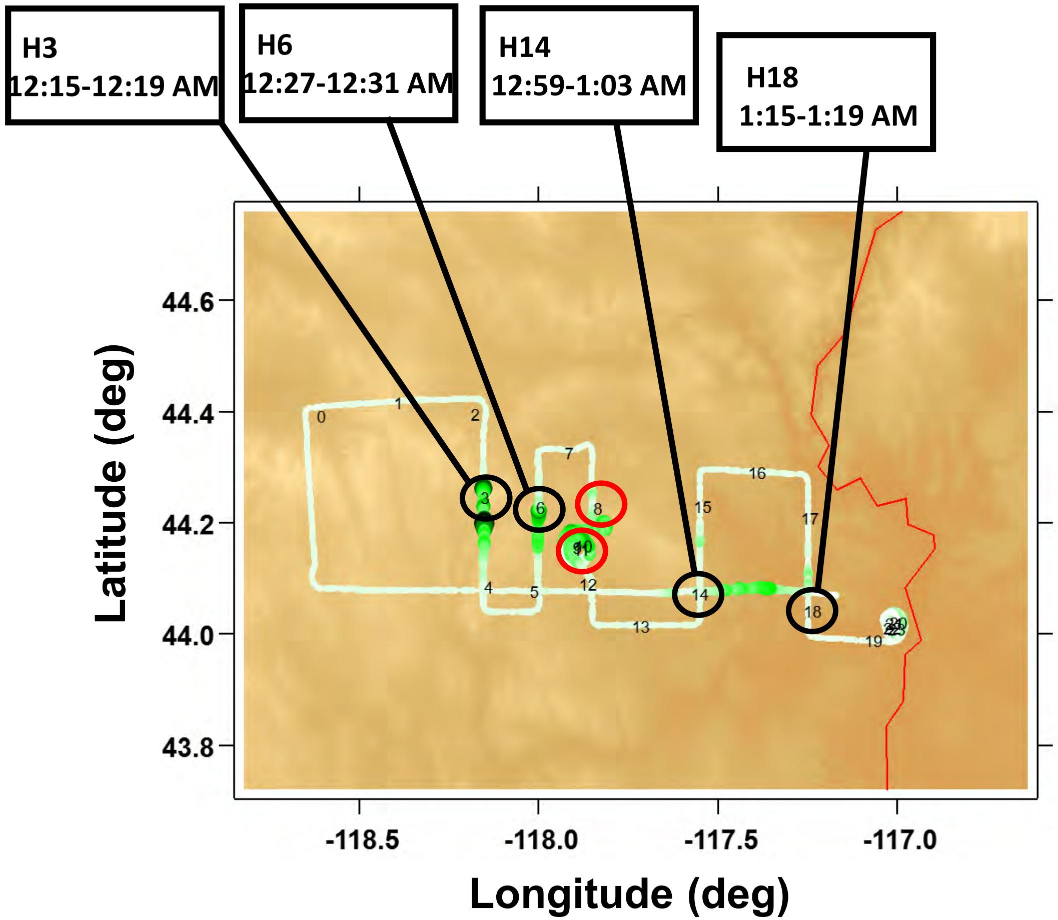
[5]Sonoma Technology, Petaluma, CA, USA

[6]Department of Chemistry, York University, Toronto, ON, Canada

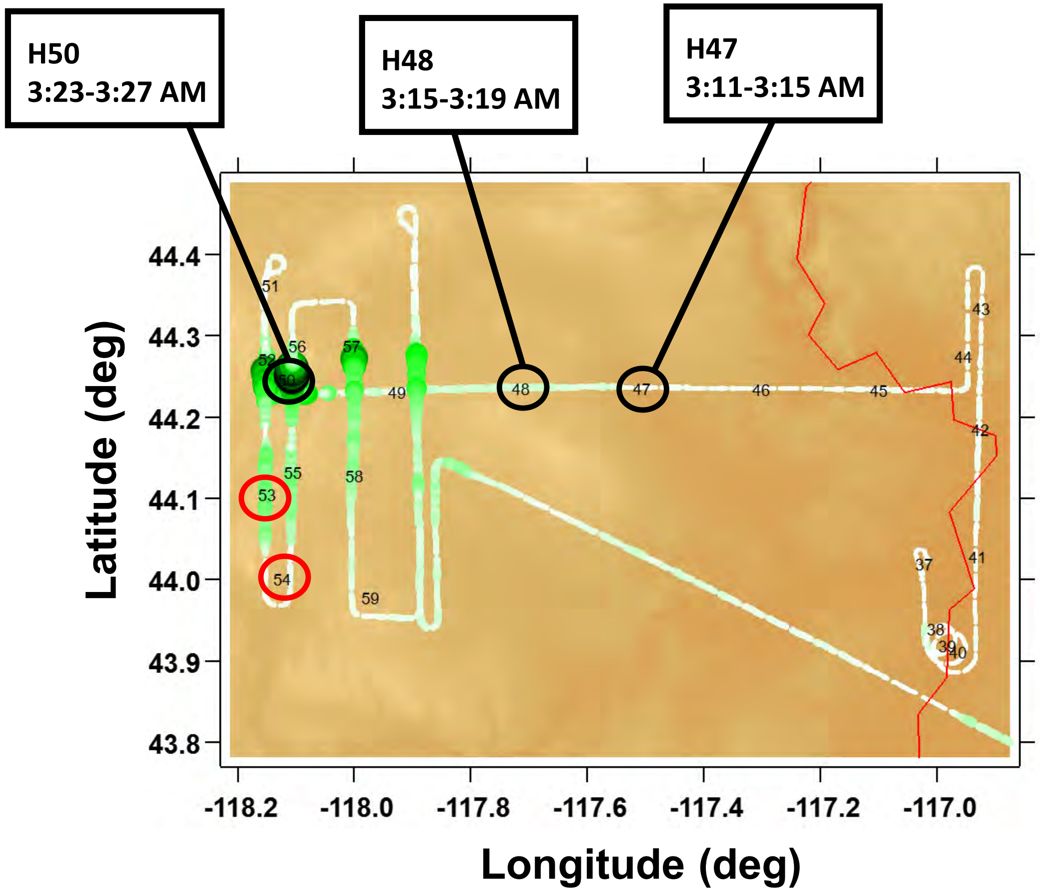
[7]National Center for Atmospheric Research, Boulder, CO, USA

Sampling: 8-28-2019

L2 Flight: Afternoon



L3 Flight: Nighttime



Analyzed Samples: H3, H6, H8, H9, H10, H14, and H18
Samples for STXM Analysis: H3, H6, H14, and H18

Analyzed Samples: H47, H48, H50, H53, and H54
Samples for STXM Analysis: H47, H48, and H50

Key Results: Chemical Imaging Analysis

PURDUE UNIVERSITY Chemistry

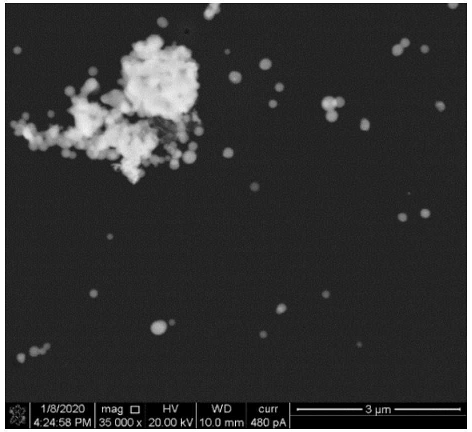
CCSEM/EDX particle-type grouping based on <u>elemental composition</u>

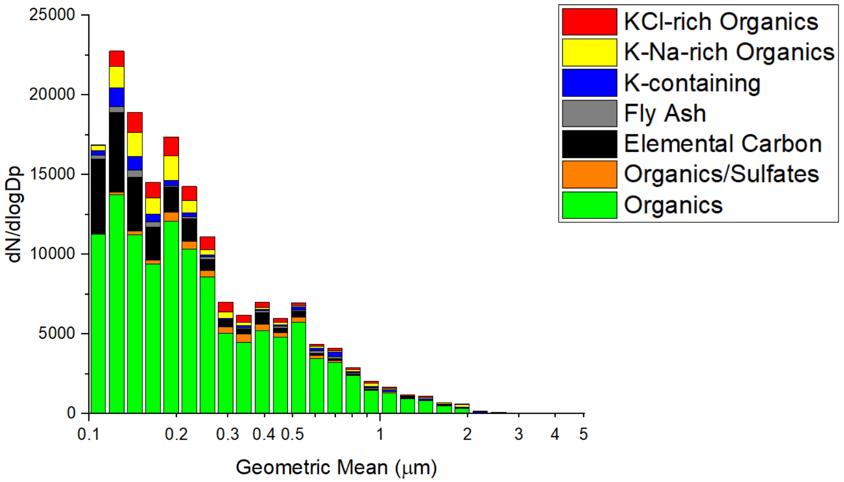
illustrates *external mixing* of individual particles: organic particles dominate; EC (soot) particles apparent in the daytime samples; inorganic salts are more significant in the nighttime samples.

