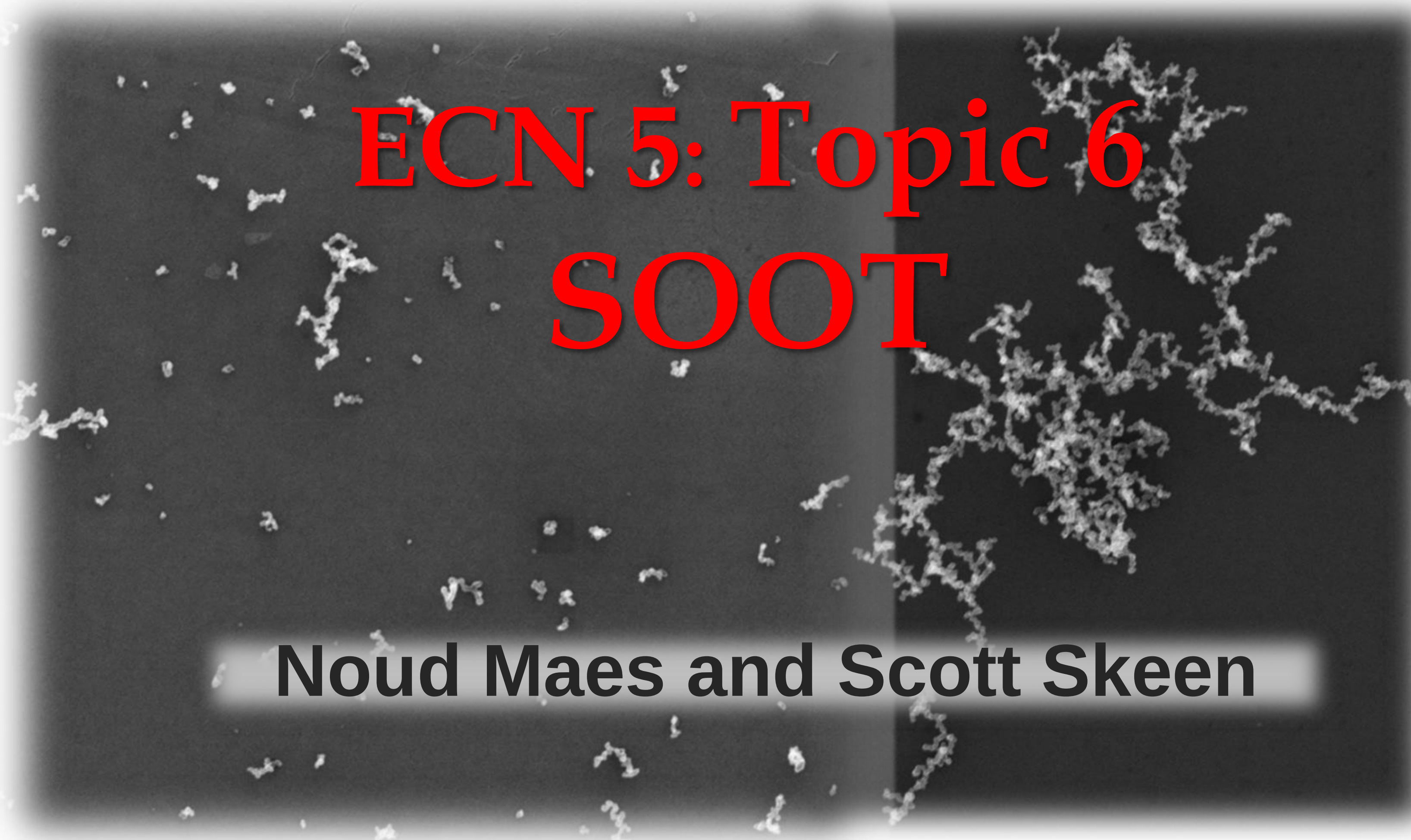
ECN 5: Topic 6
SOOT



Noud Maes and Scott Skeen

Outline

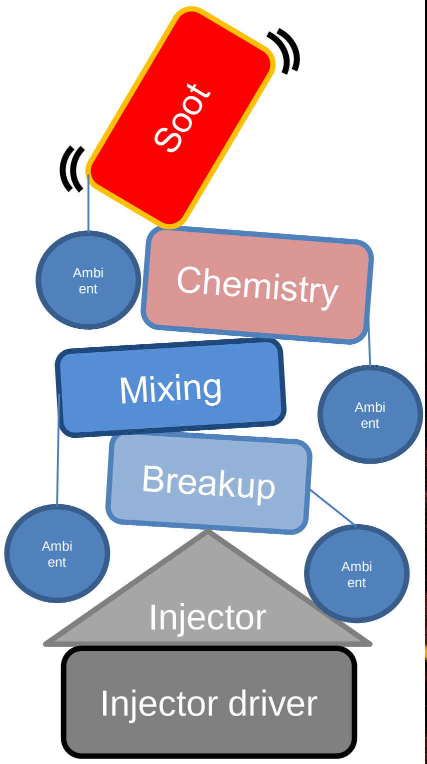
- Introduction to New Experimental Data
  - Focus on comparison of Spray C/D Data Across Institutions
  - Discussion of new Spray A data from GMR and Sandia
- Experimental Setups
- Data acquisition and Post-processing Challenges
  - Negative Image Lag
  - Beam Steering
  - Incident wavelength—soot refractive index
- Spray C/D Coparison
  - IFPEN, CAT, Sandia
- Spray C/D – 1200K short injections
- Spray A pre-burn vs. O_2/N_2: Experiments and Modeling

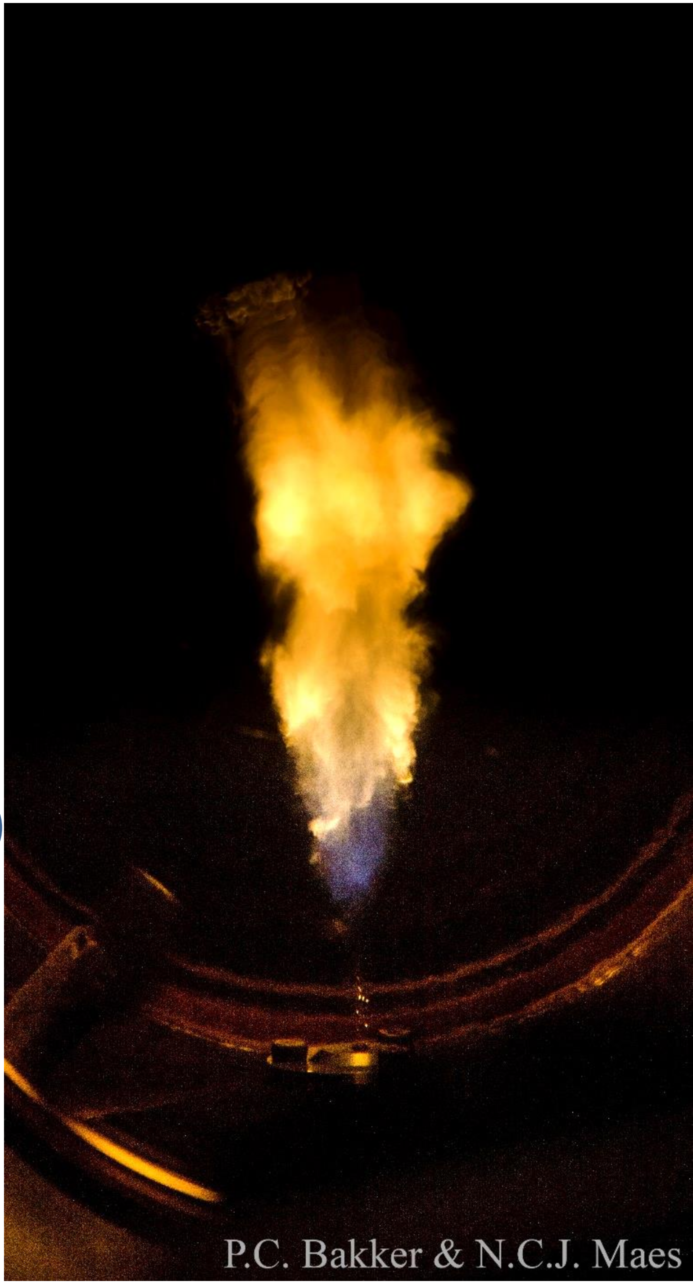
Introduction to ECN5 Experiments

- Previous Soot Efforts
  - ECN1: Nothing
  - ECN2: IFPEN LII
  - ECN3: Soot DBI vs. LII
  - ECN4: Soot DBI and modeling from Argonne, POLIMI, Wisconsin, ETHZ, UNSW…
- List of new experimental measurements
  - Caterpillar: Spray C/D 900K, 950K, 1000K at 22.8 kg/m^3, 15% O_2 (900K, 21% O_2), Constant Pressure Vessel
  - IFPEN: Spray C/D 850K, 900K, 1000K, 1100K at 22.8 kg/m^3, 15% O_2 (900K, 400 bar injection pressure)
  - Sandia: Spray C/D 850K, 900K, 1000K, 1100K at 22.8 kg/m^3, 15% O_2 (900K, 400 bar injection pressure)
  - General Motors Research: Spray A Nozzle 306-04, Constant Pressure Vessel
  - Sandia: Spray A Nozzle 210370, 30+ recent repeats at Spray A baseline

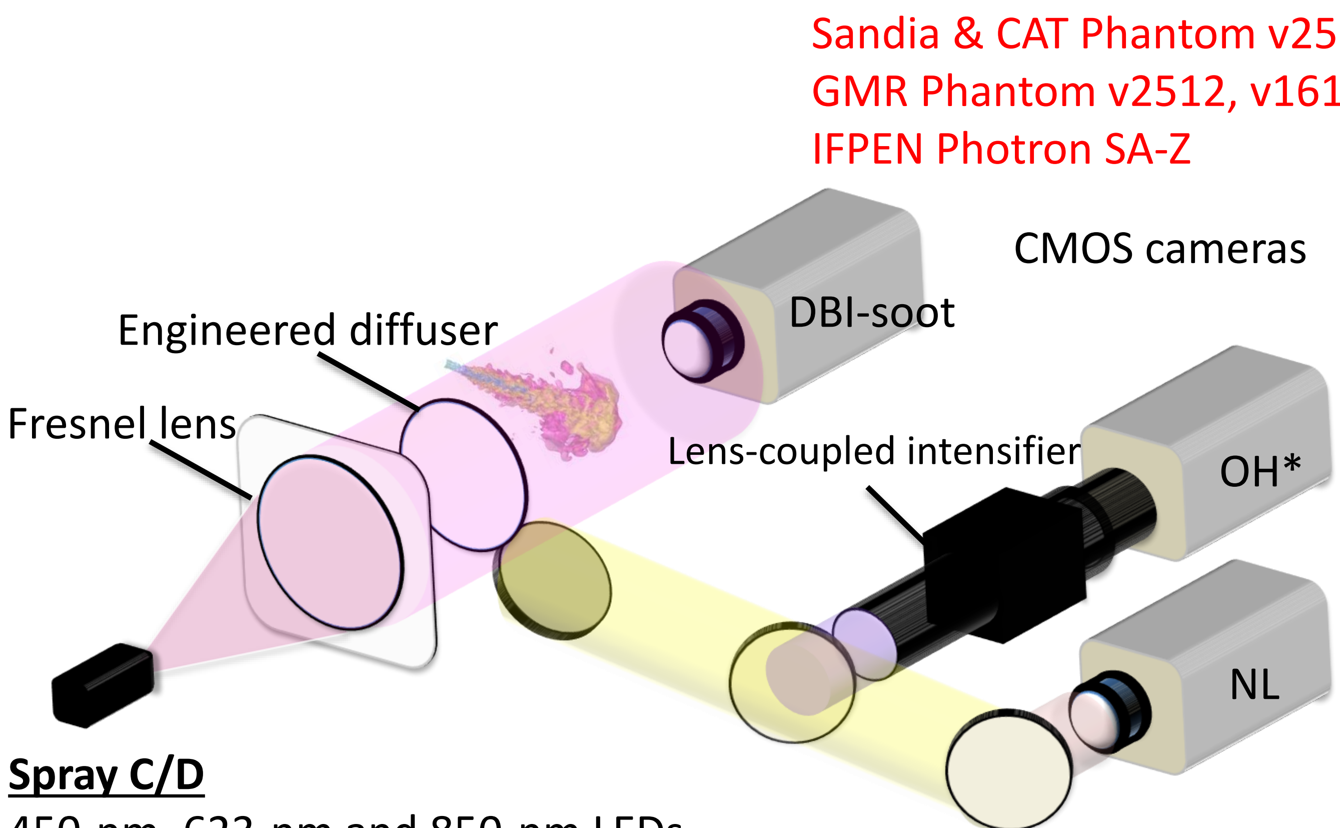
Can we compare them?

- Can we compare the data?
  - Soot is on top of the building blocks
- Different setups
  - CVP vs. CPF & FOV
  - Injector drivers
  - LED vs. laser
  - Different cameras
  - Wavelength influence
- Different post-processing
  - Obtain correct KL
    - Corrections, artifacts
  - Estimate k_e and ρ_{soot}
    - k_e (λ, refractive index, absorption, scattering, morphology, …)
    - At least: same k_e & ρ_{soot}





Diffused back illumination extinction imaging provides time-resolved measurements of soot optical thickness ∝ total soot mass



Spray C/D

450-nm, 623-nm and 850-nm LEDs

810-nm Cavitar laser (IFPEN)

Spray A

406-nm, 450-nm, 519-nm, 623-nm

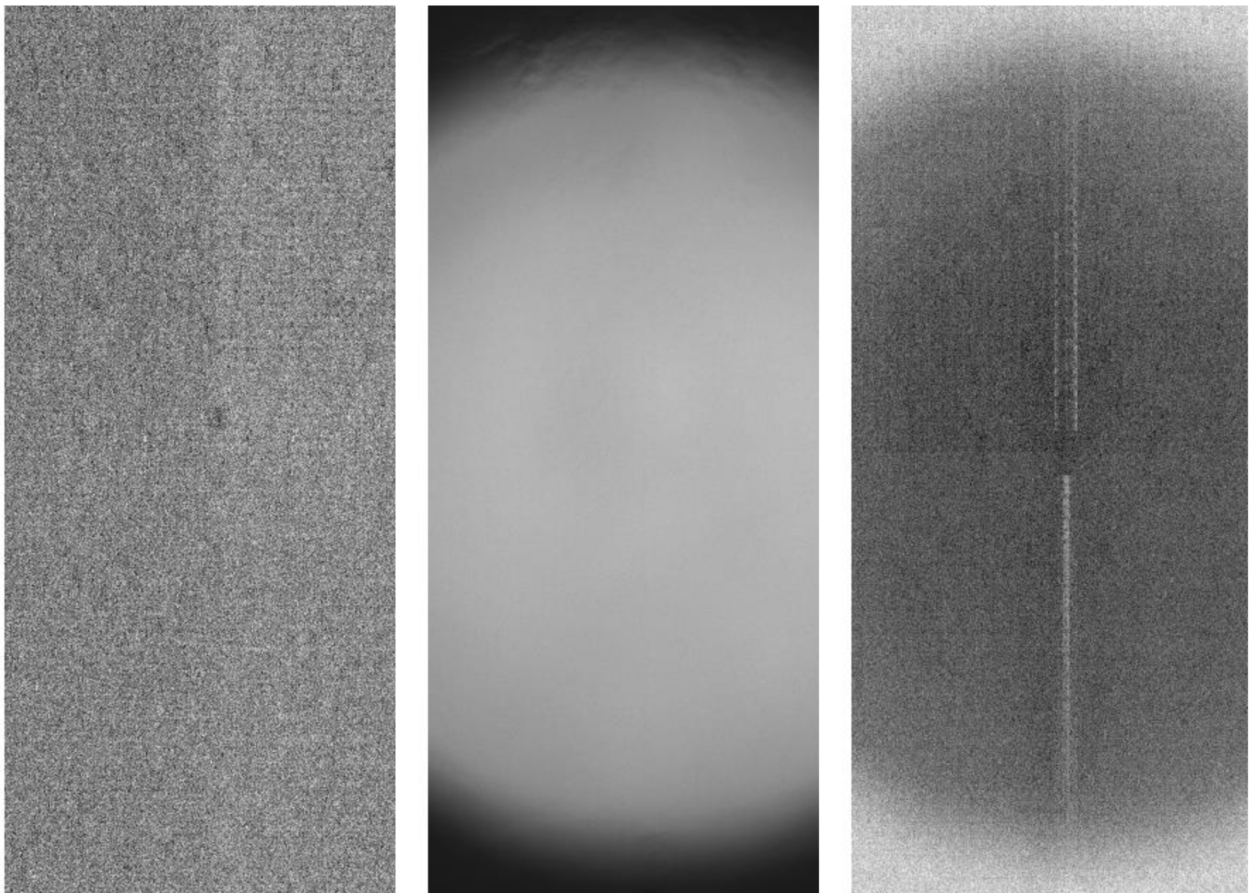
See talk by F. Westlye for more details!