STXM particle-type grouping based on <u>carbon speciation</u>

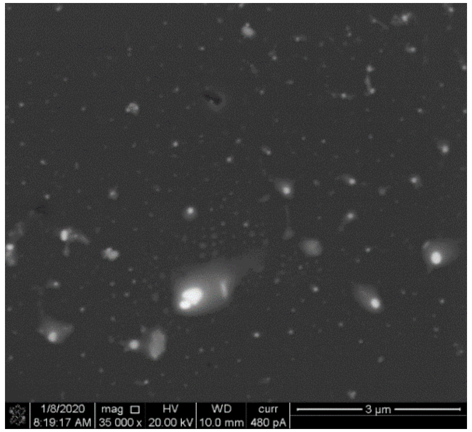
illustrates *internal mixing* of individual particles: mixtures of organic carbon (OC) and elemental carbon (EC) are common in the daytime samples; the night-time samples shows more complex mixtures of OC, EC and inorganic (IN) material

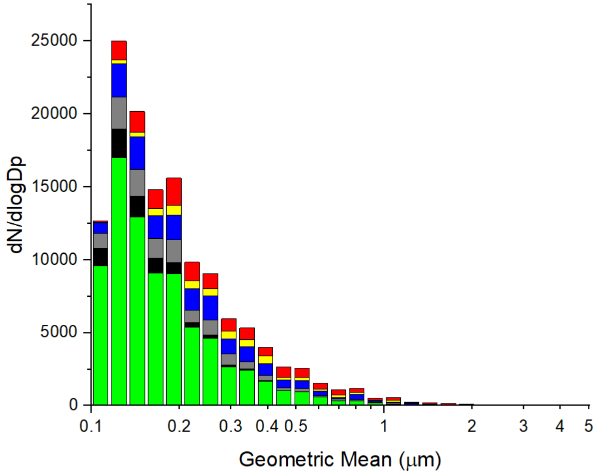
Day time

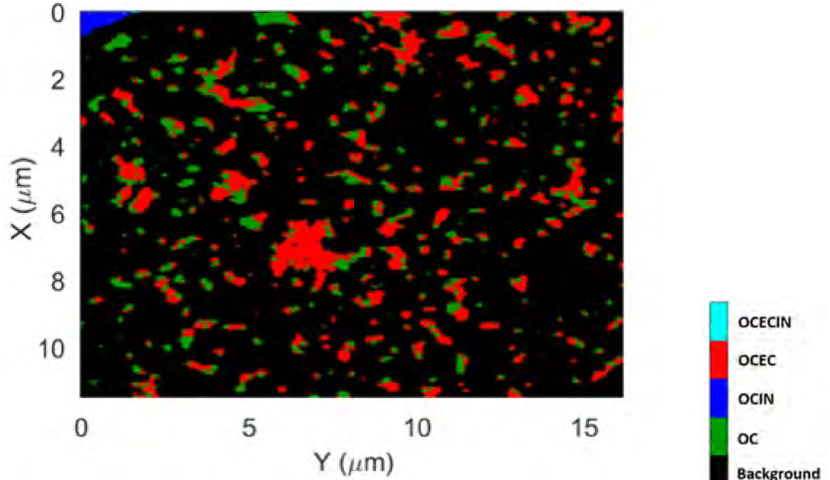


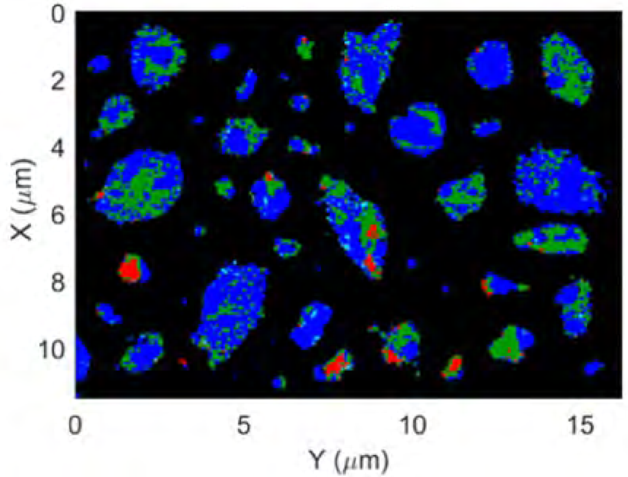


Night time





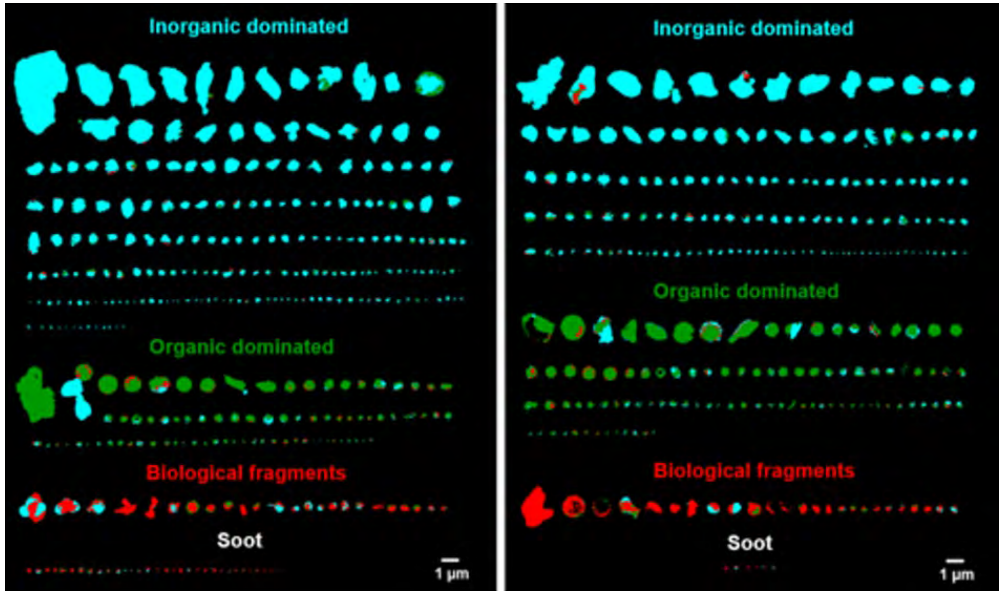




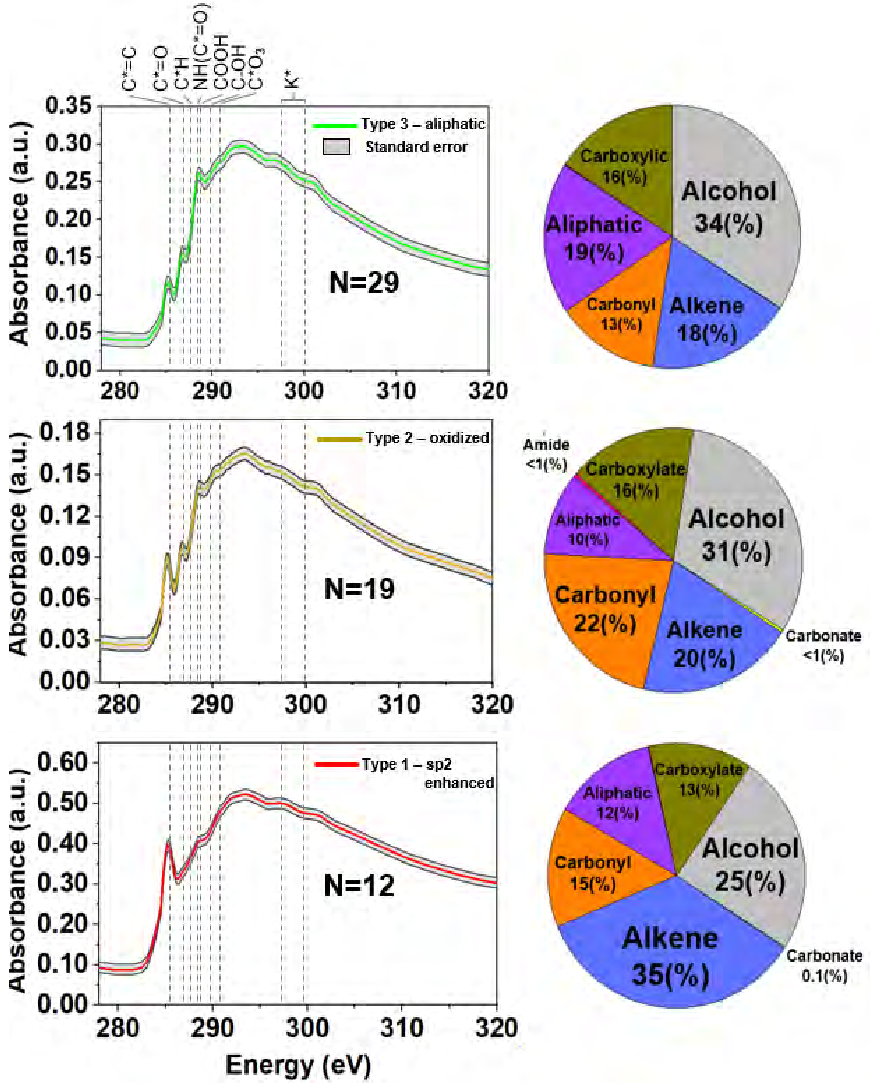
Remaining Work: STXM/NEXAFS Sample Analysis

Images from Tomlin et. al. 2020. ACS Earth Space Chem. Submitted

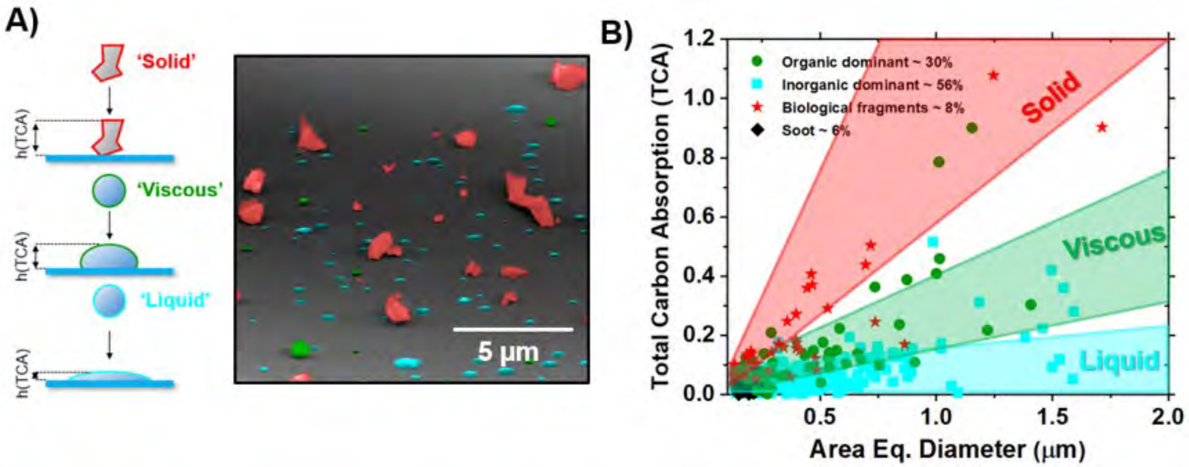
Particle internal heterogeneity and contribution of each particle type based on STXM compositional maps .



Carbon type particles differentiation and carbon functional groups contribution based on STXM absorption spectra.



Particle viscosity estimation based on Total Carbon Absorption (TCA) to further study differences in morphology between day and nighttime.

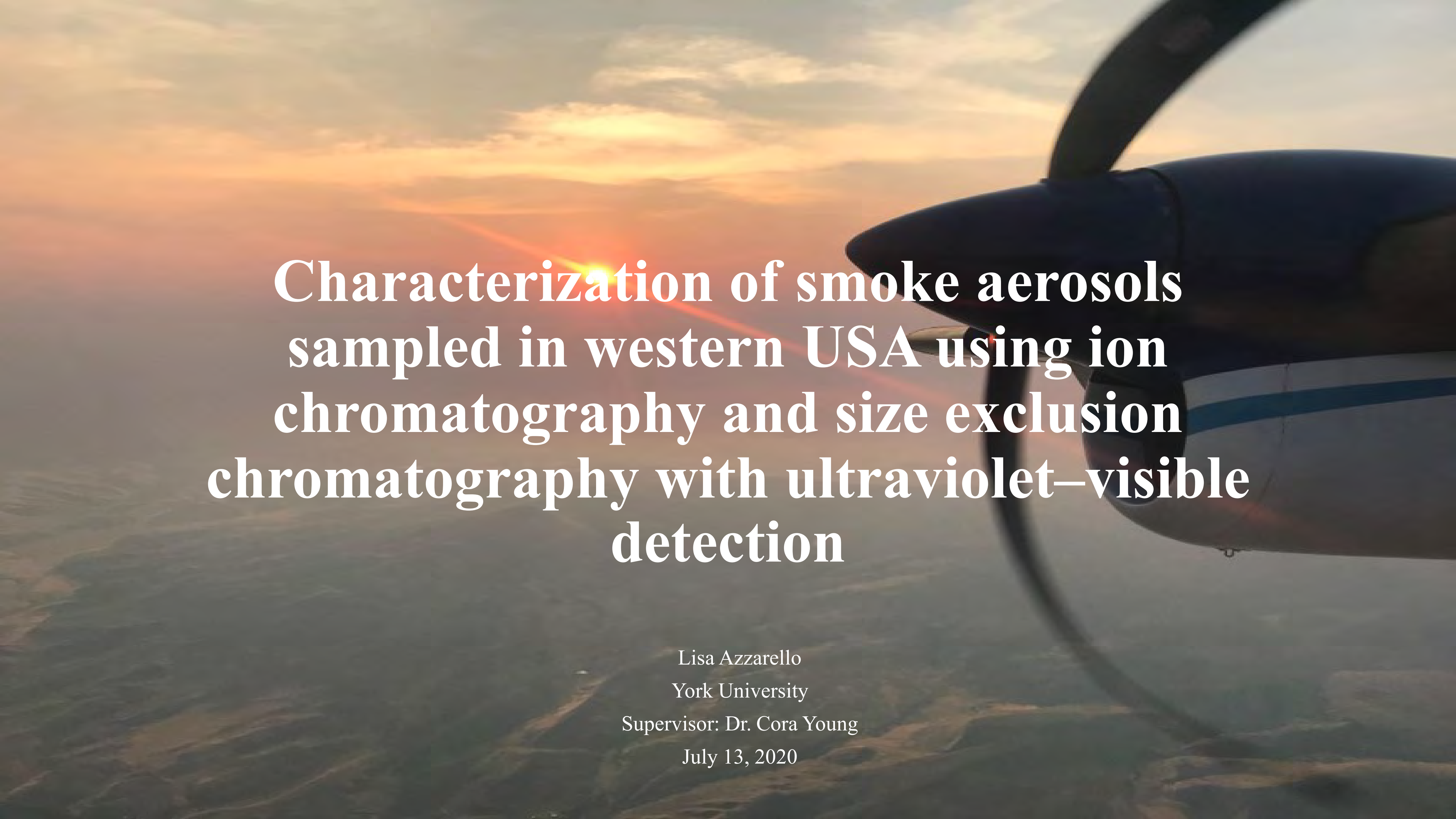


PURDUE UNIVERSITY Chemistry

Information 'Wish list' from other researches to guide our particle analysis at CLS :

- Chemical differences of particle-phase organics as detected by AMS and gas-phase organics (CIMS?)
- Differences in PSD of organics and other main aerosol types as detected by AMS and SMPS?

Thank You!



Characterization of smoke aerosols sampled in western USA using ion chromatography and size exclusion chromatography with ultraviolet–visible detection

Lisa Azzarello

York University

Supervisor: Dr. Cora Young

July 13, 2020

Sample Collection

- Brown-Carbon Particle into Liquid Sampler (BrC-PILS)
  - Collected WSOC into falcon tubes for offline analysis
- Continuous Light Absorption Photometer (CLAP): measures light absorption of particles deposited onto a filter
  - Deposition occurs on a single spot with up to 8 spots upon rotation of the solenoid valve
  - Extract spots for offline analysis

Chromatographic Separation of WSOC

- BrC-PILS collected WSOC into falcon tubes and CLAP collected filter samples for offline analysis
  - Ion Chromatography with Conductivity Detection (IC-CD)
    - Cation mode: Na^+, NH_4^+, K^+ and 11 alkylamines
    - Anion mode: Cl^- , NO_2^-, NO_3^- , SO_4^{2-} , and PO_4^{3-}
  - Size Exclusion Chromatography with UV-Vis Detection (SEC-UV)
    - SEC column: separation of molecules as a function of size
    - Diode Array Detector (DAD): provides wavelength range from 190 – 800 nm
    - Absorption spectrum based on molecular size is generated

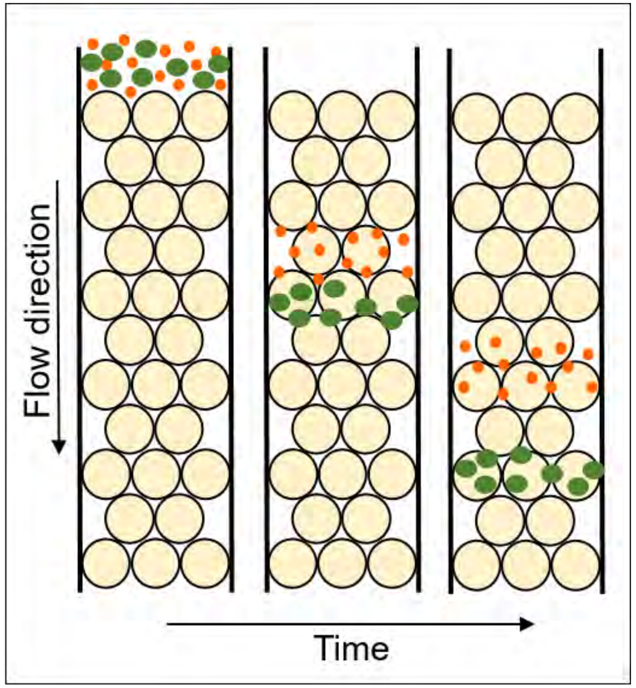


Figure 1. Principle of Size Exclusion Chromatography.