Basic implementation of DBI for soot

- Capture sequence of “zero” light frames
- Capture sequence of light “on/off” frames prior to injection
- Continue with light “on/off” sequence throughout event (may not be necessary for liquid DBI→ higher temporal resolution)
- CMOS camera response to pulsed lighting creates challenge (for some cameras)

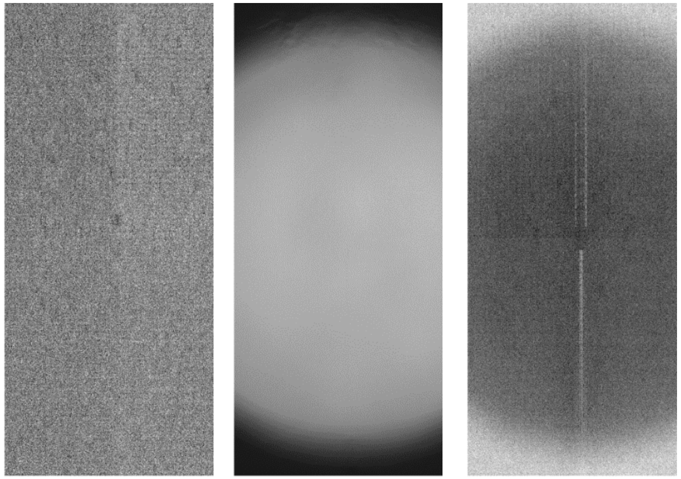
High-speed Phantom CMOS camera negative image lag requires correction

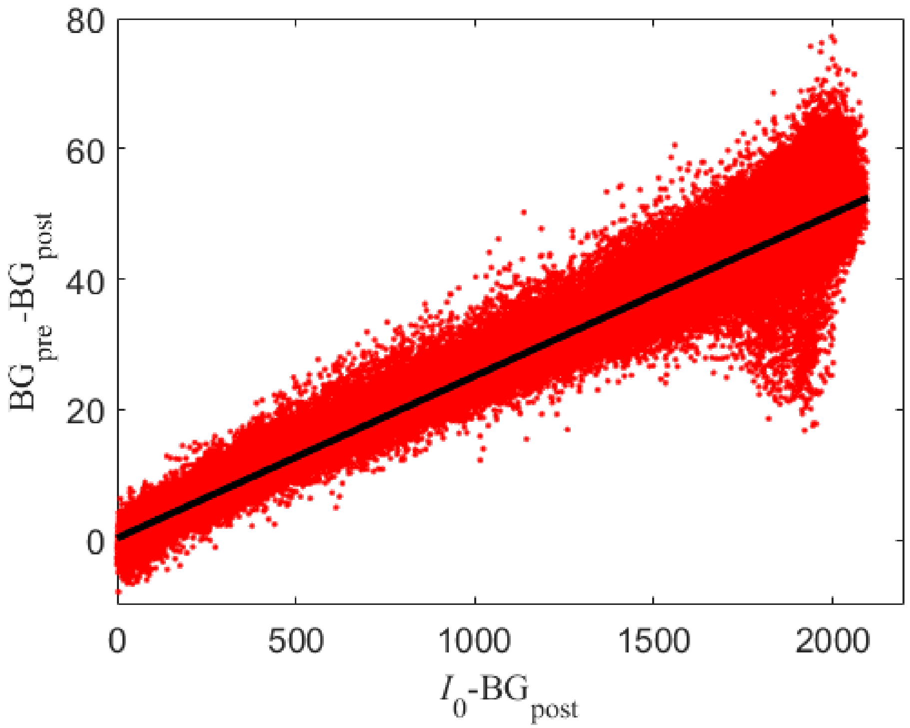
- Pre-LED dark frames acquired and averaged
- LED frames averaged for I_0
- Post-LED dark frames acquire prior to injection



High-speed CMOS camera negative image lag requires correction…even for liquid DBI measurements

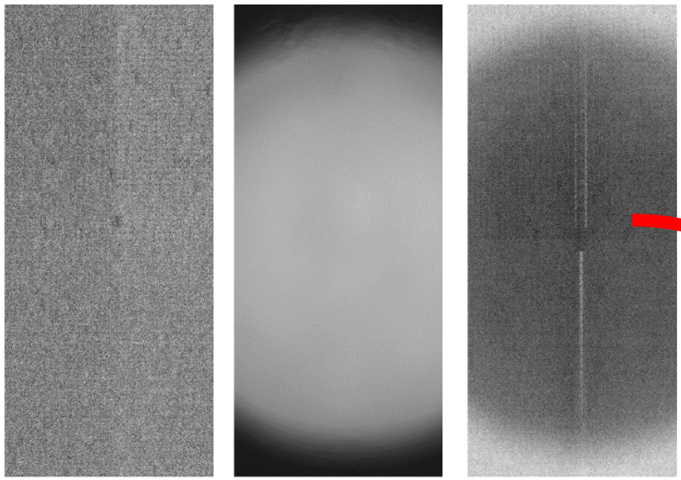
- Linear fit to scatter plot of pixel specific intensity differences generated and used to “correct” dark frames for negative image lag (NIL)

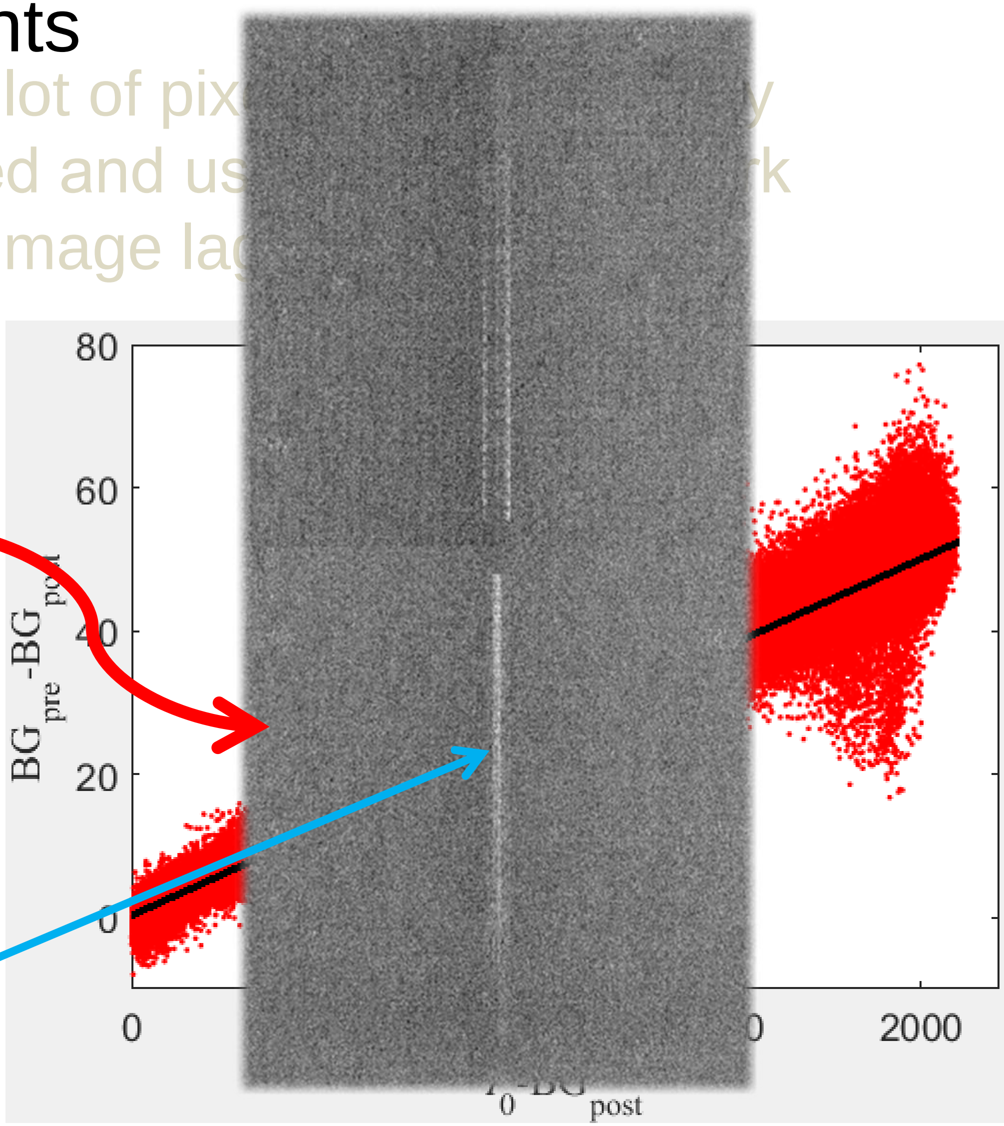




High-speed CMOS camera negative image lag requires correction…even for liquid DBI measurements

- Linear fit to scatter plot of pix
  differences generated and us
  frames for negative image la

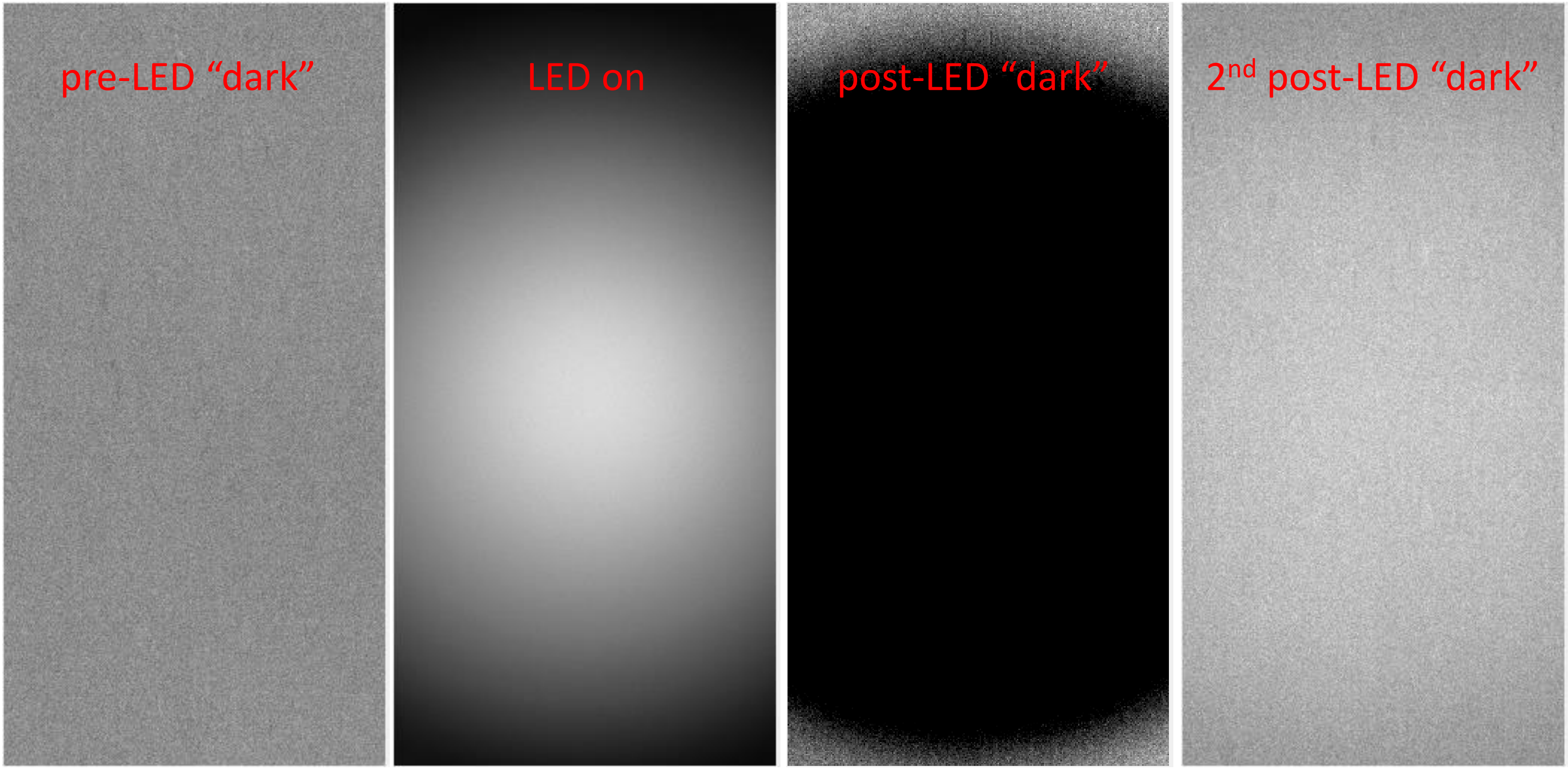




Camera still leaves some fixed pattern artifacts that require additional steps to remove…

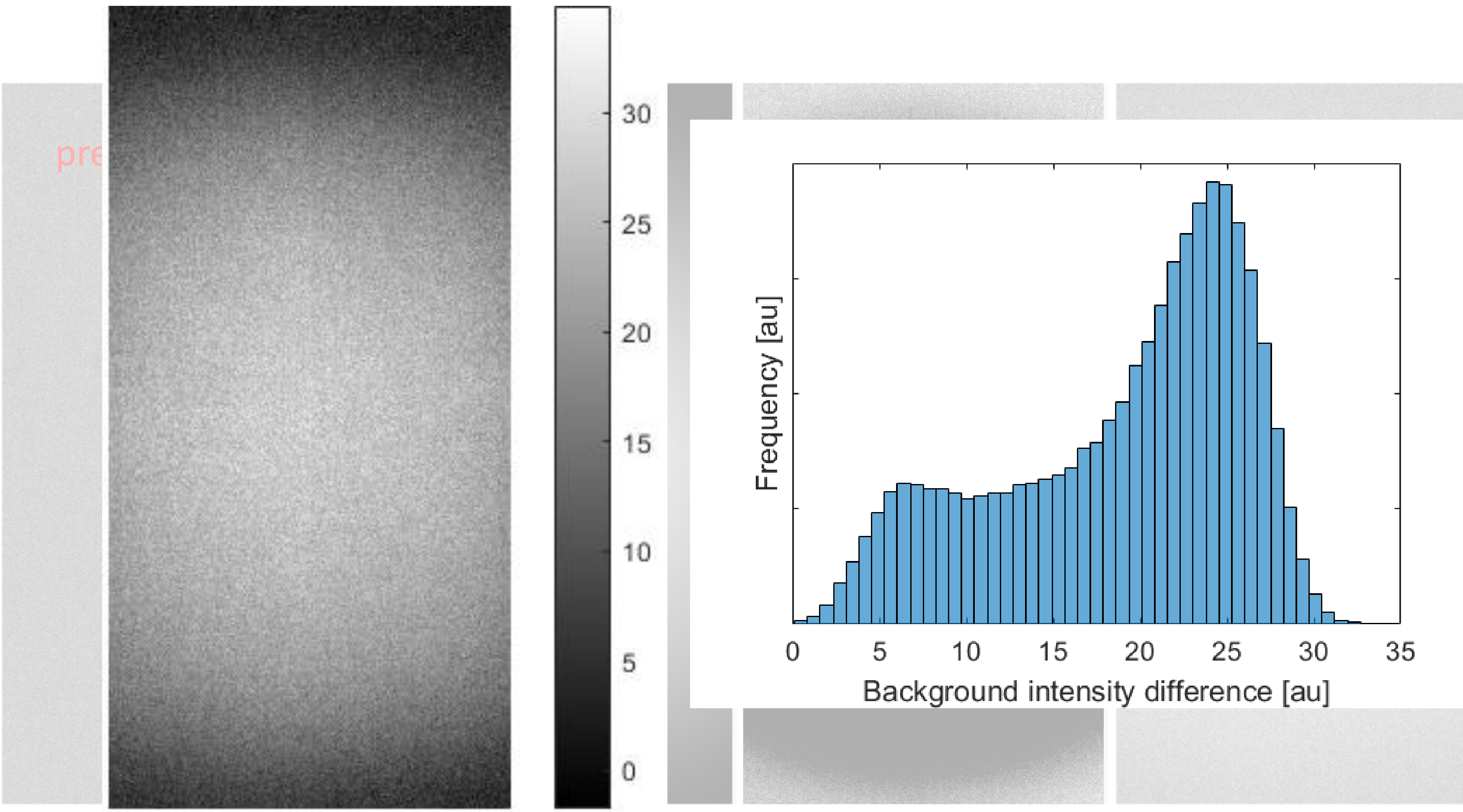
NIL correction tested by skipping two frames