Previous Results Using SEC-UV

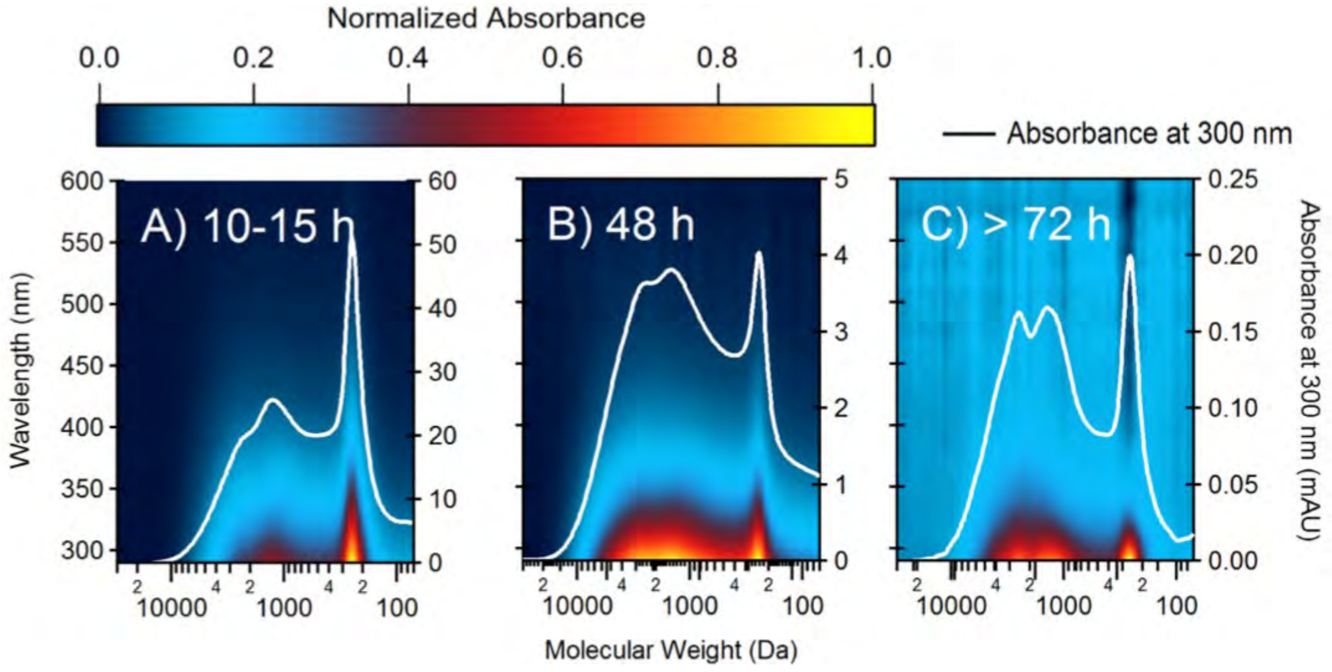


Figure 2 Absorption profiles as a function of molecular weight across various sampling regions and plume age.[1]

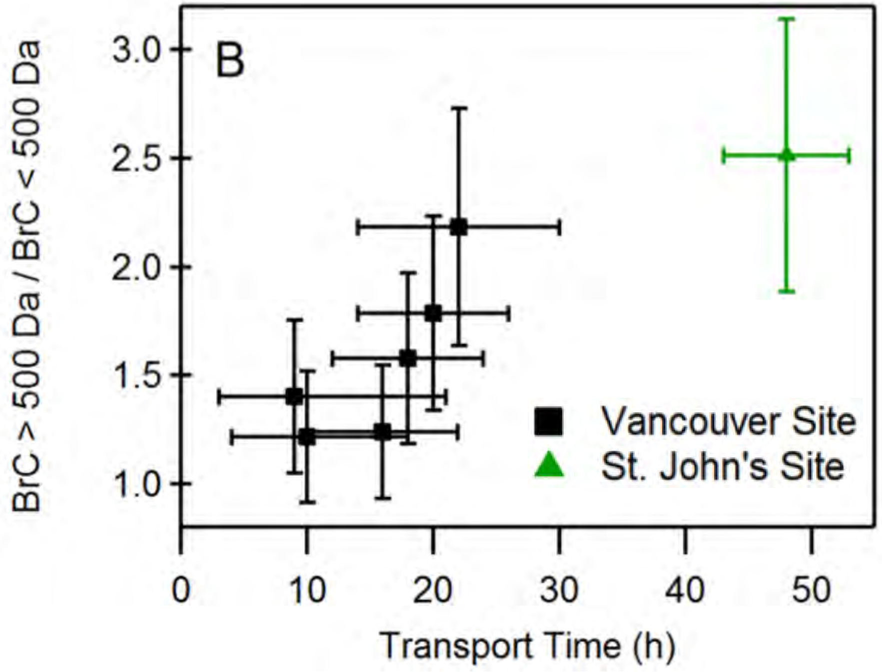


Figure 3. Absorbance ratio for large molecules (>500 Da) to small molecules (<500 Da) as a function of plume age.

[1]Di Lorenzo RA, Washenfelder RA, Attwood AR, et al. (2017) Molecular-Size-Separated Brown Carbon Absorption for Biomass-Burning Aerosol at Multiple Field Sites. *Environ Sci Technol*. 2017;51(6):3128-3137. doi:10.1021/acs.est.6b06160

Preliminary Results

Number o Samples Colle	nalyzed
281	

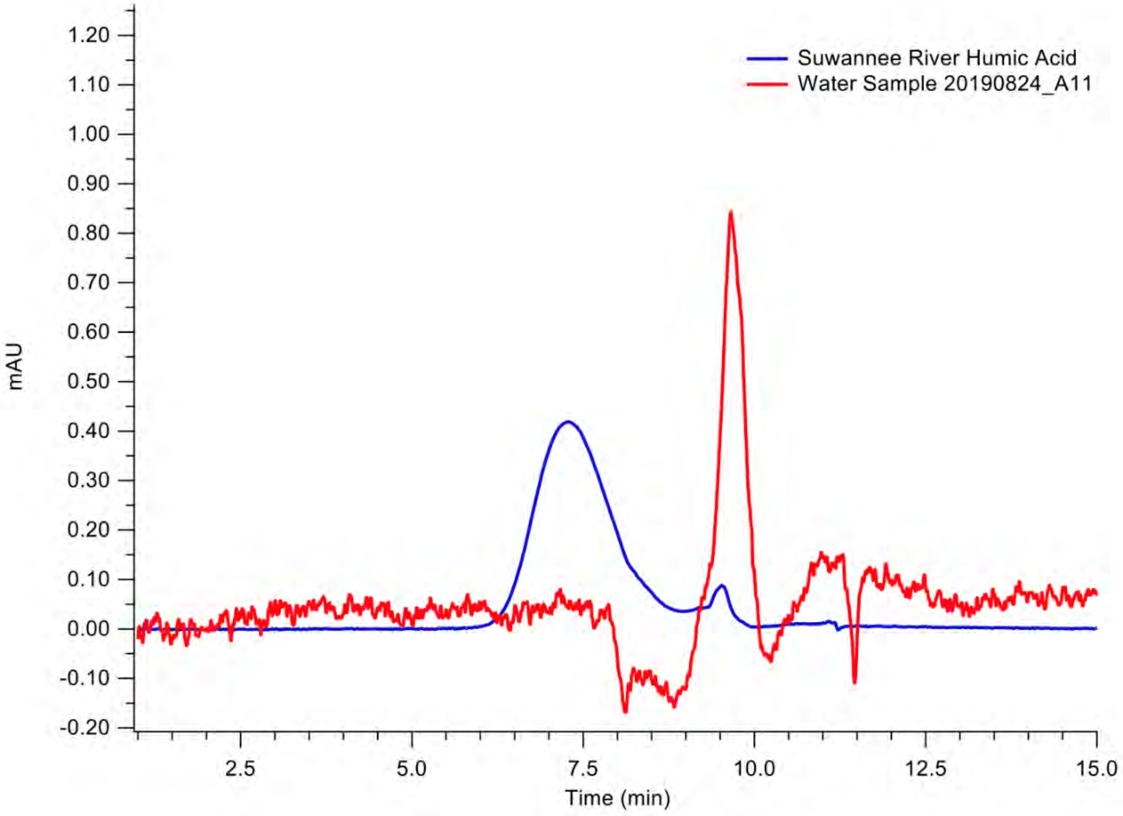
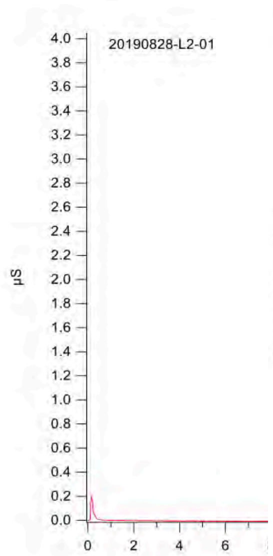


Figure 4. Absorption profiles at 300 nm of Suwannee River humic acid and a FIREX-AQ water sample collected on August 24, 2019.



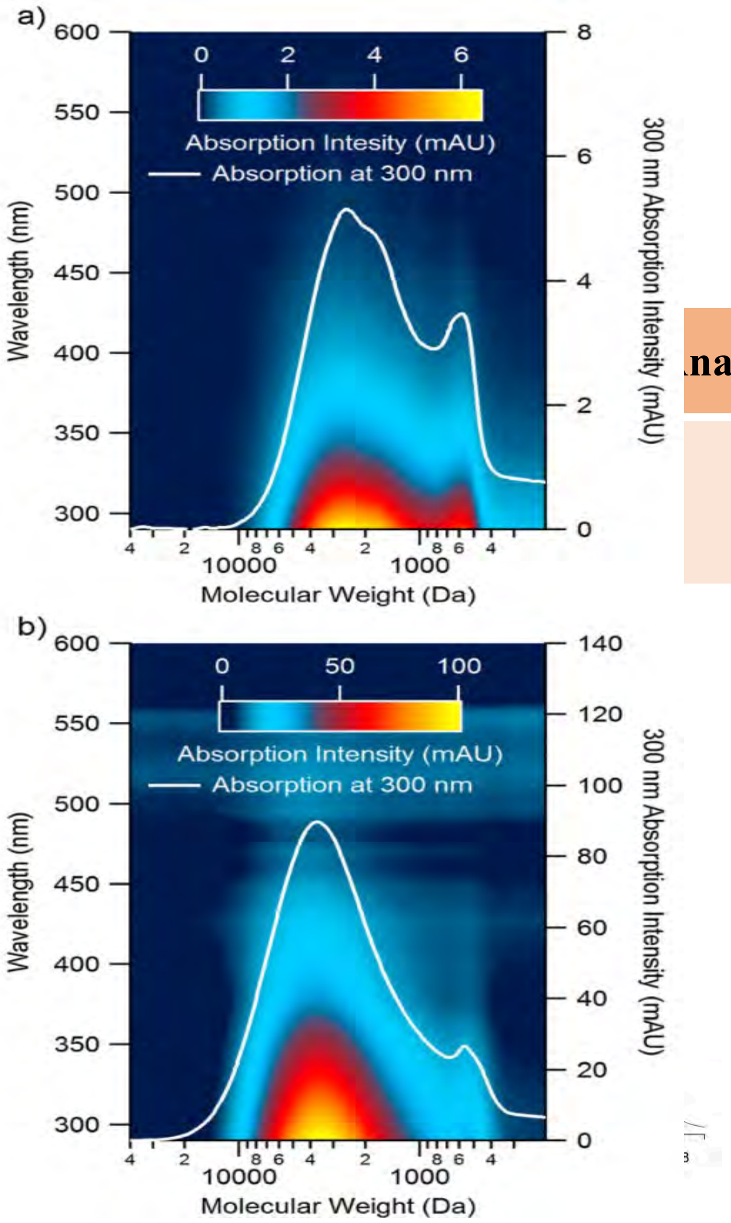


Figure 5. Absorption density plots at 300 nm of a) biomass burning extract and b) Suwannee River humic acid.[2]

Figure 6. IC-CD chromatograph of water sample collected on August 28, 2019.

[2]Di Lorenzo, R. A., and Young, C.J. (2016), size separation method for absorption characterization in brown carbon: application to an aged biomass burning samples, Geophys. Res. Lett., 43, 458-465, doi: 10.1002/2015GL066954

Remaining Work

- SEC-UV
  - Run remaining water samples
  - Extract CLAP filters
  - Continue to compare results to online absorption data
- IC-CD
  - Complete cation mode
  - Confirm presence of ions with mass spectrometry
- Deduce trends as a function of plume age
- Write and publish results

Thank you for listening!

Special thanks to:

Dr. Cora Young

Dr. Trevor VandenBoer

CJY & VDB group members

Dr. Rebecca Washenfelder

Twin Otter Crew

Effects of emissions, transport, and chemistry on prediction of air quality impacts from fires

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[2] NOAA ESRL Chemical Sciences Laboratory, USA
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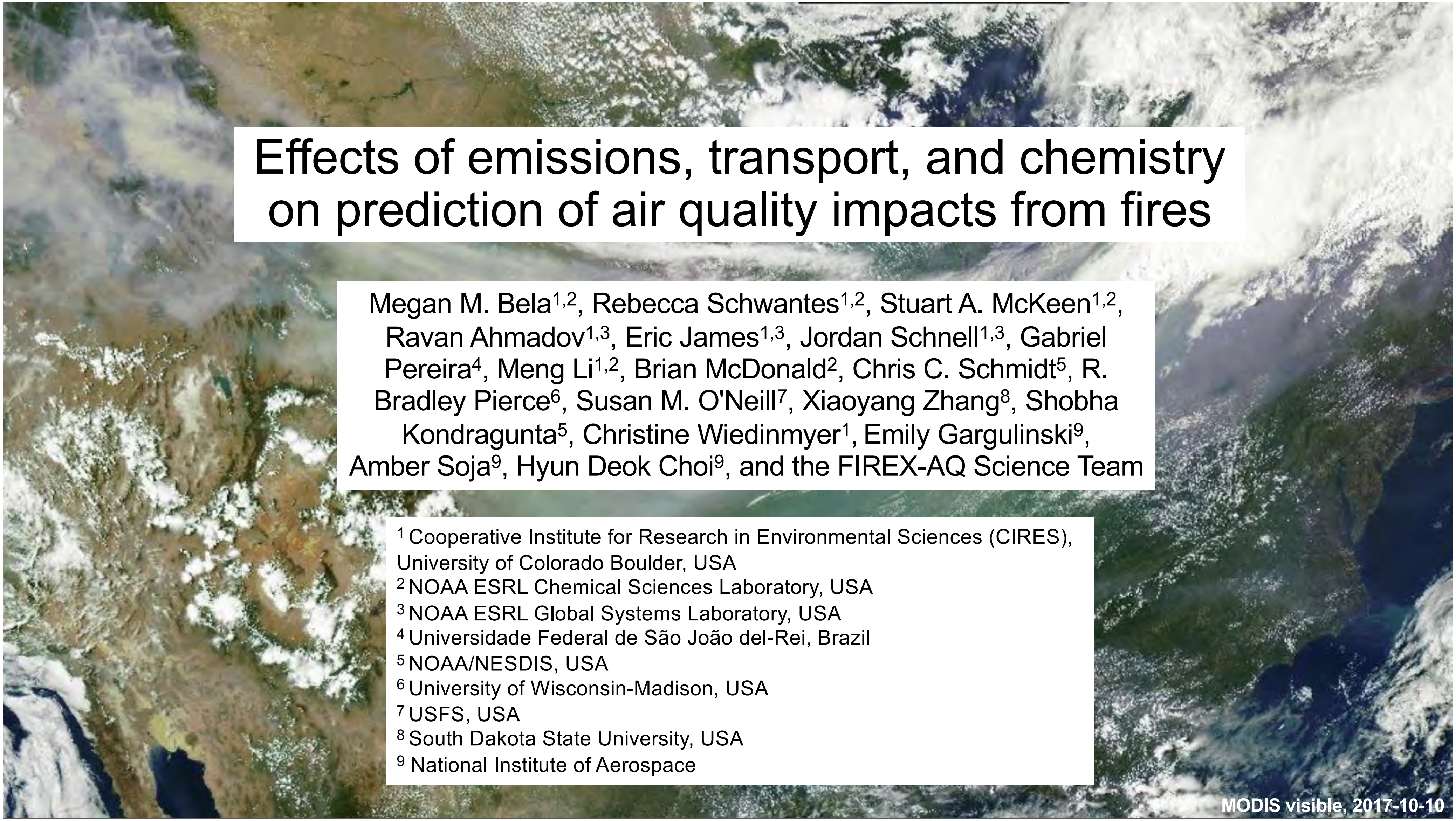
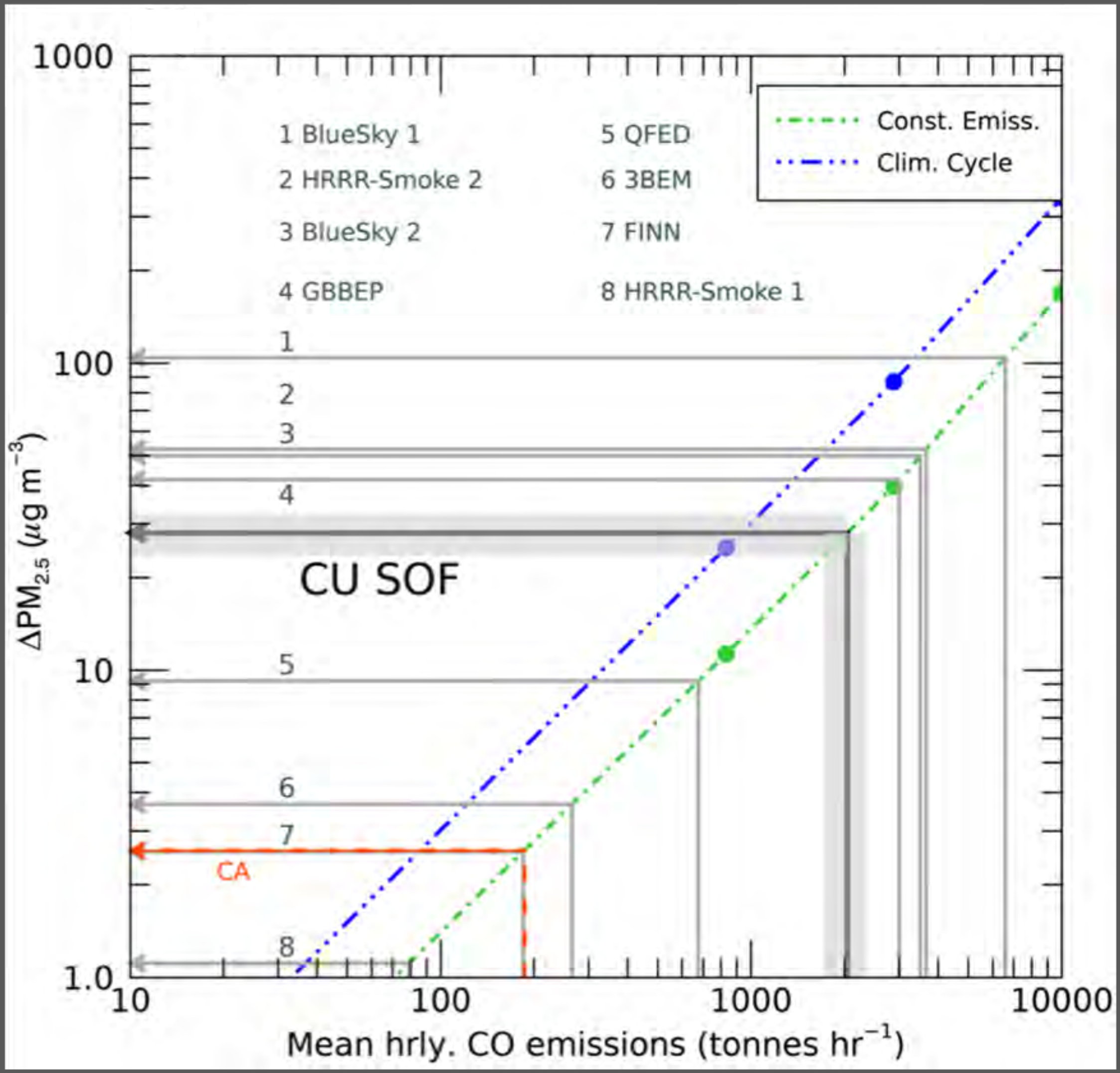
[6] University of Wisconsin-Madison, USA
[7] USFS, USA
[8] South Dakota State University, USA
[9] National Institute of Aerospace