- Data acquired at GMR by Scott Parrish



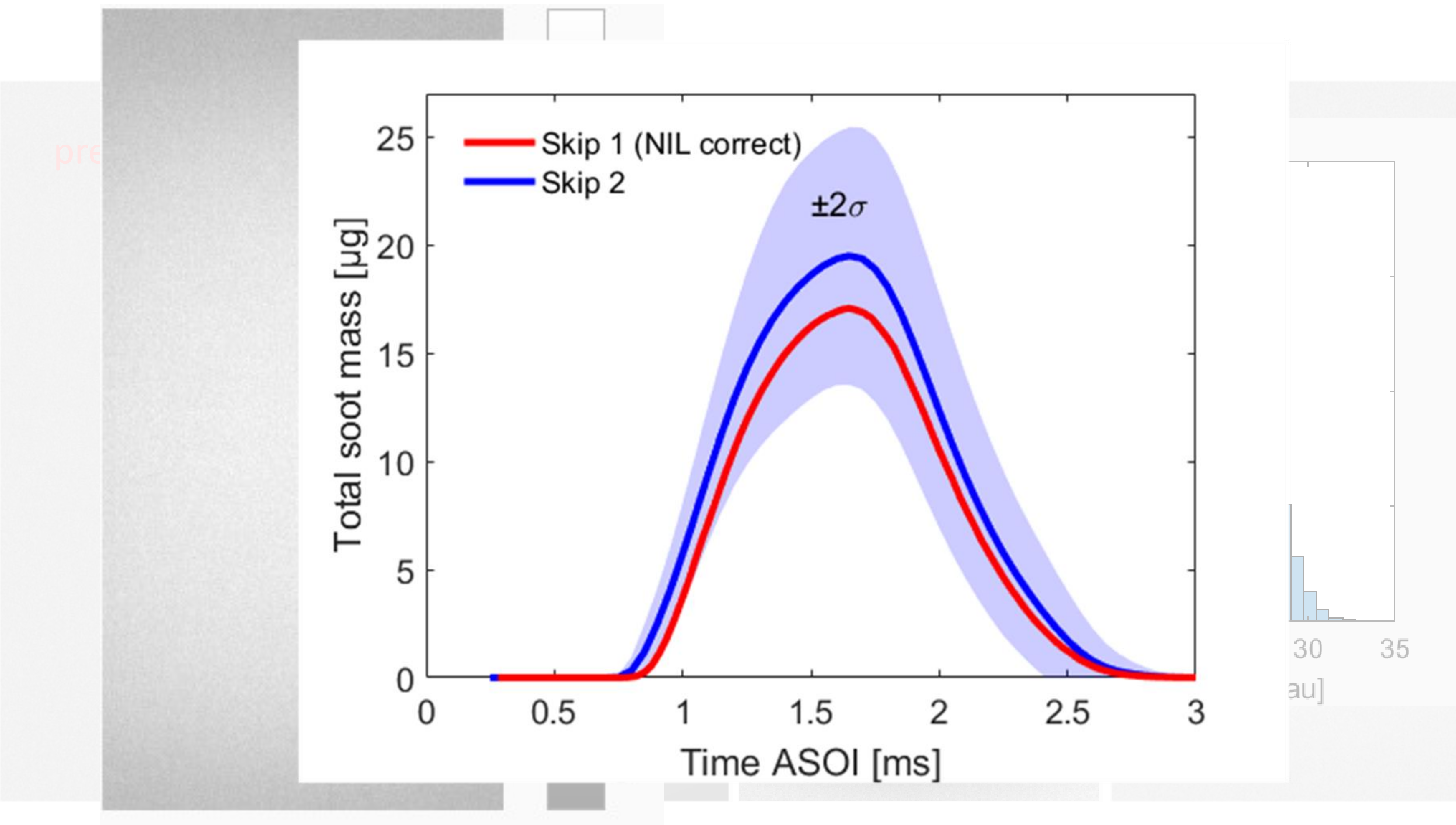
NIL correction tested by skipping two frames

- Data acquired at GMR by Scott Parrish



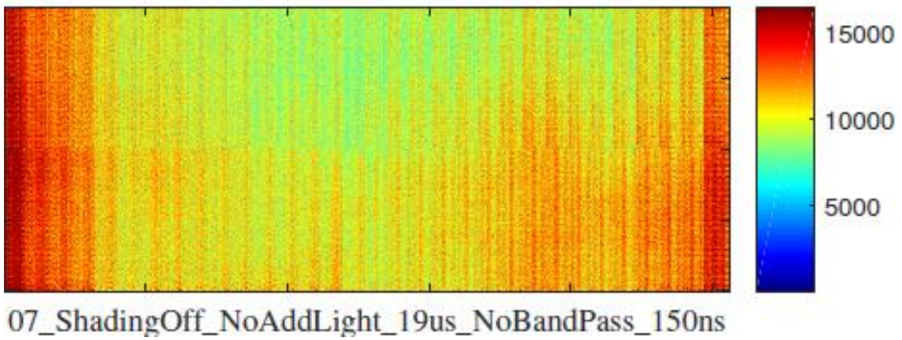
NIL correction tested by skipping two frames

- NIL corrected data are within ***σ***

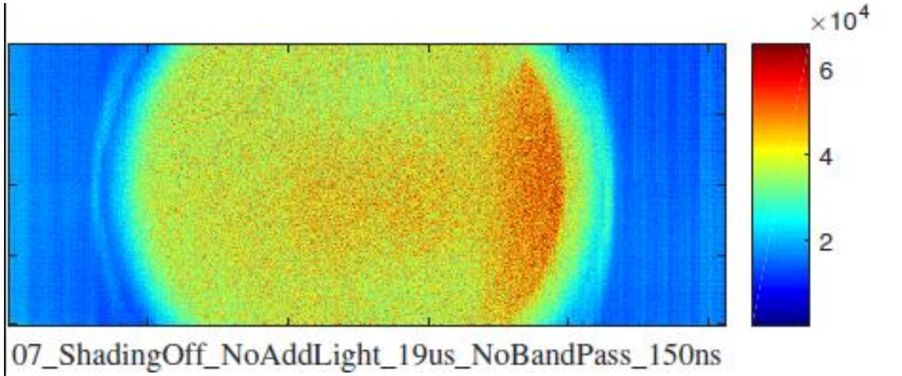


Latest Photron camera used at IFPEN (SA-Z) does not show significant NIL

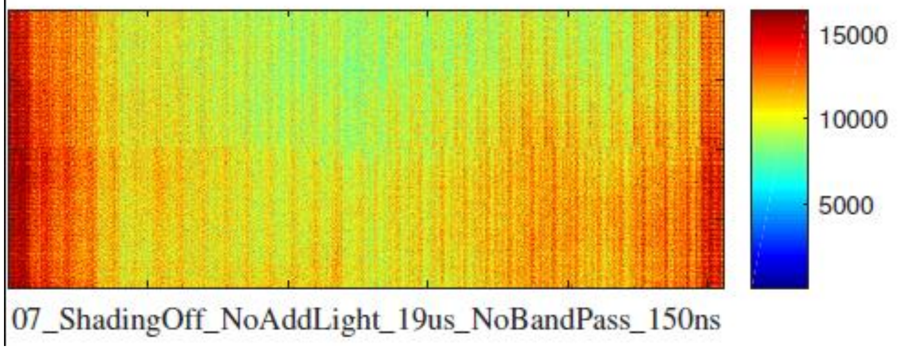
- Single pre-laser image (note: converted to 16 bit)



- Single laser image



- Single post-laser image before injection



SA-Z behavior confirmed with additional lighting to simulate flame emission, beware of changing illumination intensity

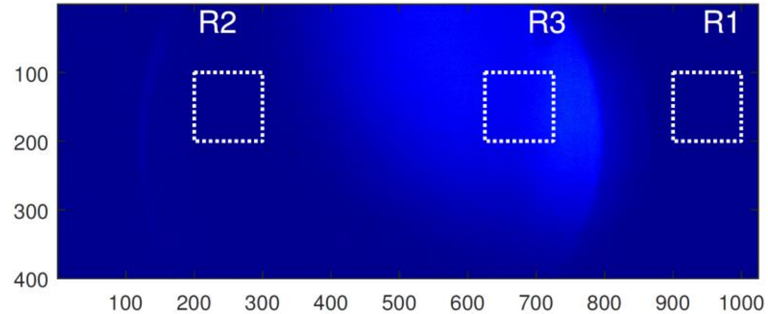
- Repeated experiments without spray

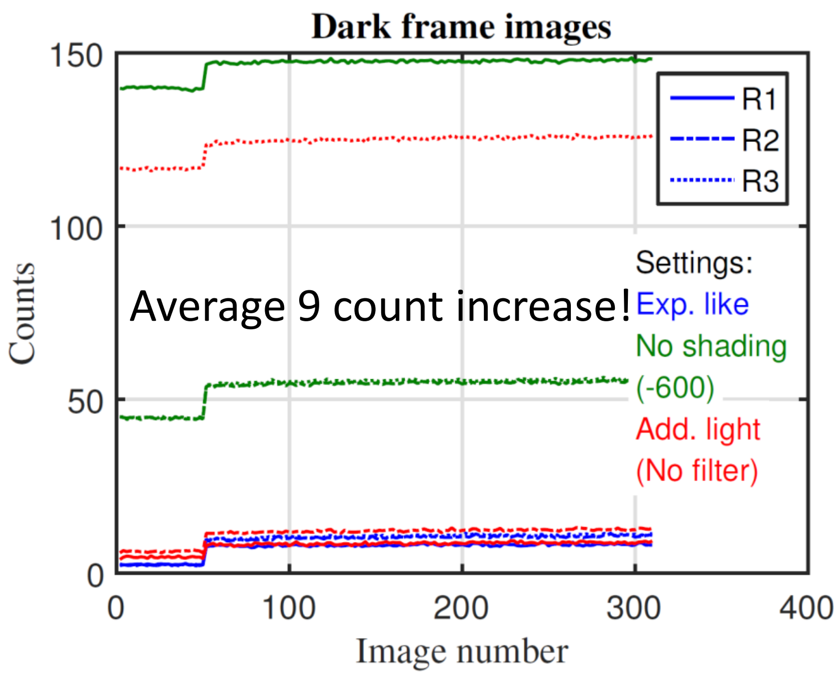
Investigated 3 regions (original 12 bit):

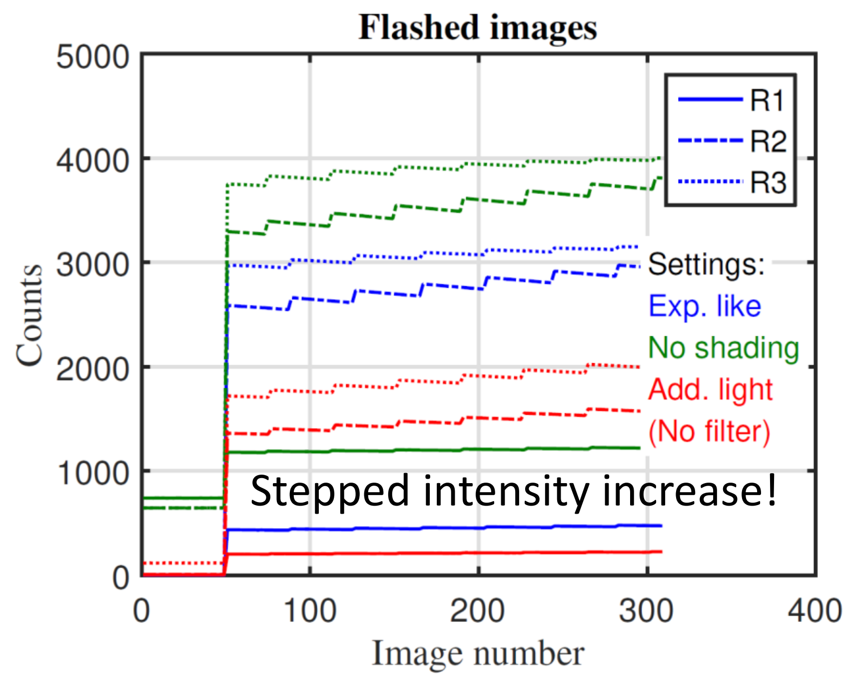
R1 = outside vessel

R2 = extinction imaging

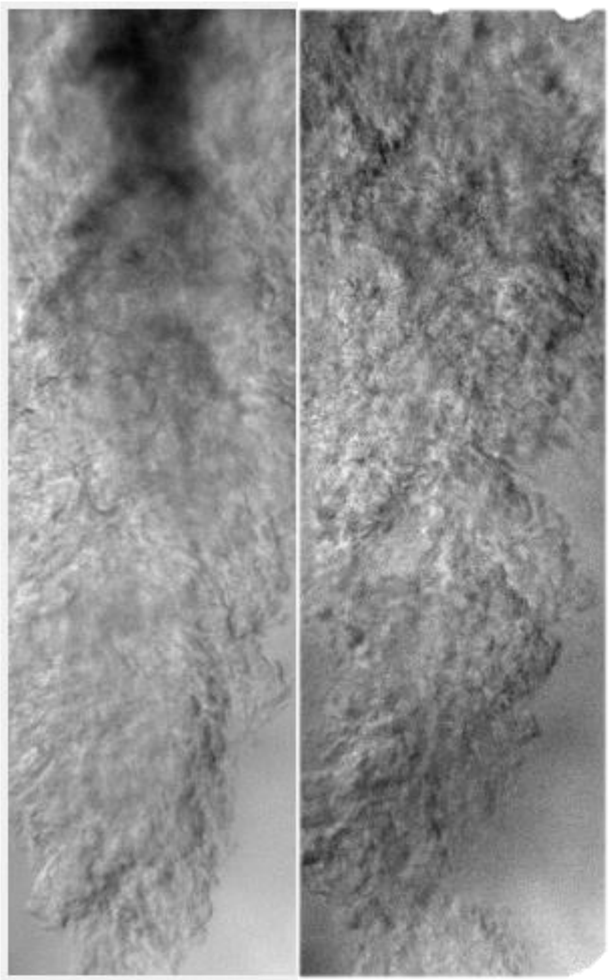
R3 = extinction imaging + "luminosity"

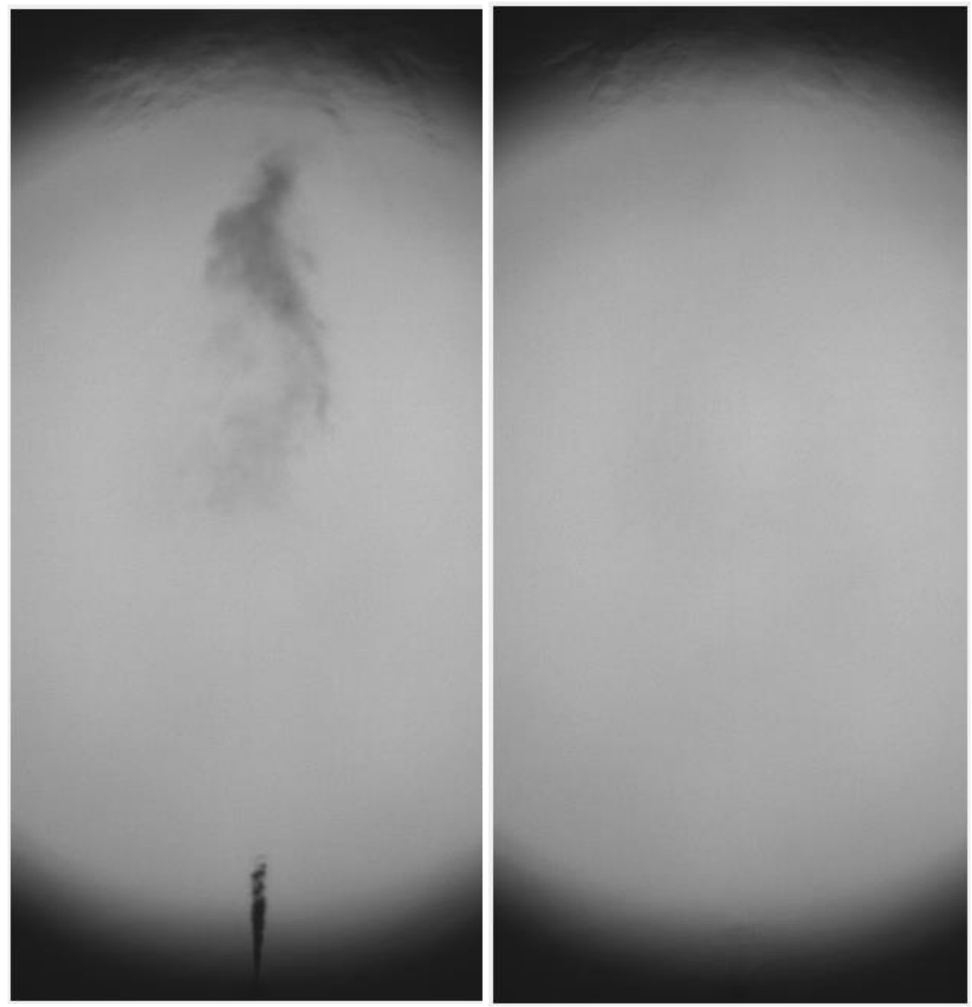






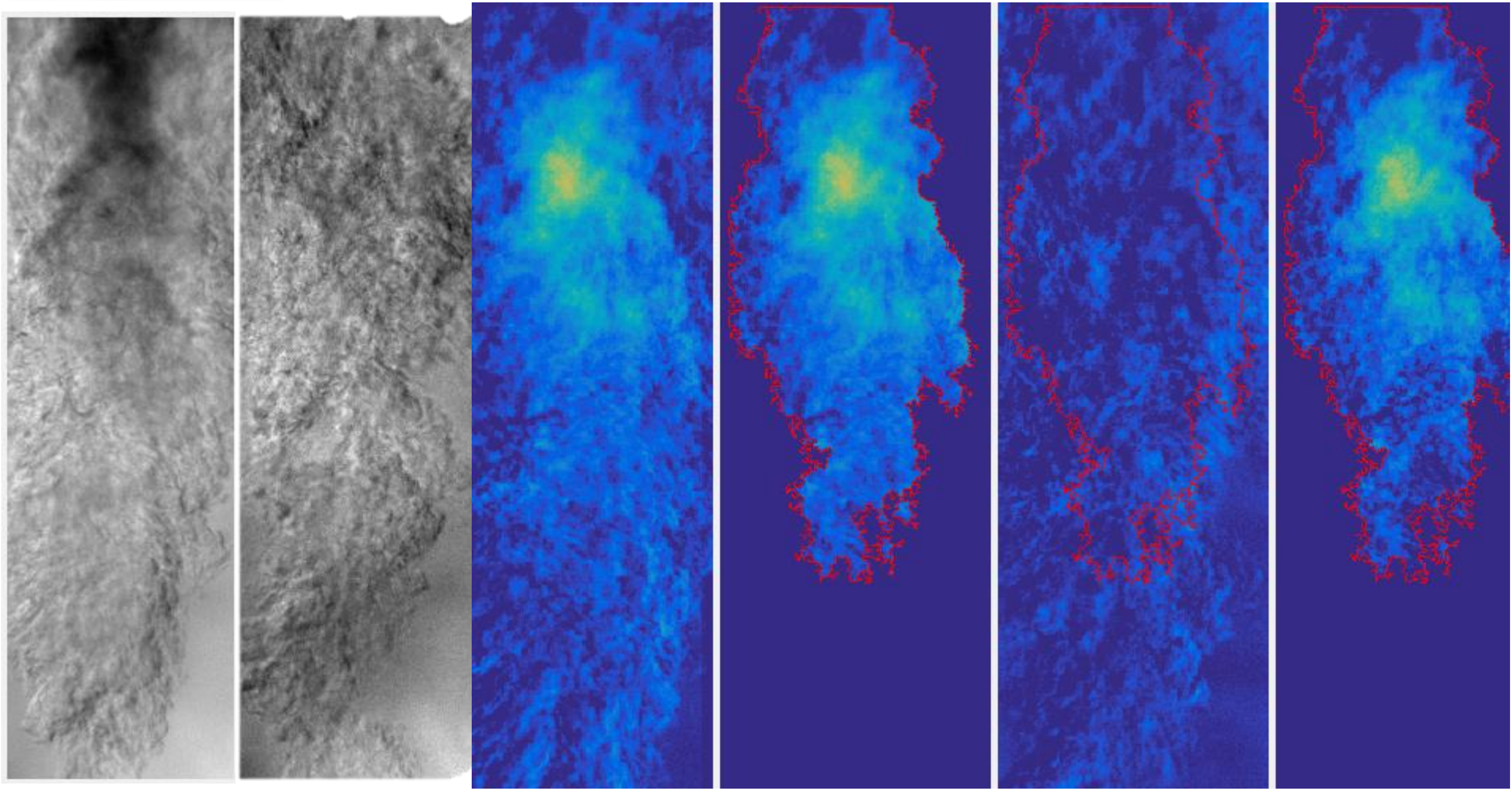
Poorly optimized DBI lighting can yield total soot mass consistent with improved DBI diagnostic





…but many corrections are required during post-processing!

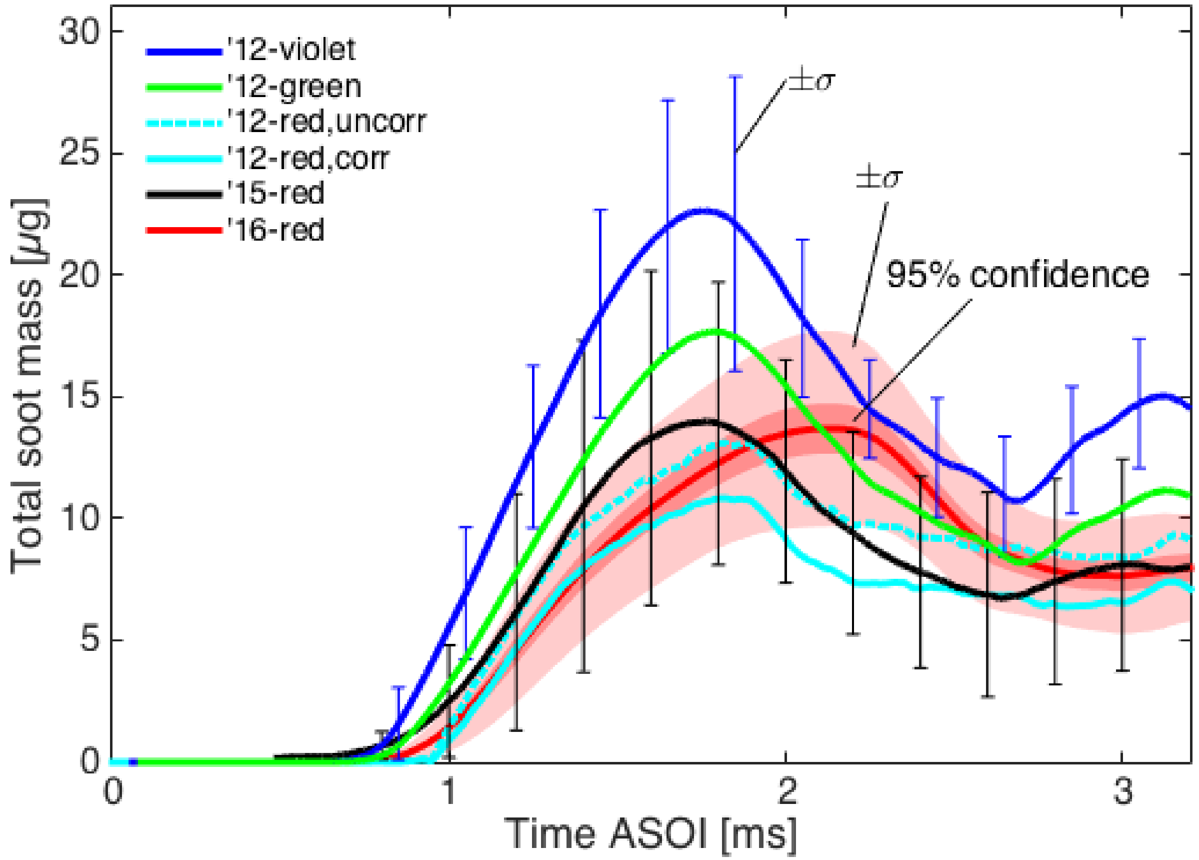
Corrections for bad DBI lighting include masking with lum frame and removal of ref. indx. grad *KL*



Not all soot causing extinction may be hot enough to emit sufficient intensity
Oxidizing soot downstream is hot and bright, but causes little extinction.

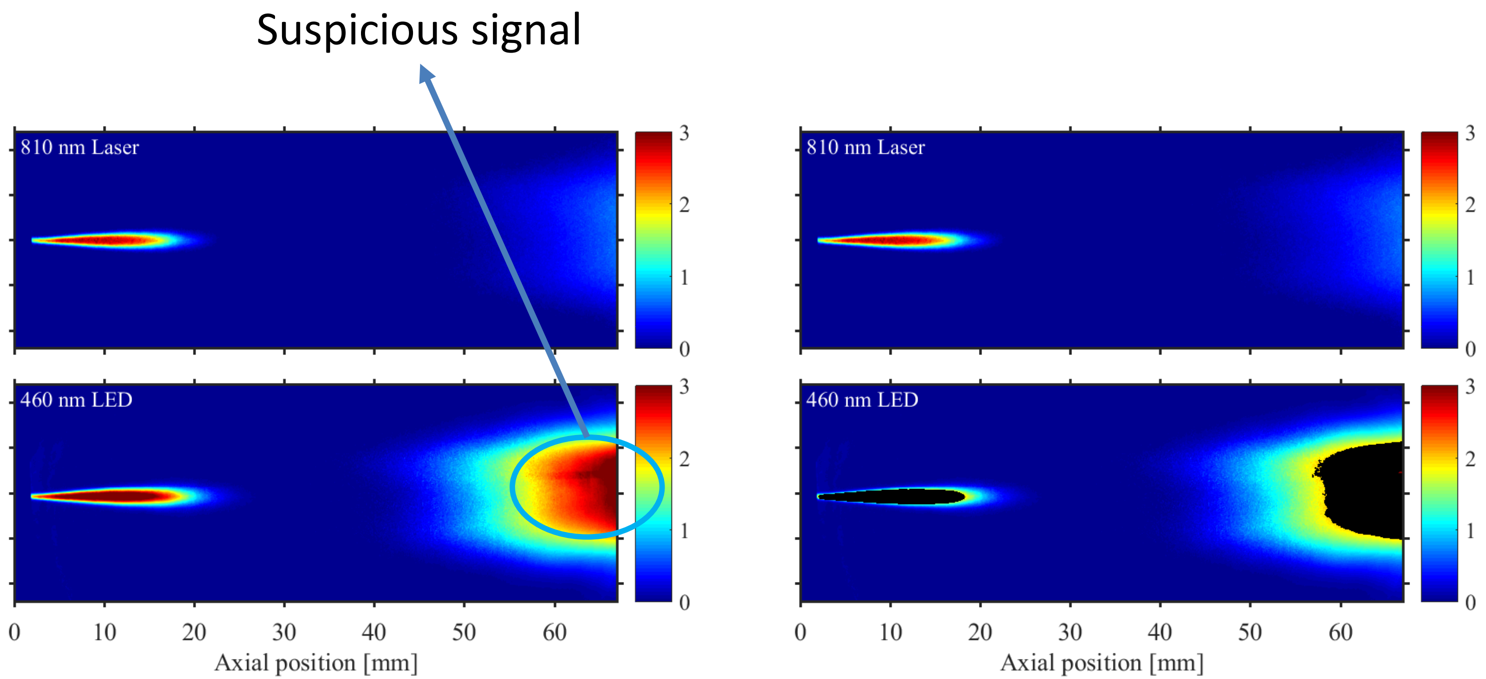
Spray A soot measurements at SNL show wavelength dependencies—red LED measurements seem to be converging

- Large number of recent repeat experiments at SNL (~60) provides excellent 95% confidence interval for the mean but also highlights significant shot-to-shot variability in total soot
- Relatively small variability in ignition delay and lift-off length, yet total soot mass spans 8-20 µg in the head!
- See talk by Koji Yasutomi tomorrow!
- Increased sensitivity for extinction not accounted for by change in k_e from Rayleigh-Debye-Gans Approximation



IFPEN Measurements at 450 nm capture soot farther upstream but reach noise limit of camera in optically thick region

- Saturation: full extinction of light, or “$I\text{-}I_{luminosity}$” below reliability threshold
- Use average of non-full extinct pixels



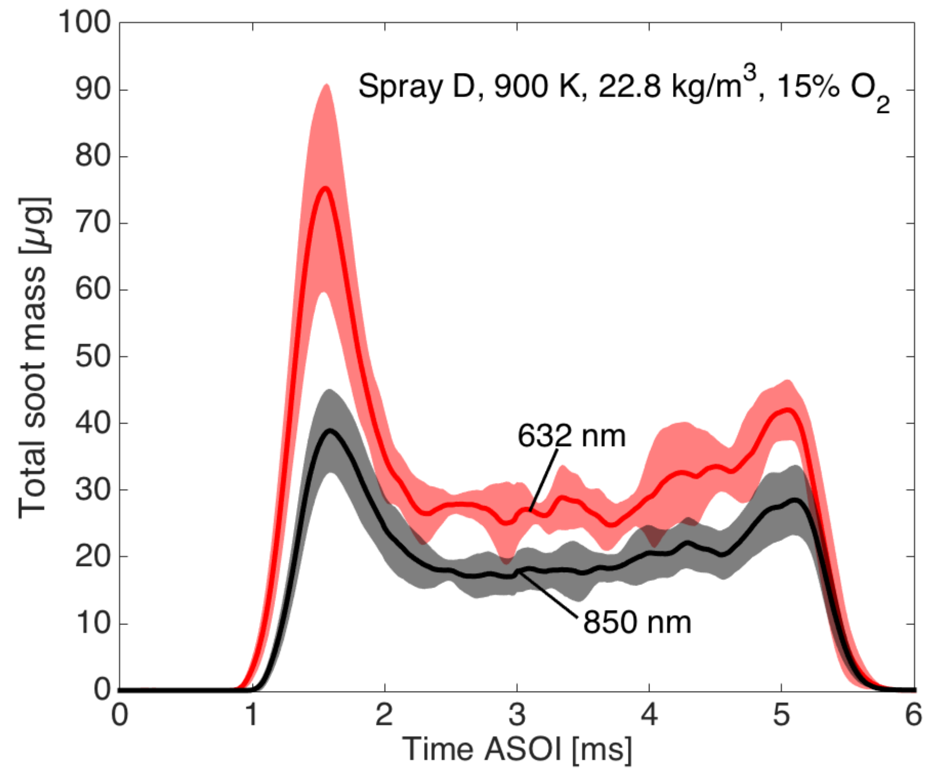
Soot measurements at SNL with 632-nm and 850-nm LEDs highlight wavelength dependence of soot refractive index

Total soot mass (in each pixel of the image) is proportional to optical thickness, *KL*

$$mass_{soot,pixel} = \rho_{soot} \int f_v \, dz \, \delta x \, \delta y = \rho_{soot} KL \frac{\lambda}{k_e} \delta x \, \delta y$$

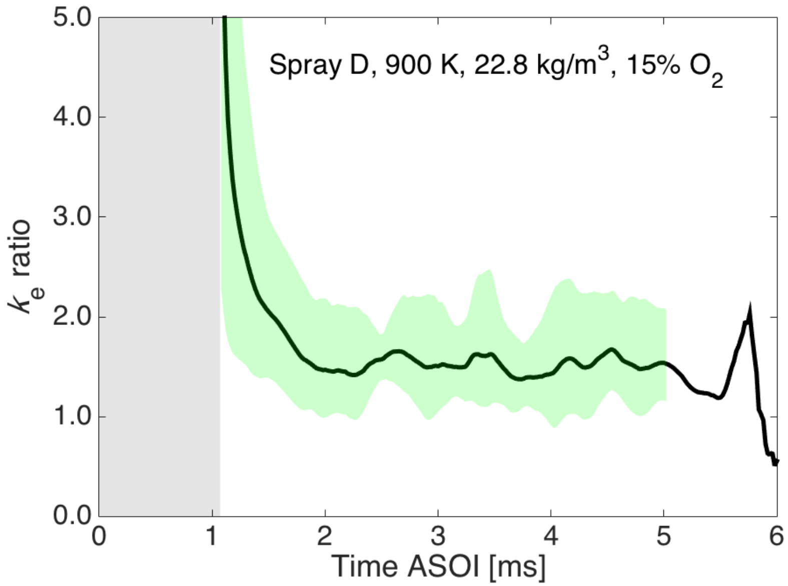
where δx and δy represent the projected pixel size in the x and y axes

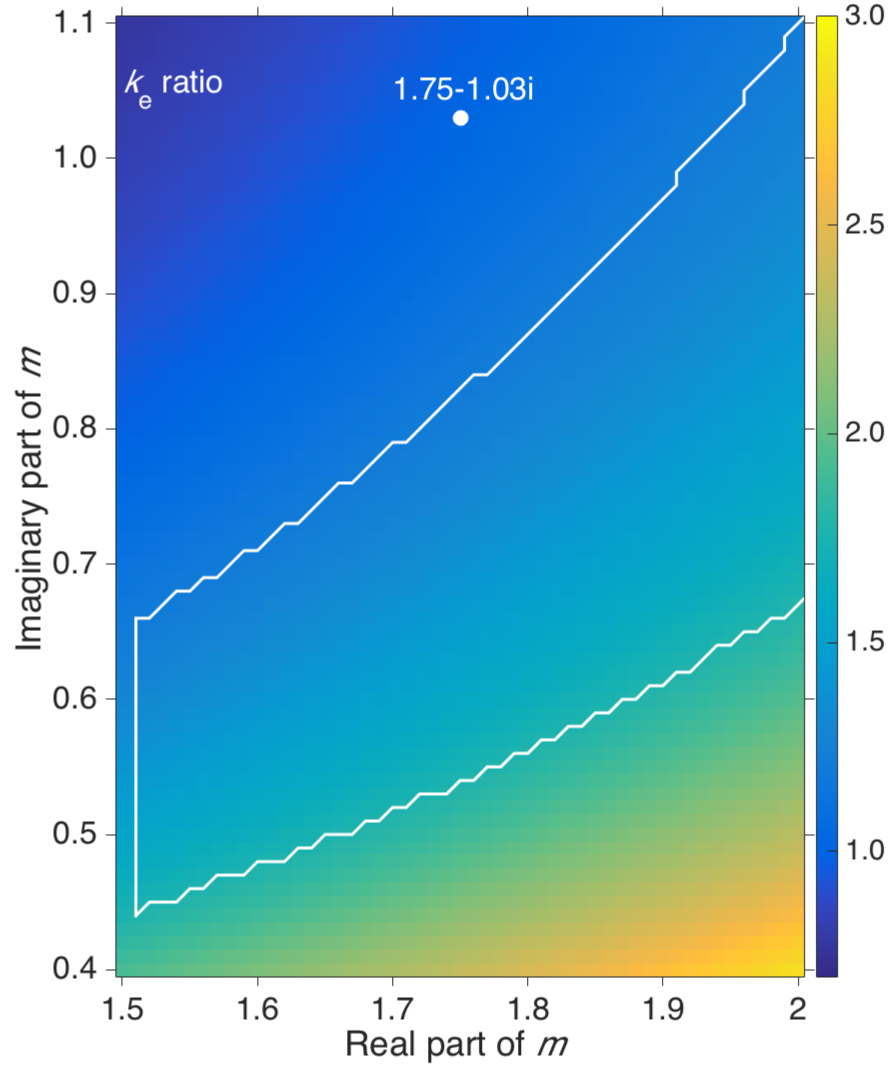
- ρ_{soot} is assumed to be 1.8 g/cm^3
- k_e is the non-dimensional extinction coefficient and is calculated from the Rayleigh-Debye-Gans approximation for fractal aggregates
- k_e depends on soot morphology and the refractive index
- We have shown previously that for sufficiently small particles, k_e is not a strong function of N_p (the # of primary particles per aggregate)
- Williams and Shaddix reported a refractive index of m=1.75 – 1.03i at 632 nm.
- Several authors have reported a modest wavelength dependence over the visible spectrum and into the near IR.