Motivation

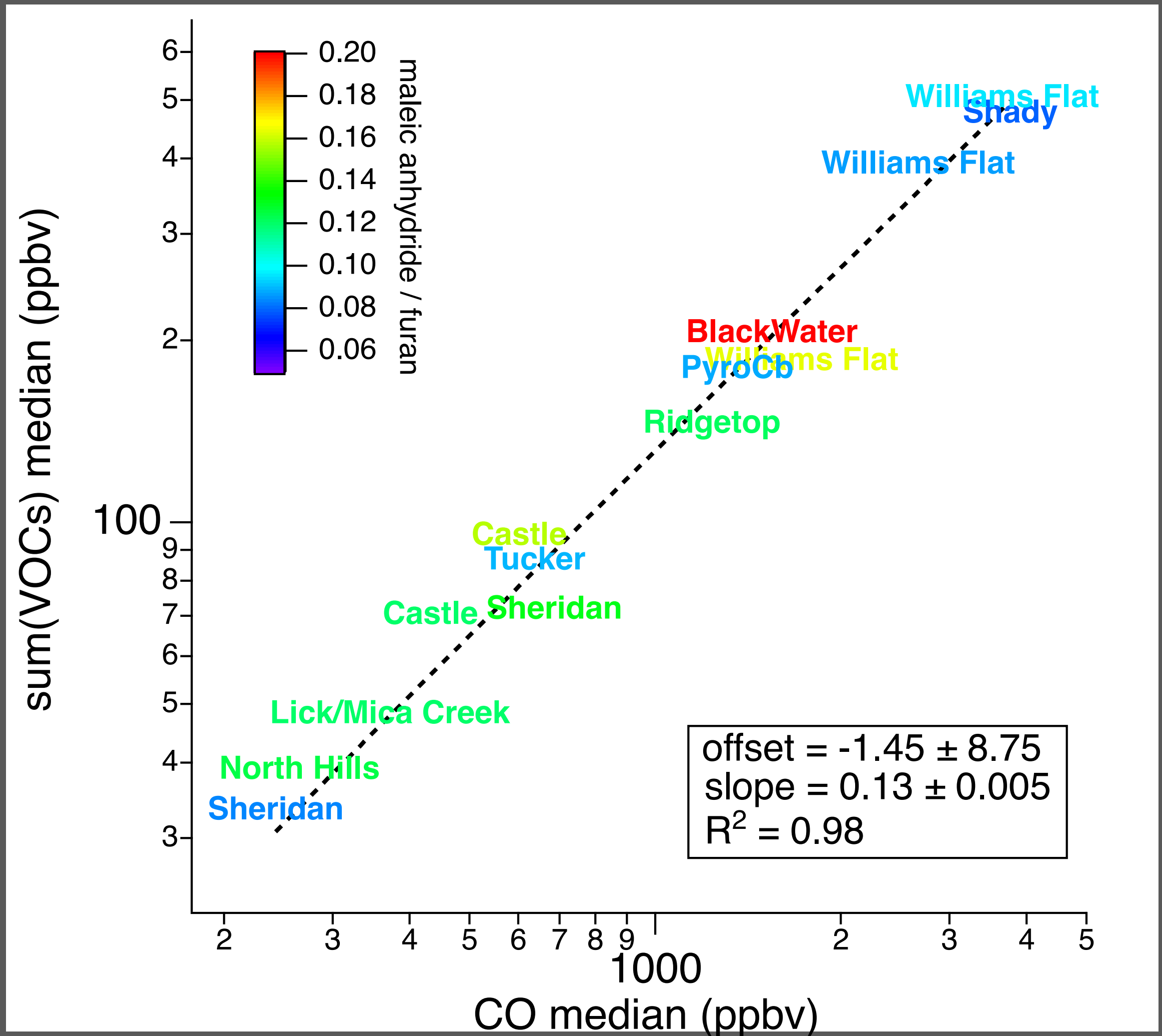
Emissions from satellite-based inventories vary widely