Use of multiple incident wavelengths highlights wavelength dependence in soot refractive index

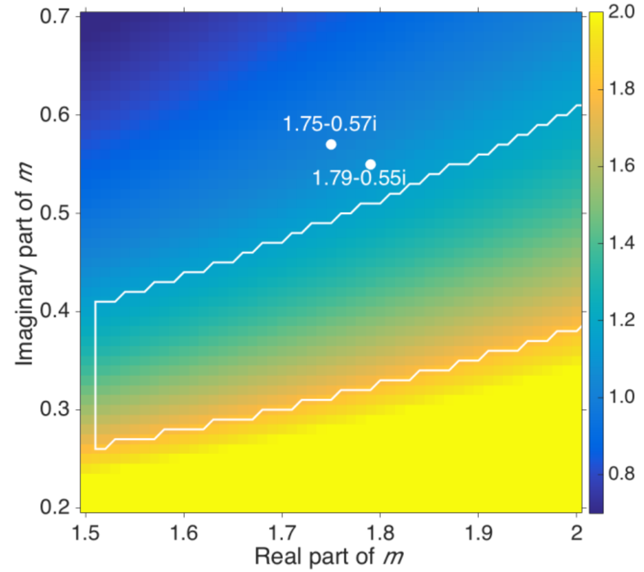
- Early (young or nascent) soot most likely characterized by high H/C ratio.
- Quasi-steady soot k_e ratio ~1.5.
- k_e ratio map based on fixed m = 1.75-1.03i for 632 nm from Williams and Shaddix, m varied for 850 nm.
- Large range of values possible to meet requirements of experiment, imaginary component needs to decrease significantly!



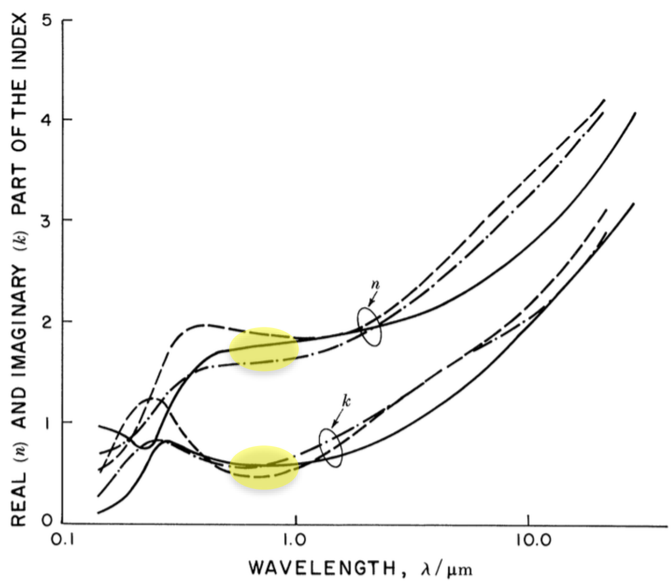


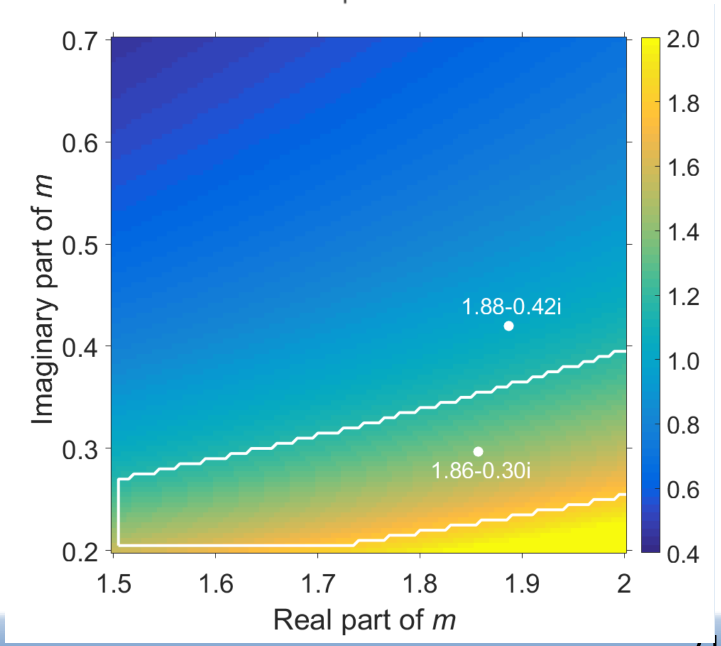
Use of multiple incident wavelengths highlights wavelength dependence in soot refractive index

- Using *m* from Chang and Charalampopoulos *Proc. Roy. Soc. Lond. A*, 1990 (430), *m*=1.75-0.57i better confines region on k_e ratio map within experimental result
- Experiments of Chang and Chara. indicate real component increases with wavelength 632->850 nm and imaginary component decreases.
- Recent work by Bescond et al. [*Journal of Aerosol Science,* vol. 101, pp. 118-132, 2016] and personal comm. from J. Yon led us to 1.88-0.42i at 623 nm and 1.86-0.3i at 850 nm
- Note that Mullins and Williams measured 1.89-0.46*i* and 1.89-0.44*i* at 632 nm for soot generated in toluene and n-heptane wick burner flames, respectively.



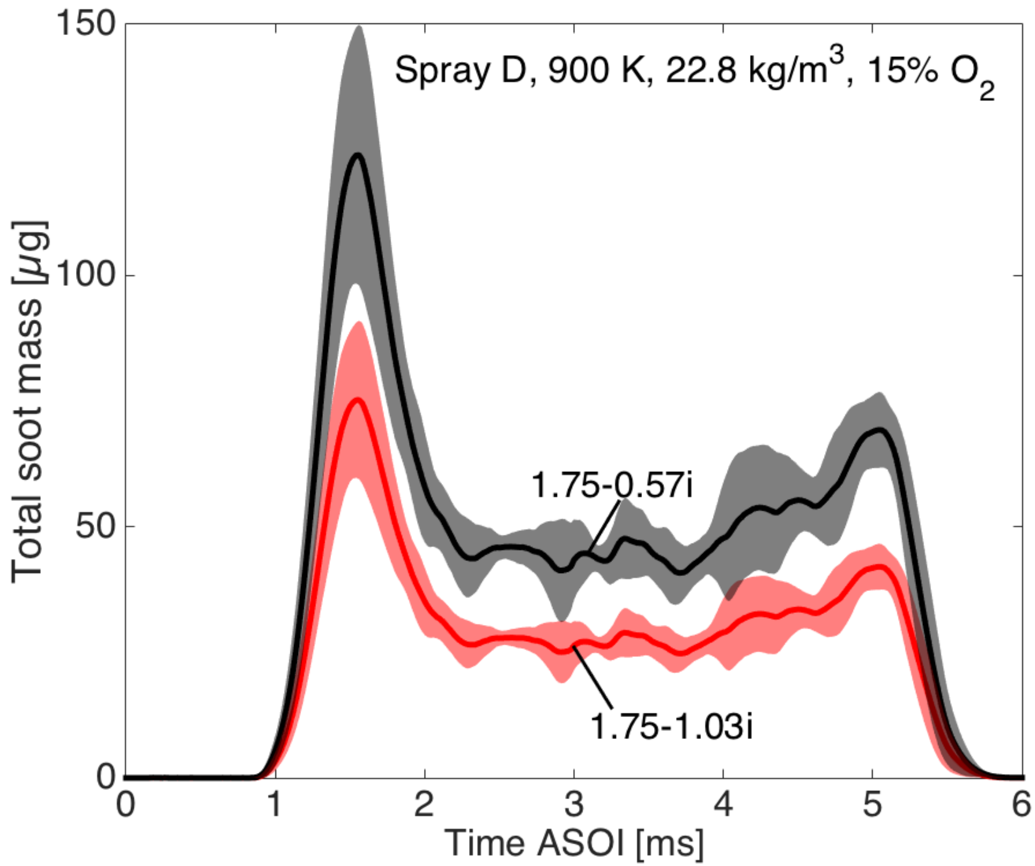
H. Chang and T. T. Charalampopoulos





Uncertainty in soot refractive index is a significant concern for accurate quantitative measurements

- Using m from Chang and Charalampopoulos *Proc. Roy. Soc. Lond. A*, 1990 (430), m=1.75-0.57i to compute k_e increases measured soot mass by 65% relative to m=1.75-1.03i of Williams and Shaddix!



Can we compare the data?

- Mass flow Spray C 1.5% higher, Spray D 3.8% lower for IFP
- Significant differences hydraulic delay amongst institutes: max. 80 µs
- Largely different incident wavelengths: max. 390 nm
- Peculiar differences ID & LOL (LOL_D really similar, ~100 µs ID diff.!)

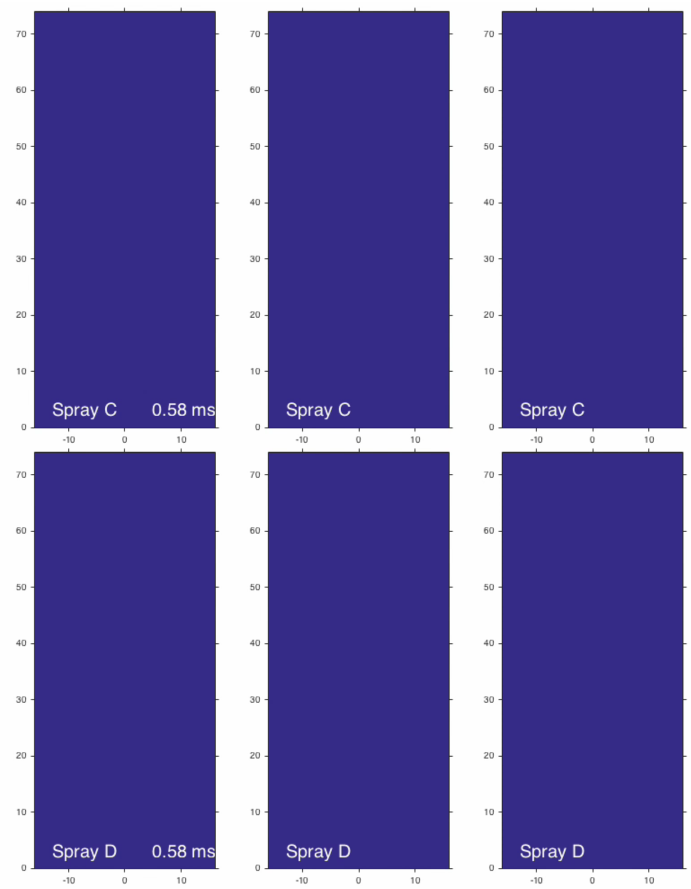
	SNL 850	SNL 623	CAT 623	IFP 810	IFP 460
Injectors	C037 & D134	D134	C037 & D134	C003 & D135	C003
Mass flow[1] [g/s]	10.10 & 11.95	11.95	10.10 & 11.95	10.26 & 11.49	10.26
Injector driver	Genotec	Genotec	Labview driven	EFS IPoD	EFS IPoD
Hydr. Delay* [s]	361 & 380	380	400	440 & 440	440 & 440
Incident [nm]	850 LED	623 LED	623 LED	810 Laser	460 LED
Camera	Phantom V2512	Phantom V2512	Phantom V2512	Photron SAZ	Photron SAZ
Filtering	ND + broad BP	ND + broad BP	ND + broad BP	ND + narrow BP	ND + broad BP
Max FOV [mm]	70	70	85	67	67
ID (900K) [us] CI 95% [us]	550 & 560 26 & 28		554 & 576 51 & 46	460 & 479 28 & 31	
LOL (900K) [mm]	24.7 & 27.2		24.1 & 26.0	21.7 & 26.8	

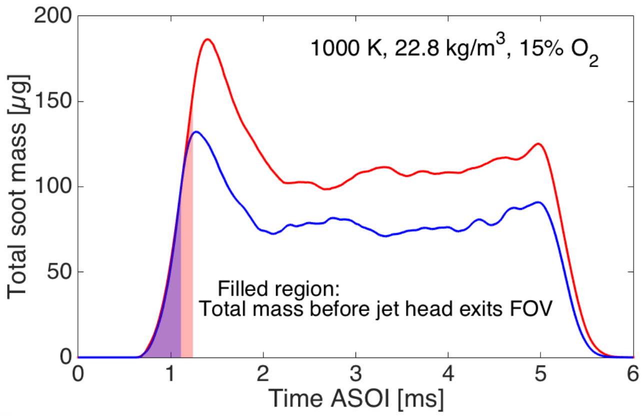
1: Fuel 180 (2016) 357–366 *: Same for all injectors according to ref. 1

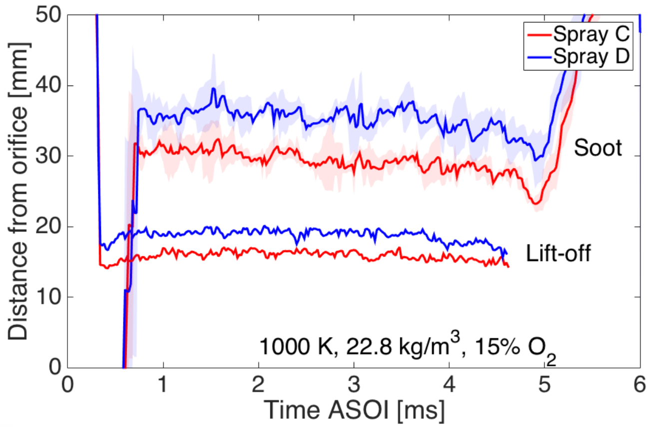
Spray C & D comparison: total soot mass

- FOV challenge (SNL-CAT-IFP)
- Missing most Soot!

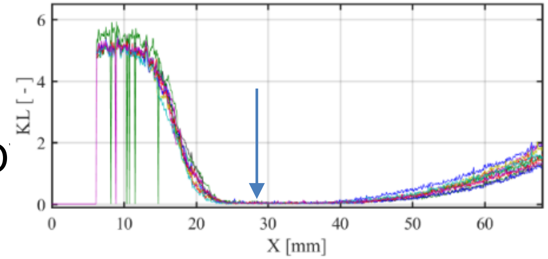
- Use onset point (C vs. D)
- Set max. window (SNL-CAT-IFP)







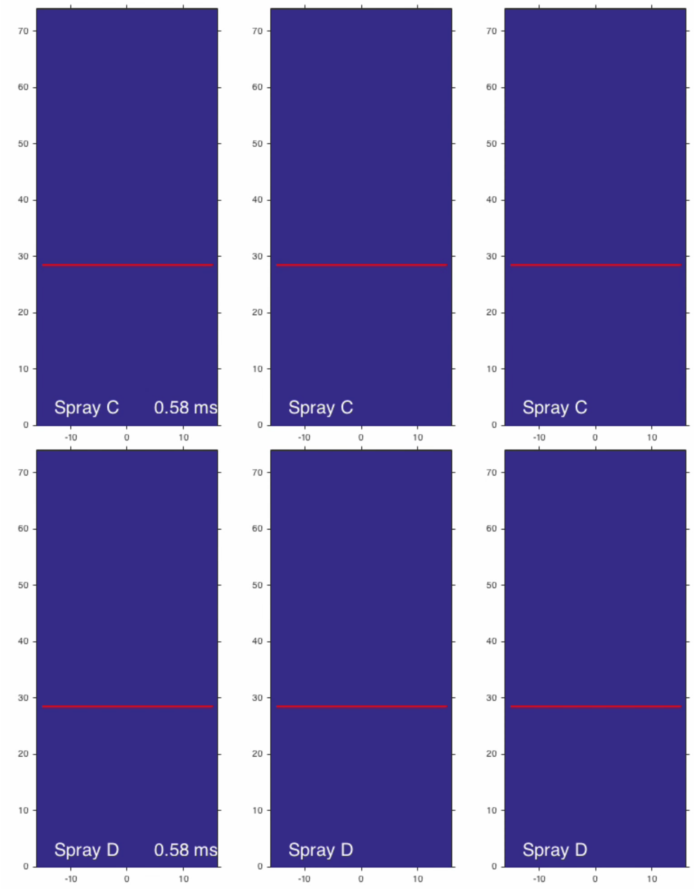
Spray C & D comparison: total soot mass



- Prevent liquid fuel in FO
- Onset point KL = 0.05
  - IFP: minimum between liquid & soot
  - SNL/CAT: luminosity as a mask + boundary funct

- Track soot for 30 mm
  - Up until common FOV!
  - Note: 30 mm not achieved at low T!

- What about the differences in mass-flow? (between Spray C & D, that is)
- What about the sooting area?



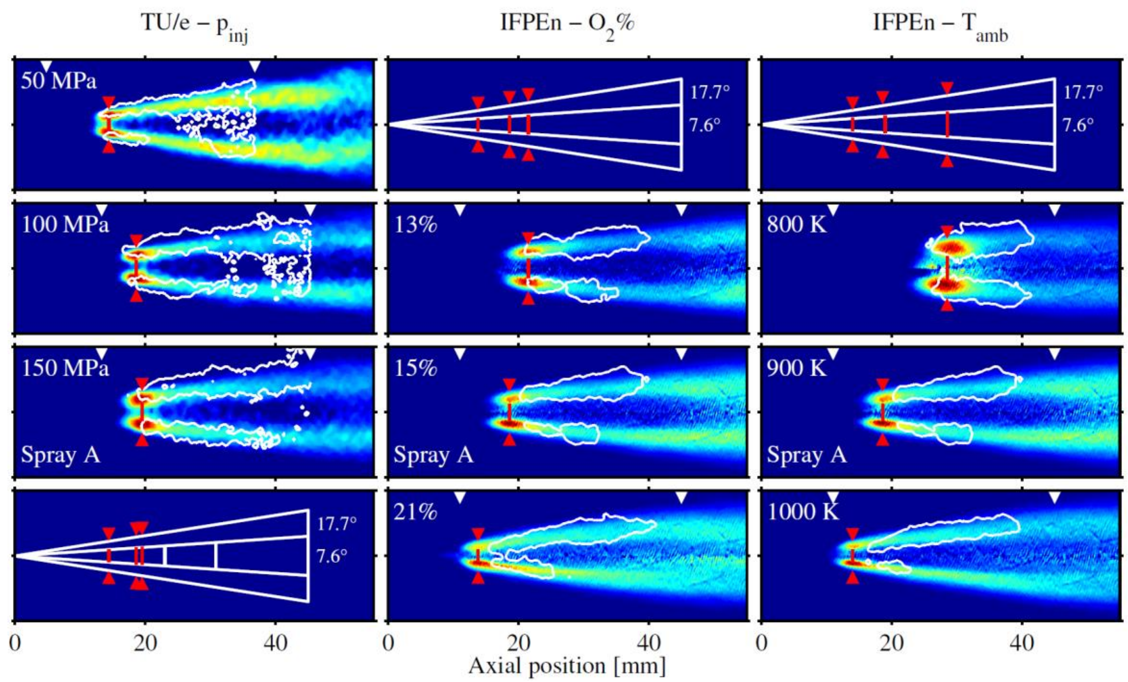
Spray C & D comparison: total soot mass

- Differences in mass-flow
  - For C003: 13% decrease compared to D135 – Take into account?

- Sooting area
  - Similar to flame structures:
    “moving upstream” →
    “confines sooting area” →
    “may influence total soot mass”
  - Limit area (instead of FOV)?

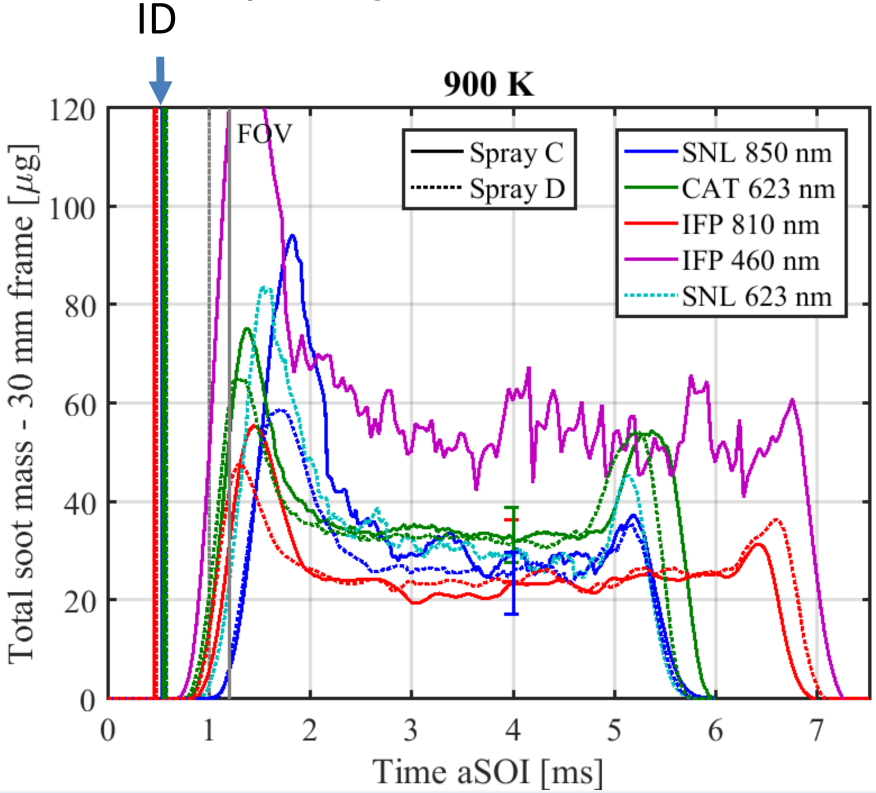
Upstream soot inception

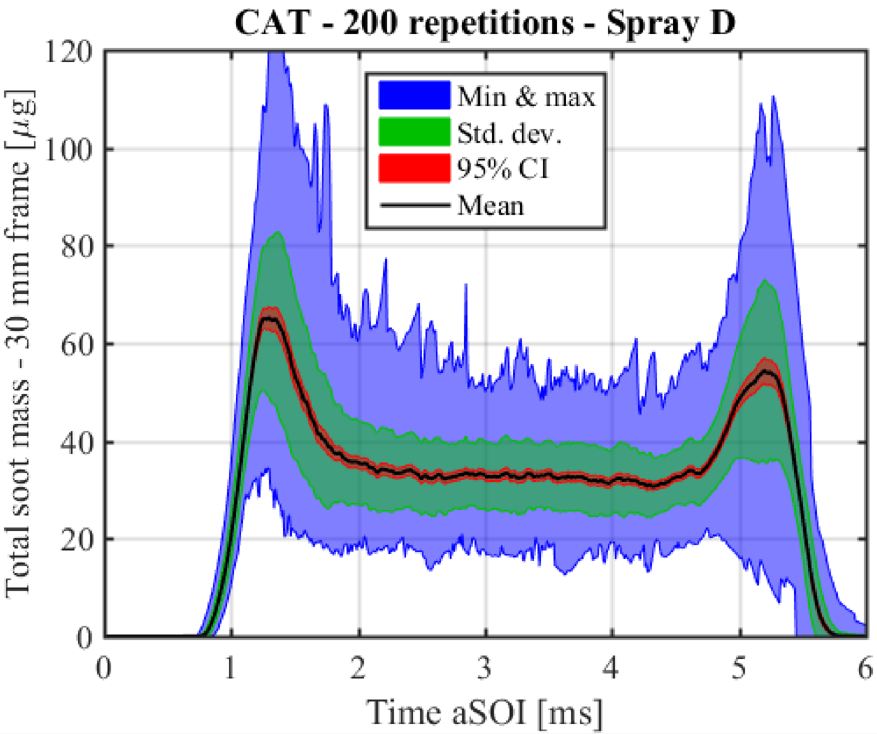
Downstream soot inception



Spray C & D comparison: total soot mass

- More soot in Spray C onset peak
- D onset before C → delayed peak in C
- QS: C&D very similar and within std dev between institutes!
- 30 mm not achieved for long-wavelengths in CVP → reduced mass!
- Capturing soot recession in IFP data due to post-processing method?

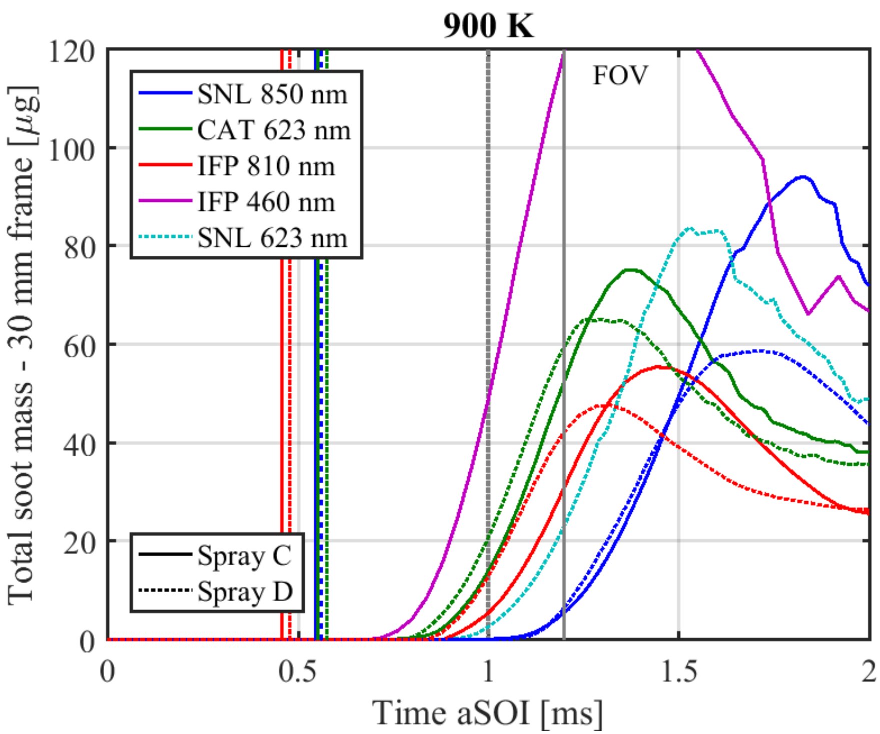


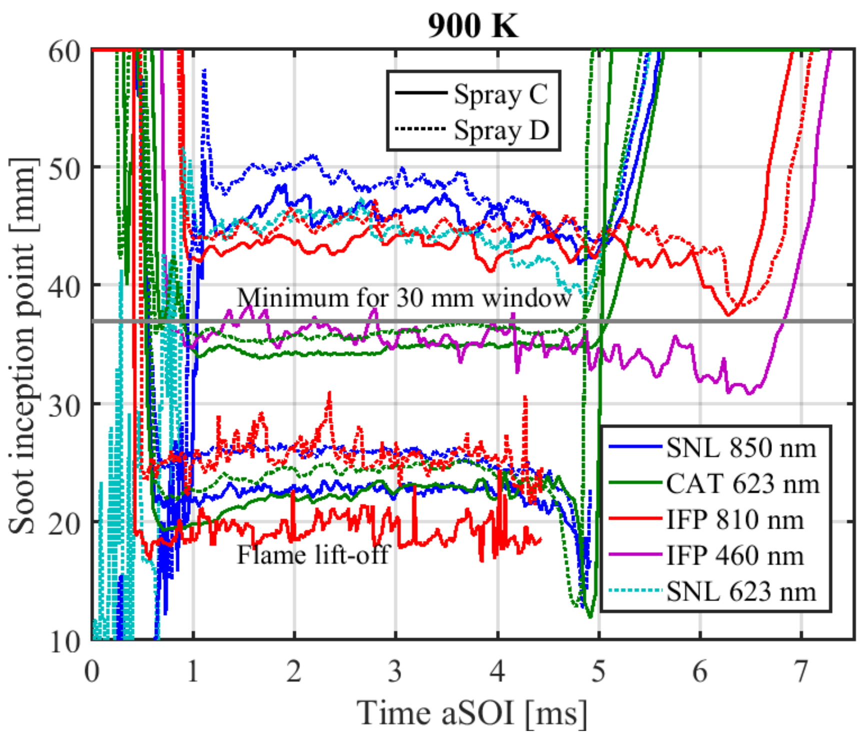


Spray C & D comparison: total soot mass

- Δt_{onset} IFP (Δ350 nm) ≈ Δt_{onset} SNL (Δ227 nm) – but note temporal limit!

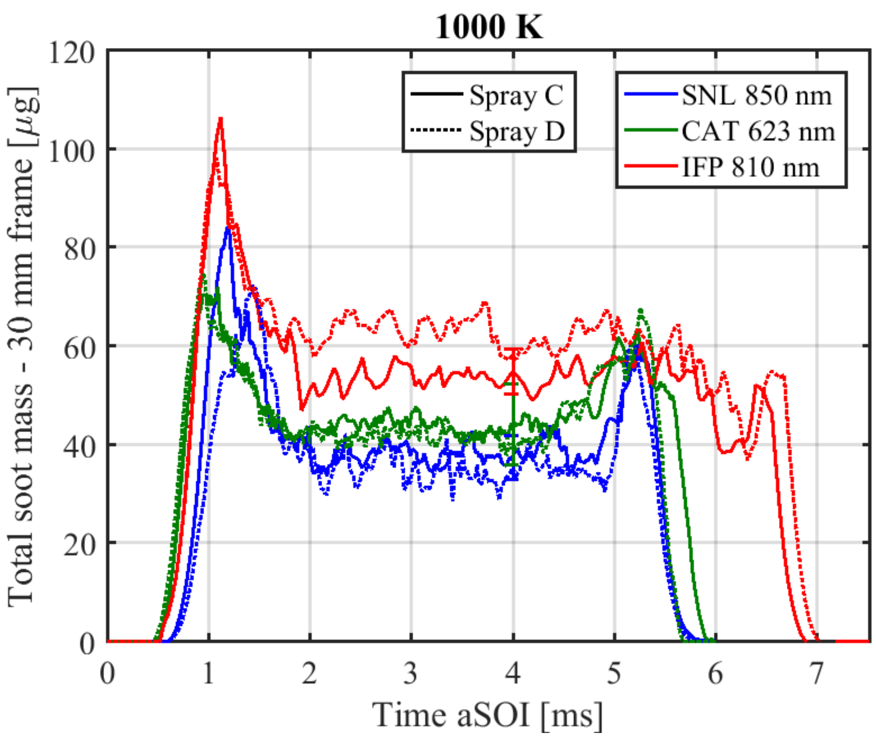
- Relatively early onset CAT + ↑ QS mass → lack of CO_2 & H_2O + FOV?
- Early onset IFP linked to ID?
- Far in the QS phase - SNL D & IFP D are similar in soot and LOL: Clearly, there is something we do not understand yet…

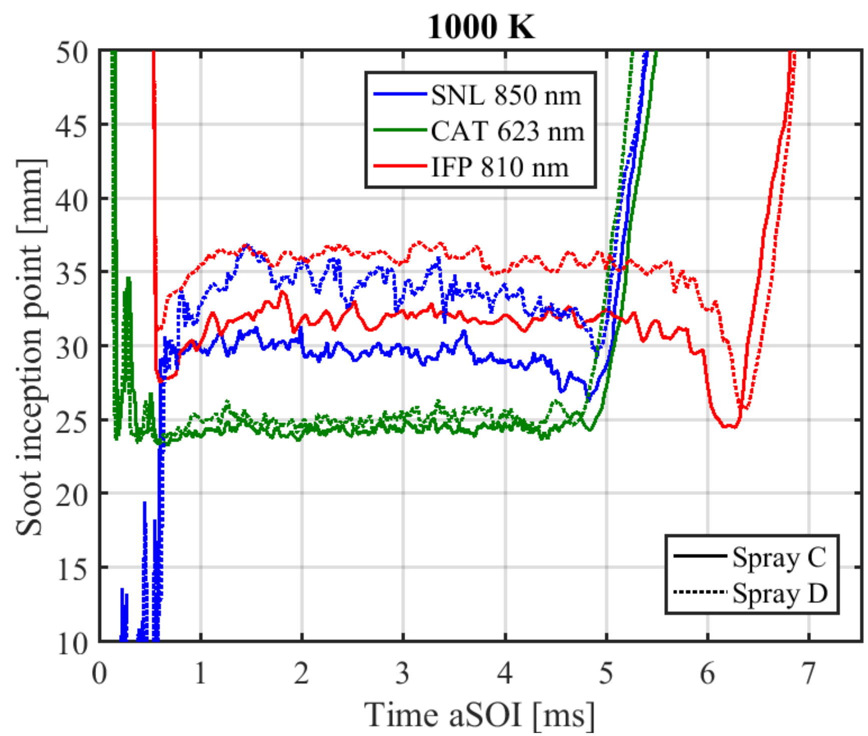




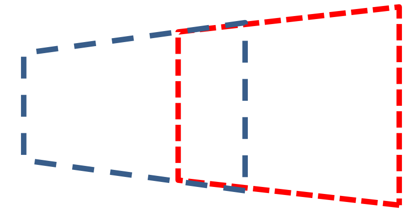
Spray C & D comparison: total soot mass

- 1000K (full FOV!): C & D peaks closer to one-another
- IFP > CAT & SNL (reversed at 900 K!) - inception point more similar!
  IFP further downstream → indicates increased production rate!
- Soot recession again only present for IFP data