Factor of 83 variation in CO emissions for Oct. 2017 N. CA fires

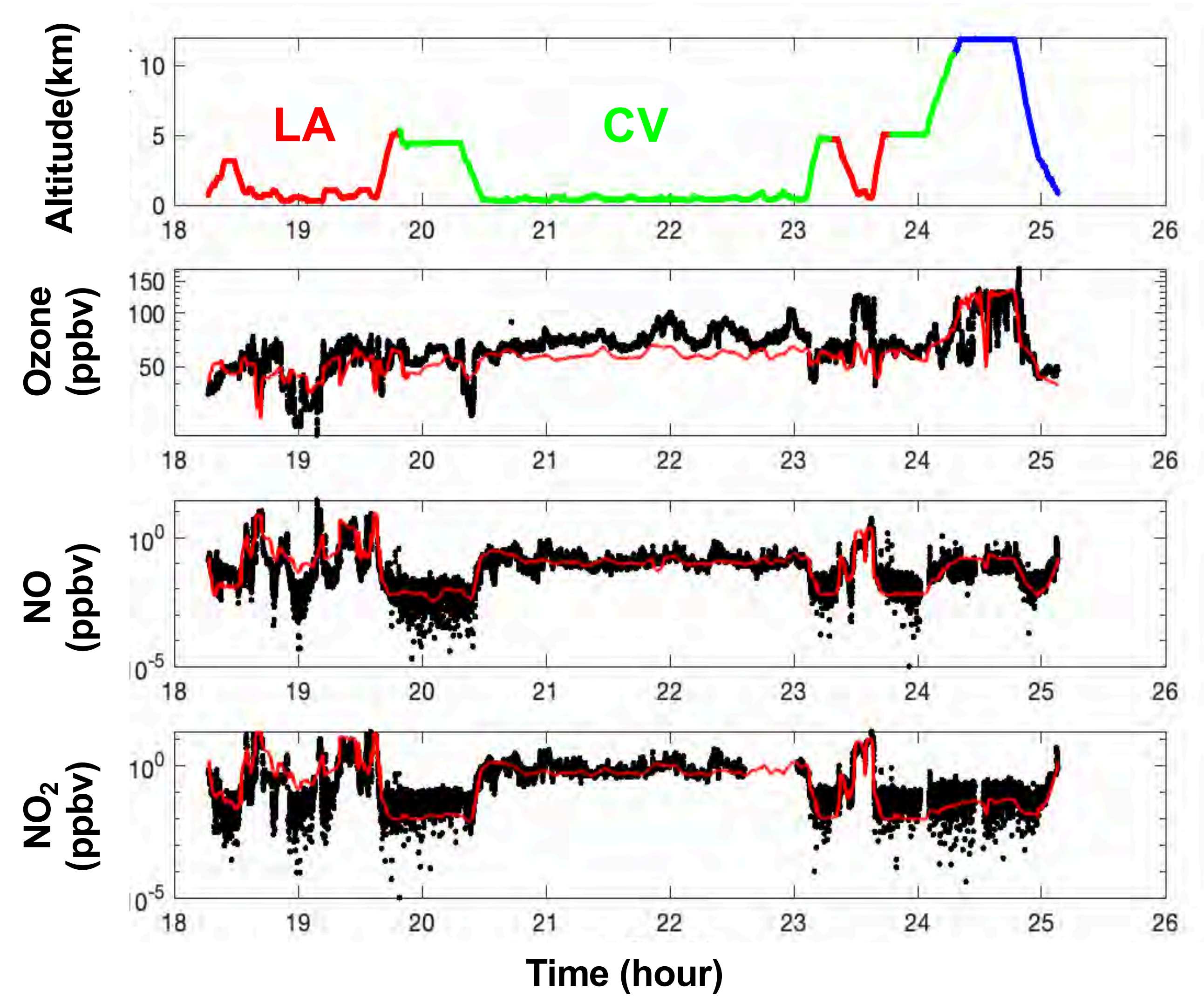
Bela et al. (2020), in prep.

Motivation **VOC fire emissions function of vegetation type, fuel amounts, fire conditions**



G. Gkatzelis

Results WRF-Chem simulations with FIVE NO_x consistent with Los Angeles samplng during FIREX-AQ 2019



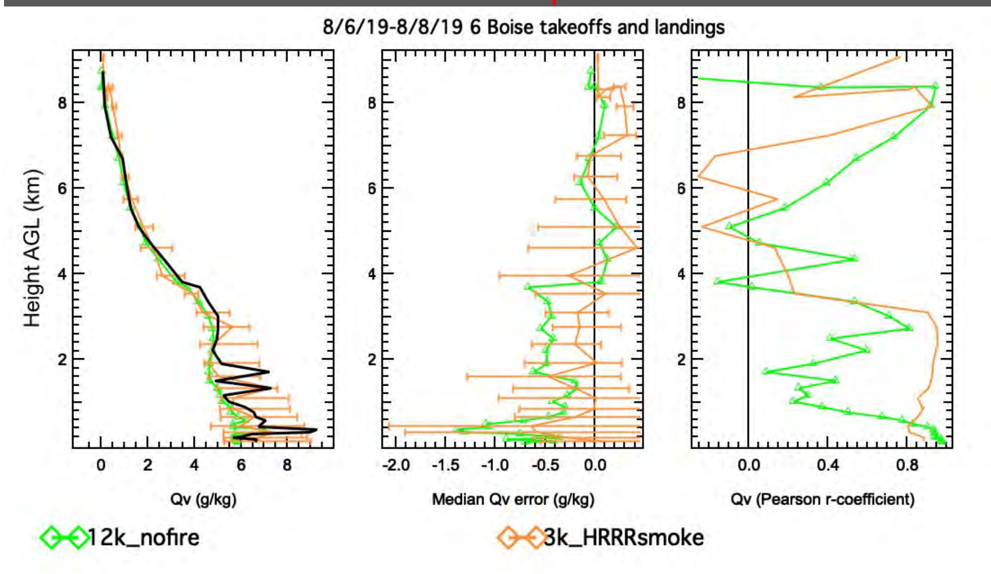
Absolute bias (ppbv)

	O_3	NO	NO_2
CV	-13.7826	-0.0179	-0.2158
LA	-8.5718	0.1711	1.3824

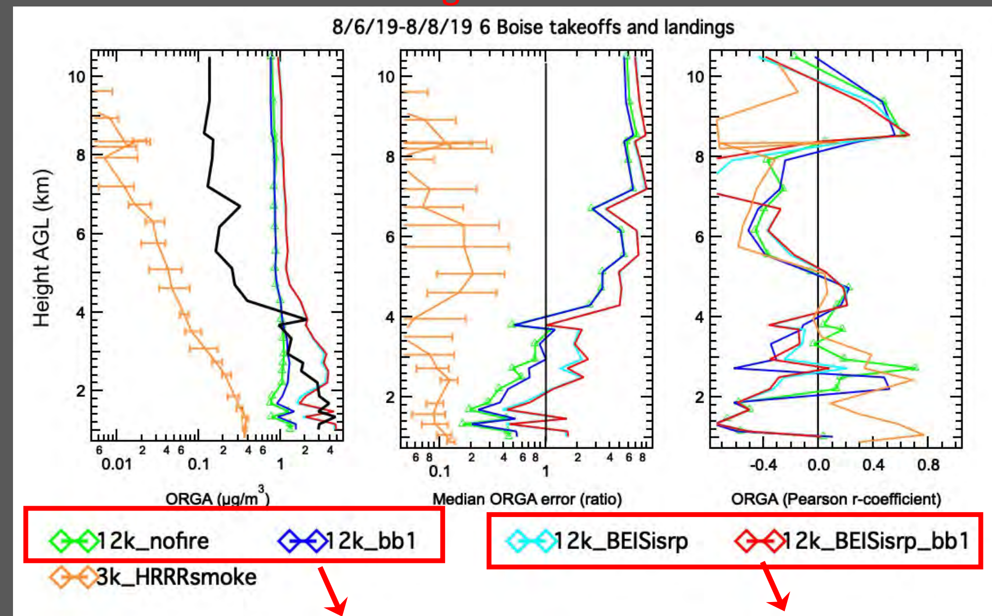
Results

Examples of DC-8 statistics for 6 Boise landing/takeoffs during the 2019 FIREX-AQ experiment (3 days of HRRR-Smoke, WRF-Chem overlap)

Water vapor



Organic Aerosol



MEGAN biog.: isoprene*0.5

BEIS biog.

Water vapor:

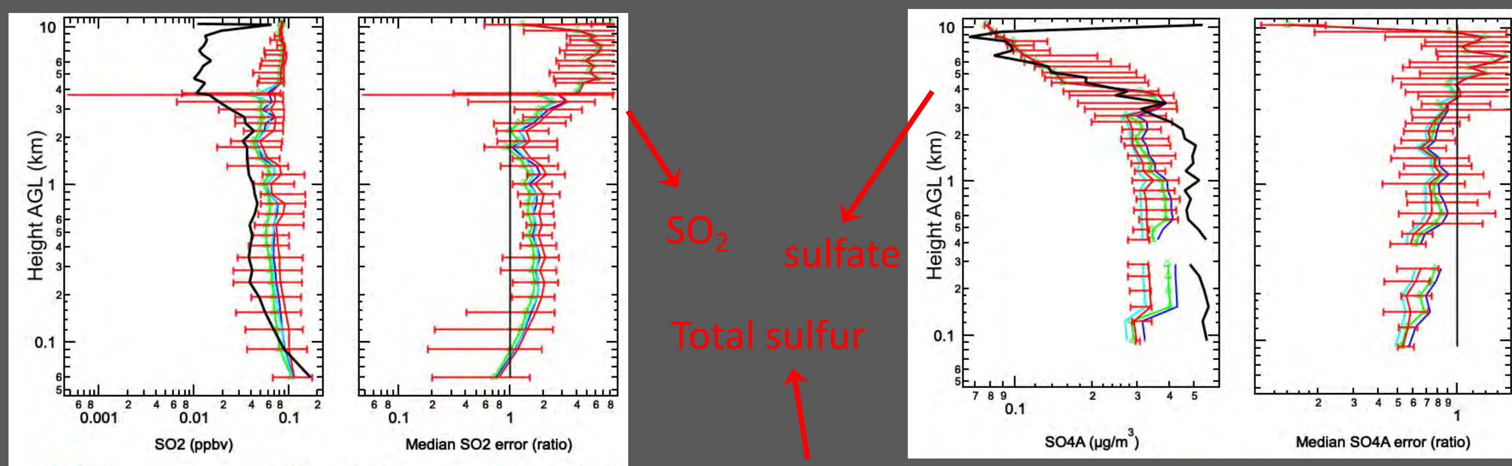
- WRF/Chem loss of correlation from .5 to 3 km AGL, Is this model resolution, PBL scheme,….??

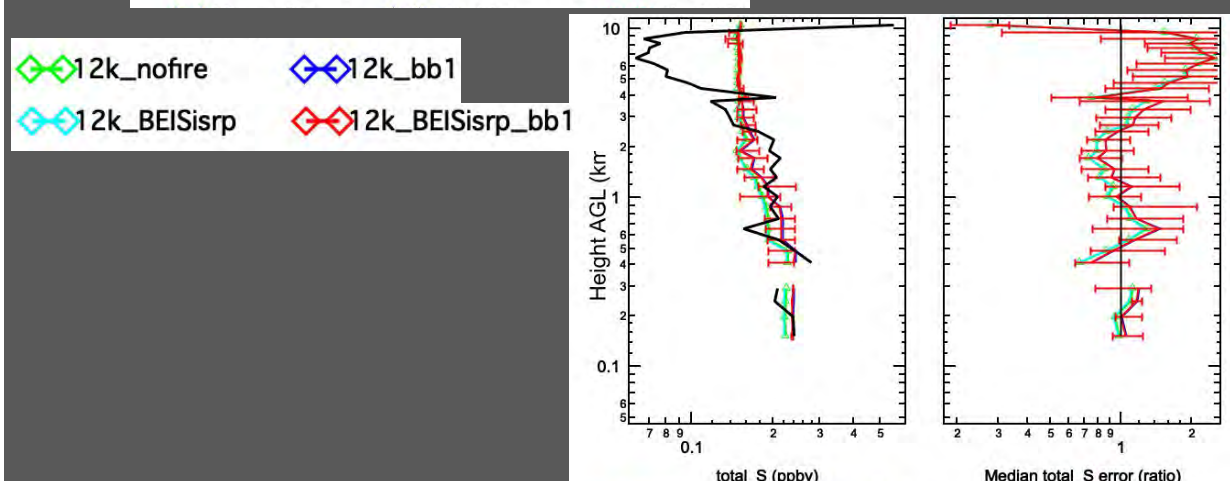
Organic Aerosol:

- Fires have little influence over Boise during FIREX-AQ
- WRF/Chem ORGA is mostly biogenic (BEIS emissions are much higher than MEGAN with .5*isoprene)
- High WRF/Chem bias above the PBL

Results

Examples of DC-8 statistics for 27 Boise landing/takeoffs during the 2019 FIREX-AQ experiment (WRF-Chem, 4 model cases, 7/22/19-8/16/19)



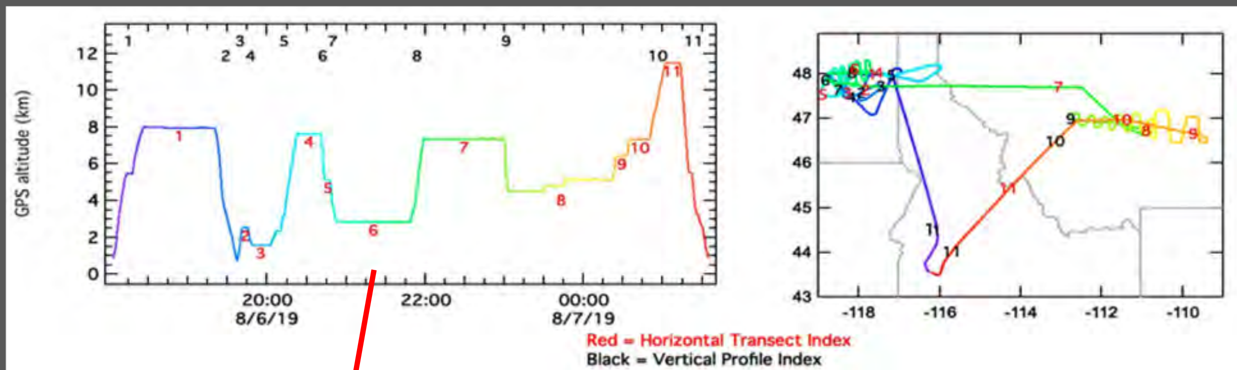


Below 2-km:

- SO_2 a factor of 2 too high
- Sulfate ~ 30% too low
- Total sulfur within ~25%
- A biogenic inventory dependency to partitioning
- Comparisons can be used to validate cloud oxidation in model

Results

Examples of DC-8 oxidant comparisons (PAN HNO_3, and CH_2O) during FIREX-AQ (WRF/Chem, 4 model cases, 7/22/19-8/16/19)



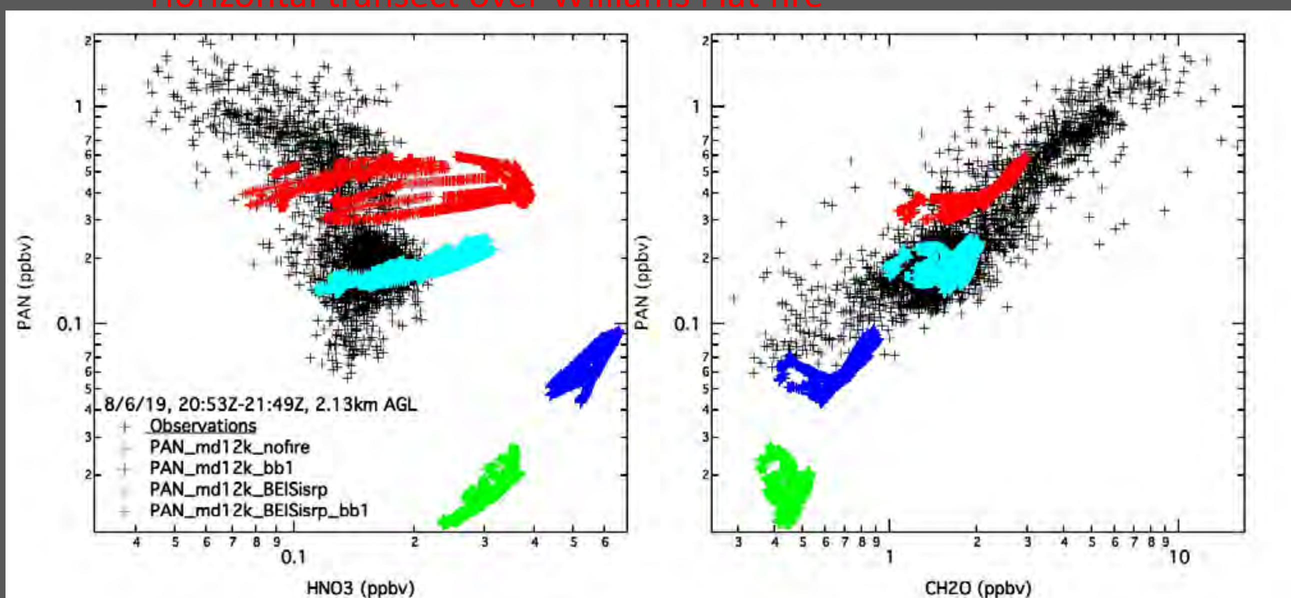
(MEGAN, .5*isoprene)

12k_nofire 12k_bb1

(BEIS)

12k_BEISisrp 12k_BEISisrp_bb1

Horizontal transect over Williams Flat fire



Below 2-km:

- Biogenic inventory dependence
- Fire dependence reasonable for PAN versus CH_2O (using BEIS)
- PAN versus HNO_3 inconsistent between observations and model

Remaining work

CSL Fire Emissions Research and Development

Emission factors (EFs)

- Update prep_chem_src EFs with values from literature and FIREX-AQ
- Add new speciation/species
- Update vegetation data and classification
- Scale emissions based on FIREX-AQ observations

Emissions for FIREX-AQ period

- Compare emissions from satellite-based inventories (FINNv2, GBBEPx, Bluesky, GFAS, QFED, GFED, Soja et al.)
- FRP emissions and plume rise for full chemistry
- GOES-16 diurnal cycle

WRF-Chem simulations at 12 and 4 km for FIREX-AQ period

- T1 chemistry
- New species/reactions
- Evaluation against FIREX-AQ observations
- Air quality impacts