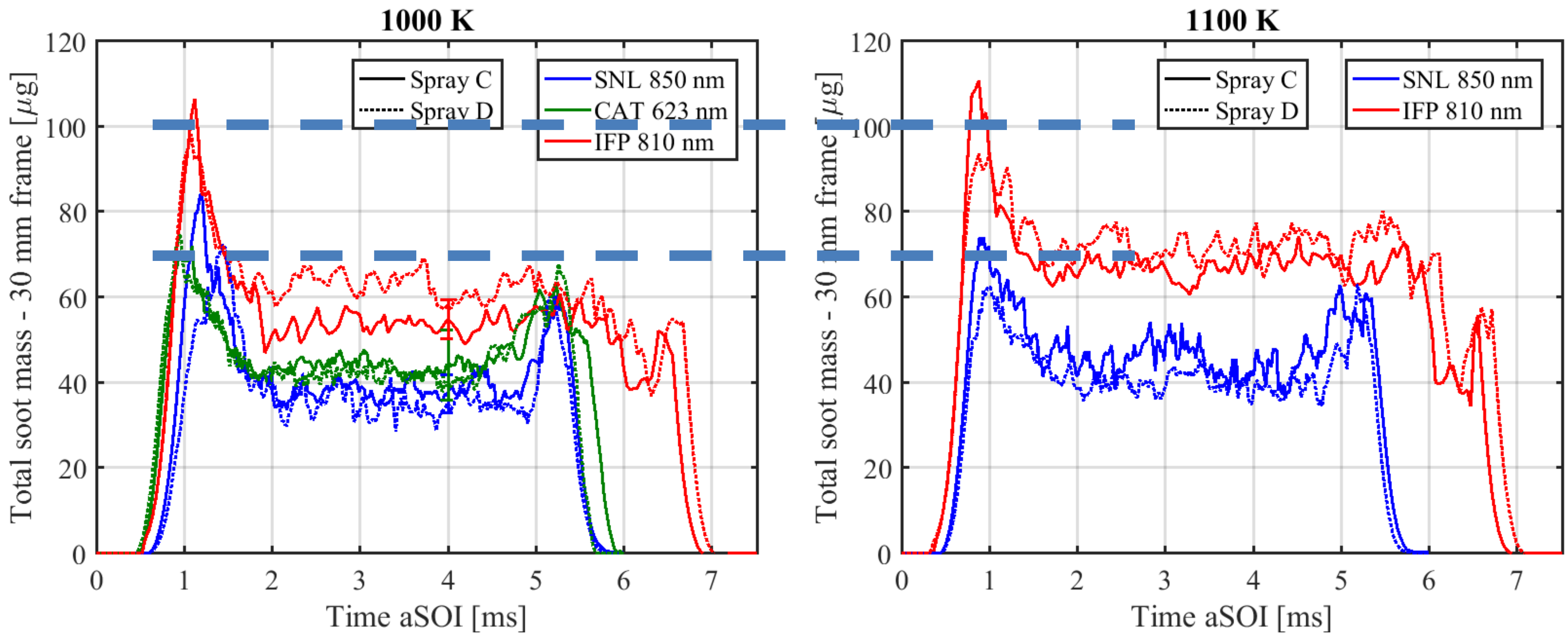




Spray C & D comparison: total soot mass

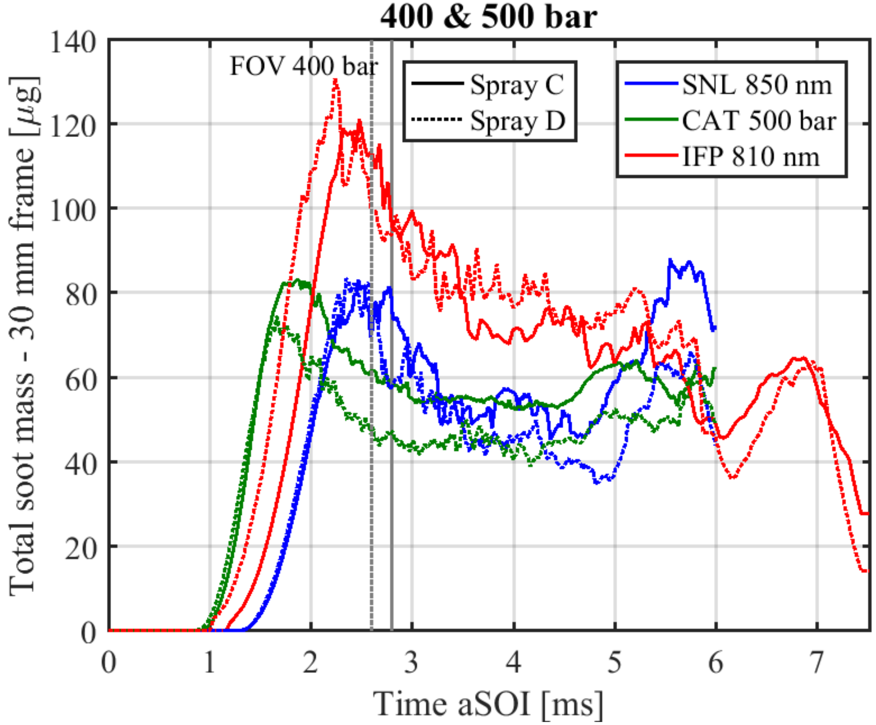


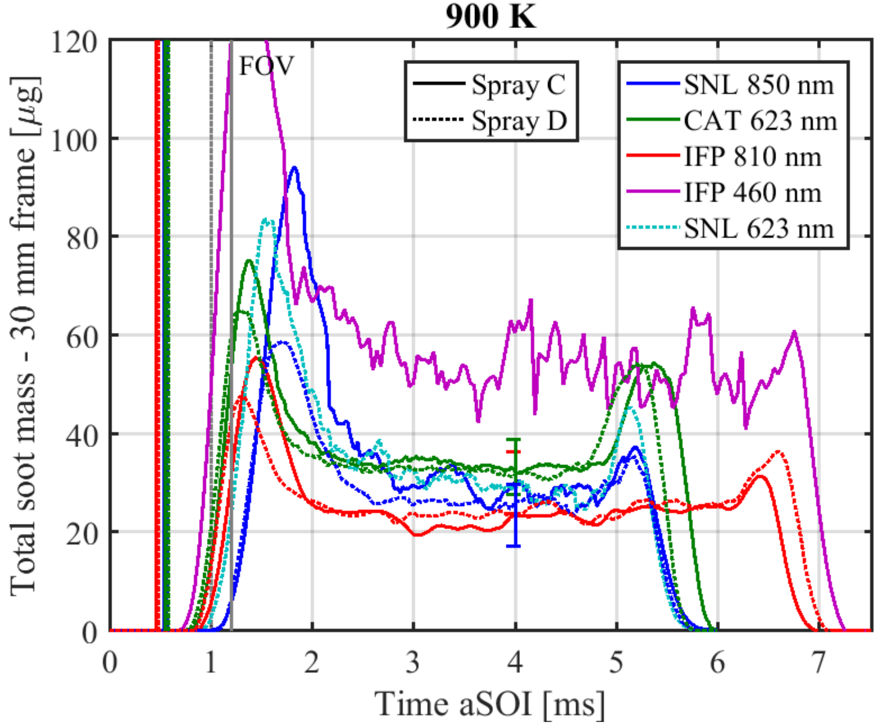
- Peak 1000 K & peak 1100 K similar → area confinement?
- QS: ~10 µg increase for both IFP & SNL
- Large difference IFP C & D @ 1000K, not for other conditions!



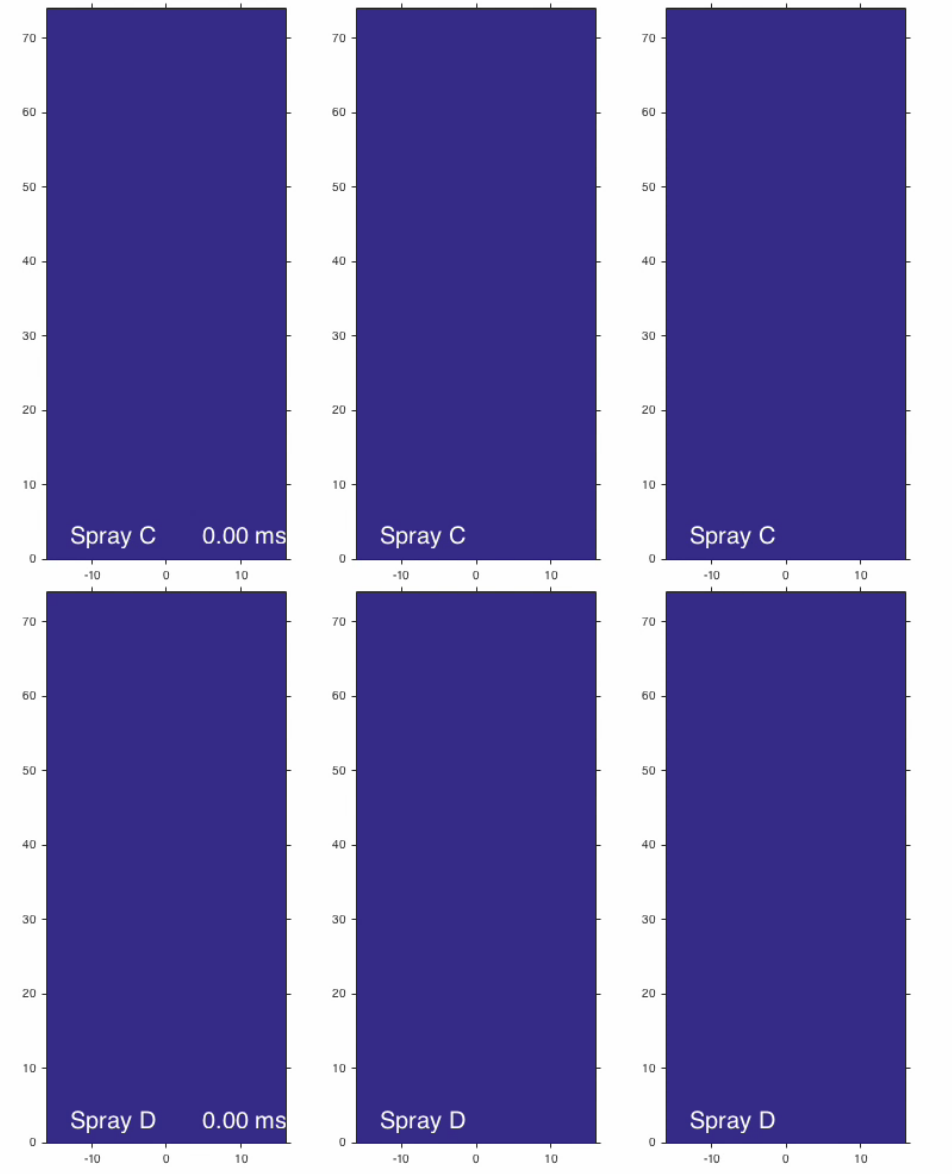
Spray C & D comparison: total soot mass

- Reduced Δp → reduced impact cavitation
- IFP: relative change in start of soot!
- SNL & IFP: not stabilizing → hydraulic influence?
- IFP: increasing sooting propensity significantly
- CAT & SNL: even reducing soot mass → area confinement





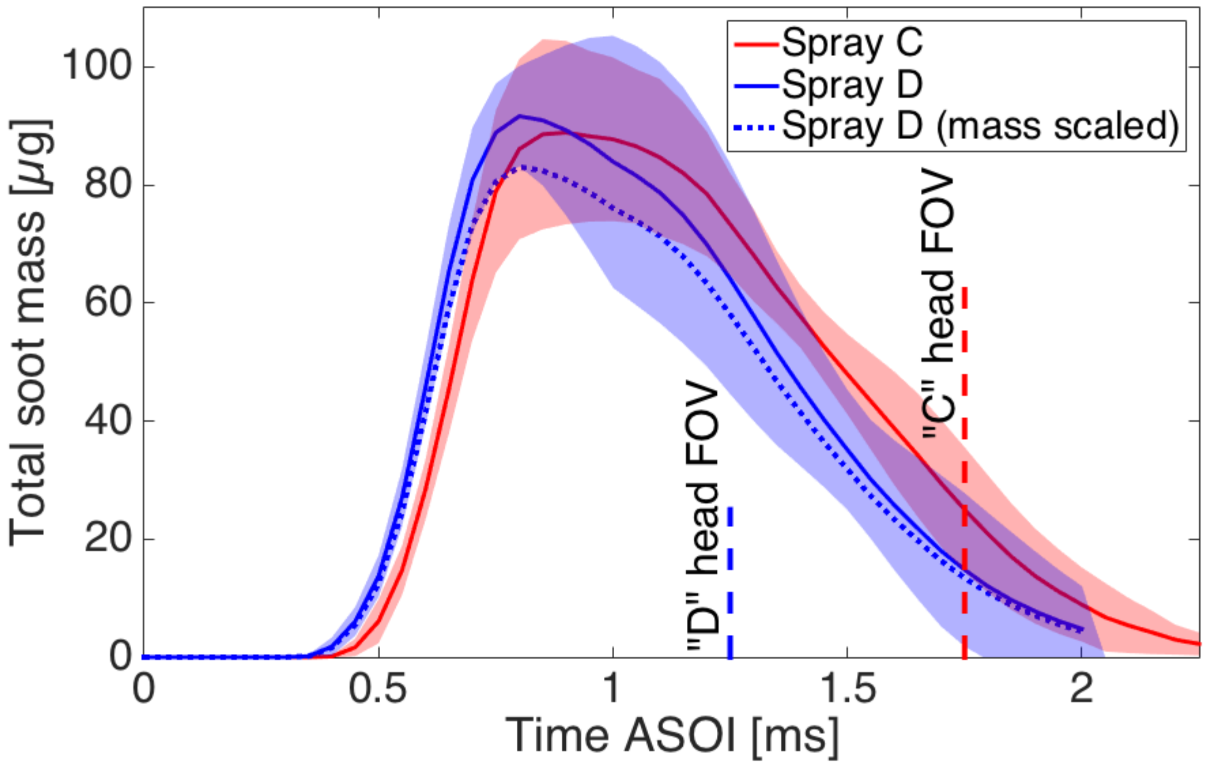
Targeted short injections at high temperature (1200 K) to capture entire event within FOV



- Selected electronic timing based on 30 kPa pressure rise target
- Mean peak pressure
  - Spray C: 29.9 kPa +/- 4.2 kPa (350 µs hydraulic)
  - Spray D: 33.0 kPa +/- 2.6 kPa (330 µs hydraulic)
- Losing some information for Spray D as jet head still reaches extent of FOV before burnout

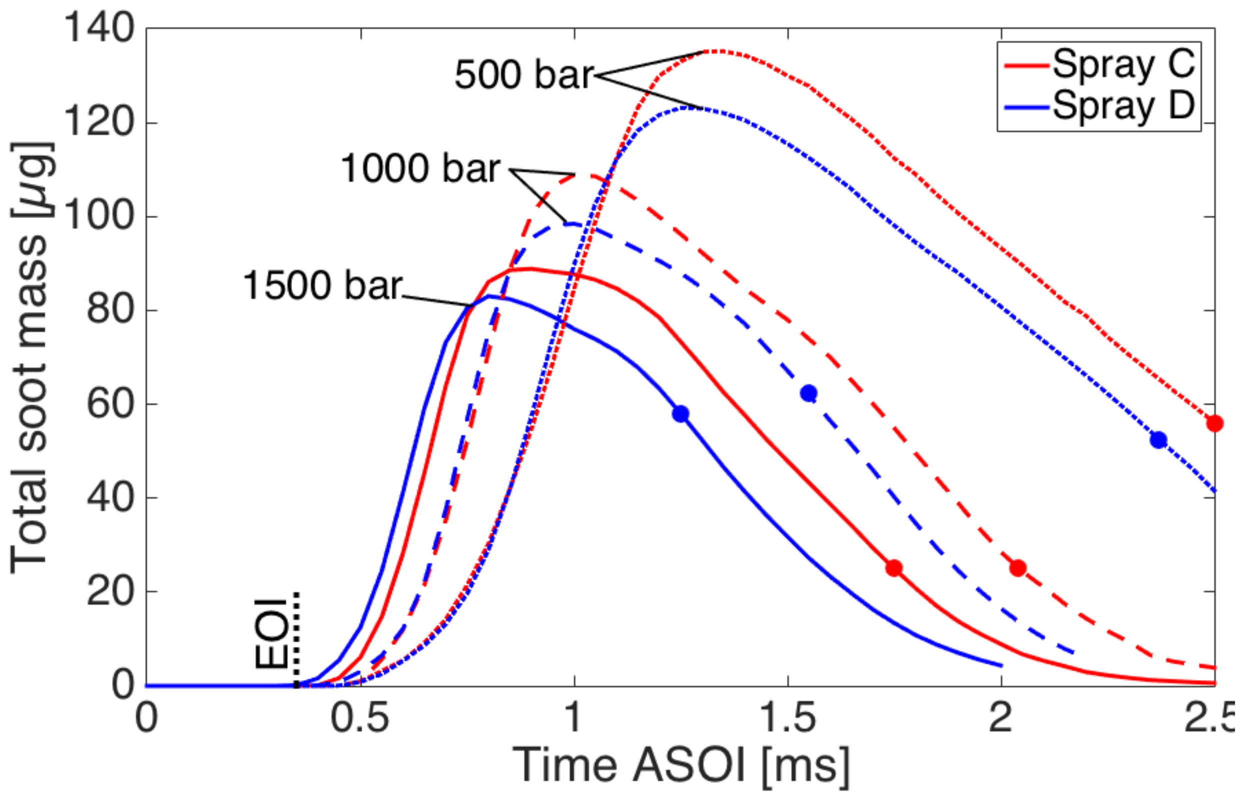
Spray D may form less soot than Spray C when scaled by total energy input (maximum ΔP)

- At 1500 bar injection pressure, total input fuel mass slightly different for C and D (scaling required)



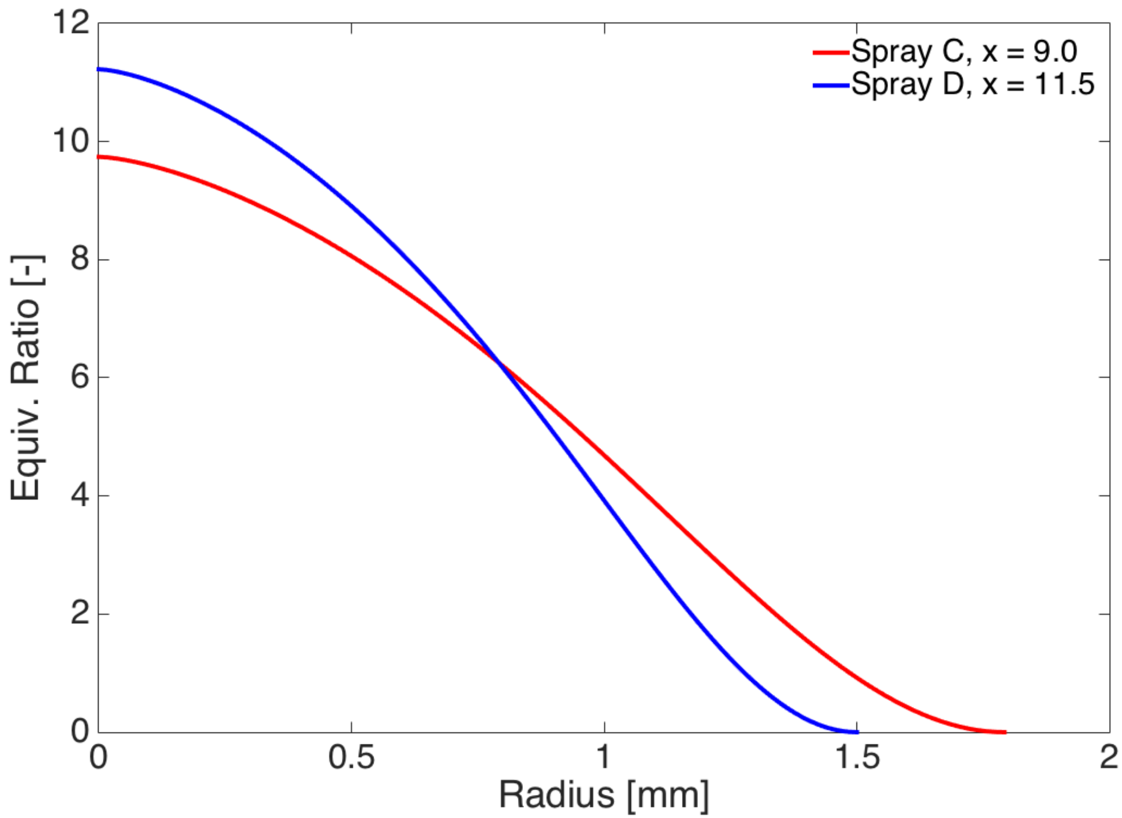
Spray D forms less soot than Spray C when sweeping injection pressure at constant total fuel mass

- Soot forms after EOI, what role does entrainment wave play?
- Interesting that injection pressure does not seem to dramatically impact rate of oxidation (Spray C).



Spray D forms less soot in short injection cases even though spray head is more fuel rich

- At 1200 K lift-off lengths, Spray D is more fuel-rich in the core
- Is core too rich/cold such that soot formation is controlled by regions outside?

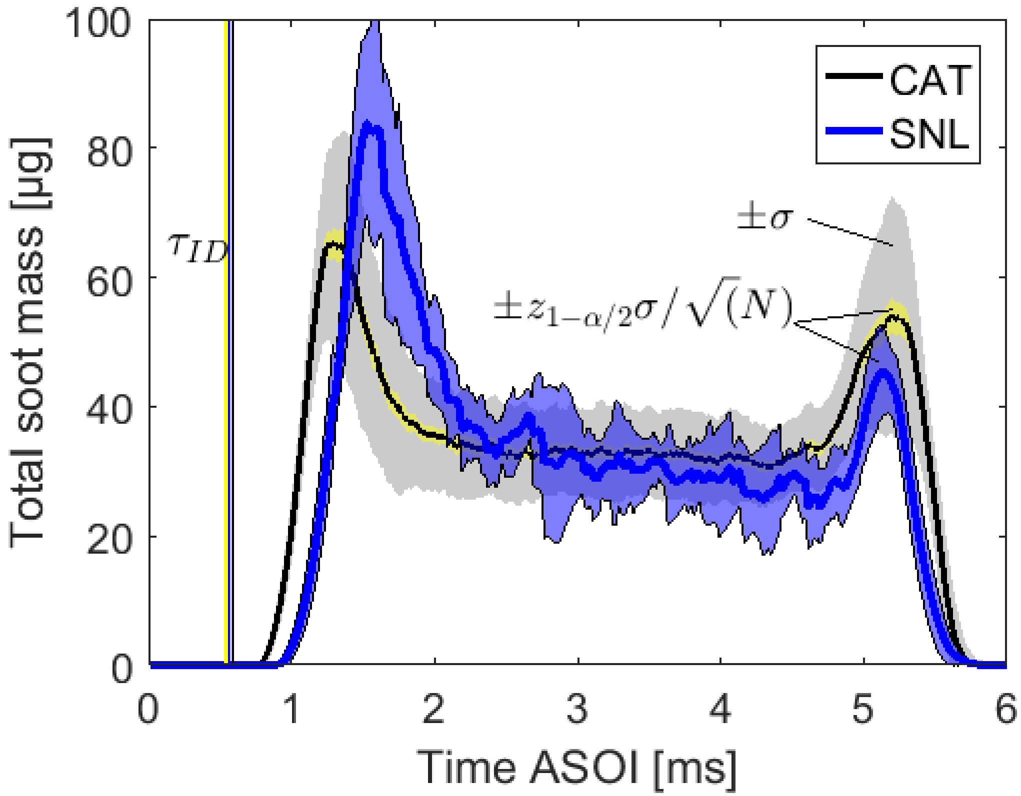


Introduction to ECN5 Modeling

- Modeling focus on pre-burn vs. no-preburn products effects
  - Spray A soot data from GMR constant pressure vessel indicate an earlier onset time and slightly higher total soot mass than data collected at Sandia
    - Investigate ignition delay times and lift-off lengths
    - Soot onset location and characteristics
  - Spray D soot data from CAT constant pressure vessel also show earlier soot onset times but similar quasi-steady total soot mass to Sandia data
- Modelers asked to focus on Spray A baseline condition with only 15%/85% O_2/N_2 mix and with standard pre-burn products that include H_2O and CO_2
  - POLIMI
  - Penn State
  - ETHZ
  - UNSW
  - UW

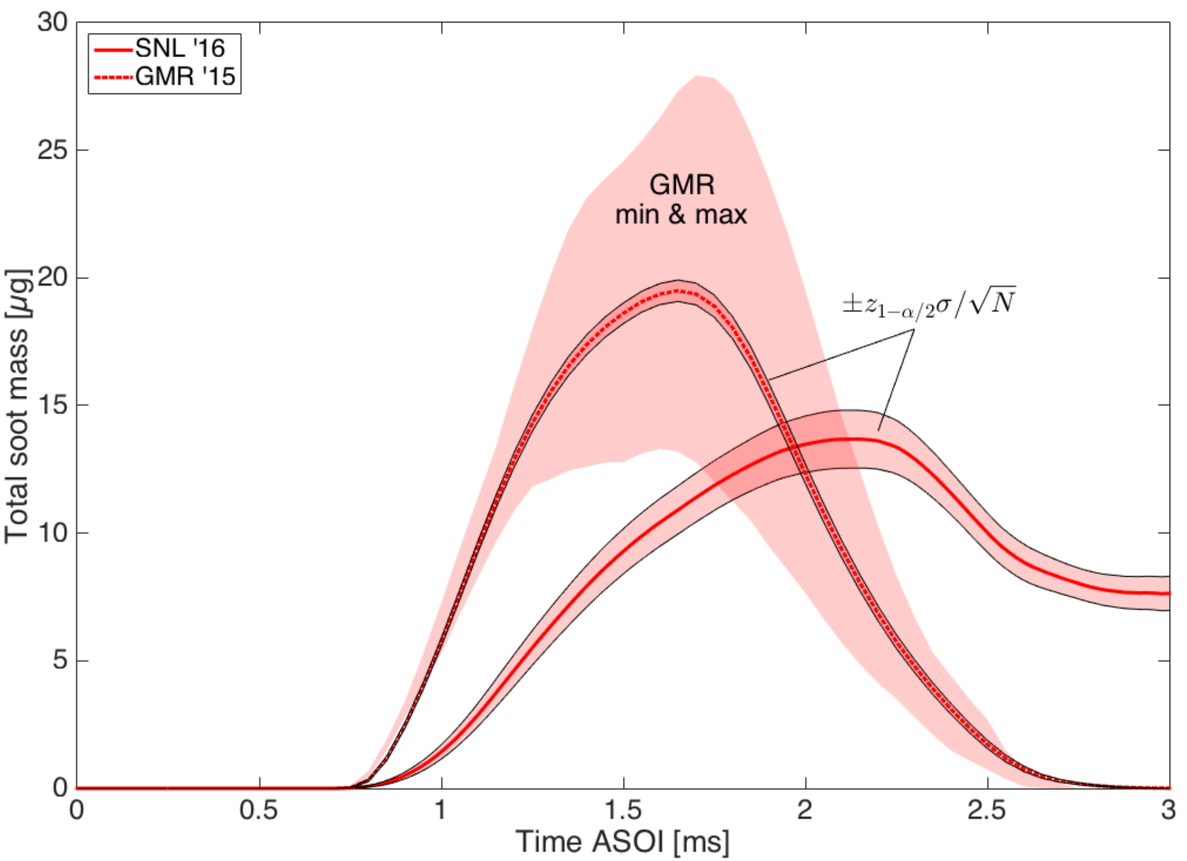
Spray D: Constant pressure vessel with only O_2/N_2 characterized by an earlier soot onset time but similar soot mass in head and quasi-steady

- 200 inj. conducted at CAT with Spray D provide significant statistics and an opportunity to consider shot-to-shot variability
- Constant pressure vessel with only O_2/N_2 characterized by an earlier soot onset time; however peak soot in head is lower and quasi-steady soot is similar
- SNL: ID = **560 μs ± 26 μs**
  - **LOL: 26.4 mm**
- CAT:
  - **Head-on: ID = 544 μs ± 3 μs**
  - **Side view: ID = 576 μs ± 33 μs (different experiments)**
    - **LOL: 25.7 mm**
- Again, note how consistent formation time and rate are in the CAT data—indicative of deterministic behavior



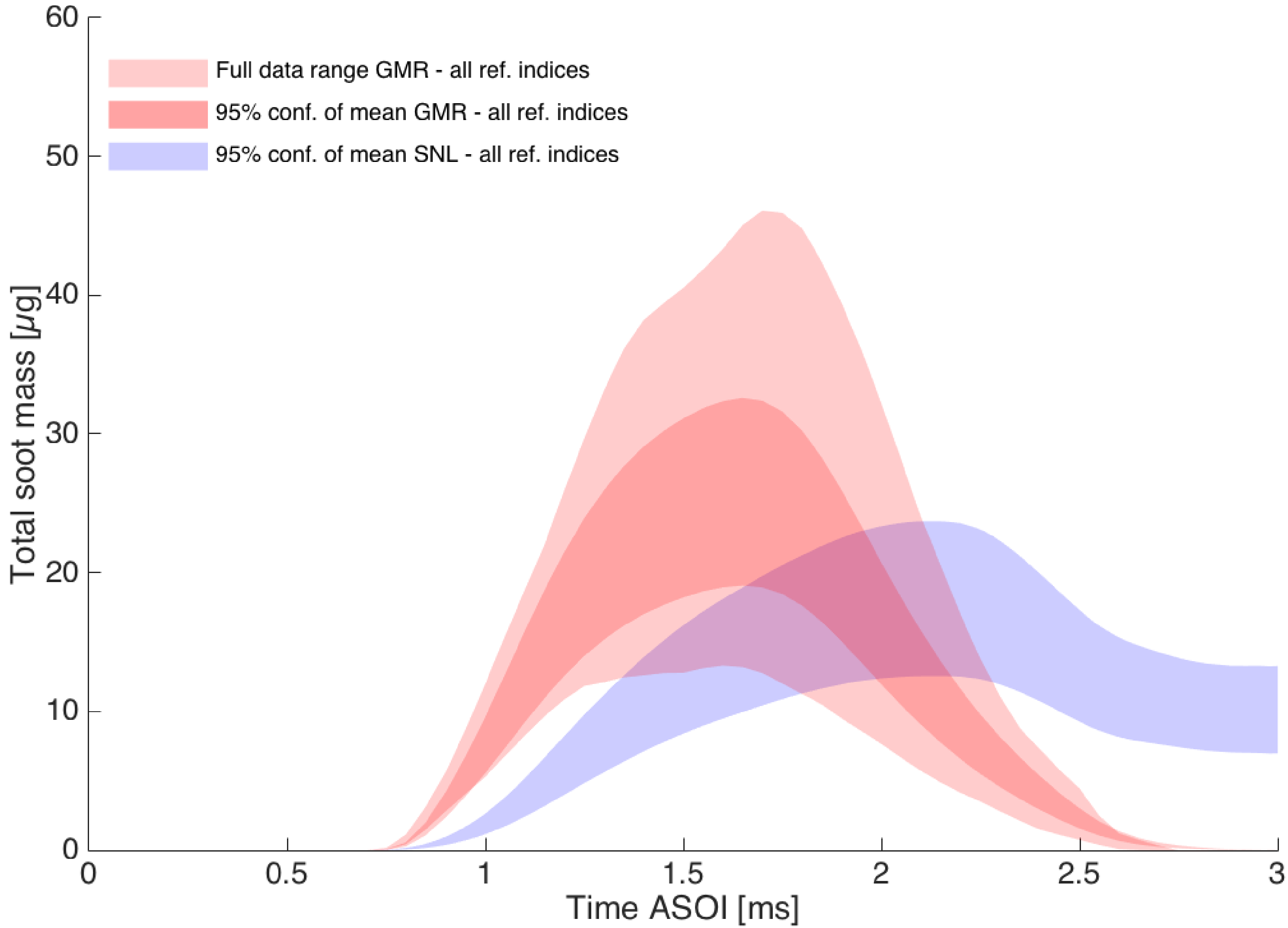
Spray A: Constant pressure vessel with only O_2/N_2 characterized by an earlier soot onset time and a higher total soot mass in the head

- 200 injections conducted at GMR with Spray A provide significant statistics and an opportunity to consider shot-to-shot variability
- Constant pressure vessel with only O_2/N_2 characterized by an earlier soot onset time and a higher total soot mass in the head
- SNL: ID = 423 μs ± 11 μs
- GMR:
  - **Head-on: ID = 408 μs ± 2 μs**
  - **Side view: ID = 380 μs (different experiments)**
- Note how consistent formation time and rate are in the GMR data—indicative of deterministic behavior

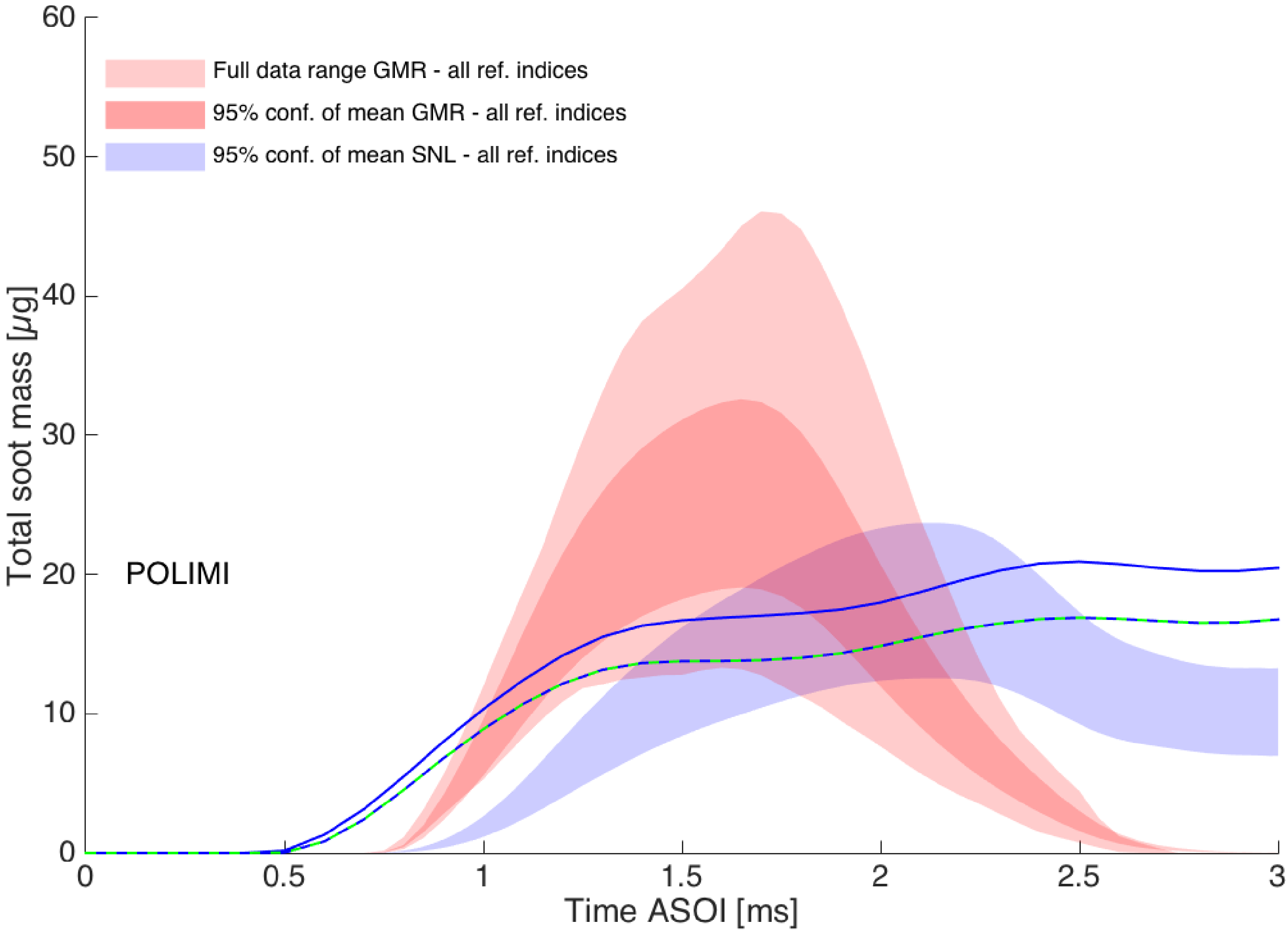


<u>What do the models say about the influence of the pre-burn products?</u>

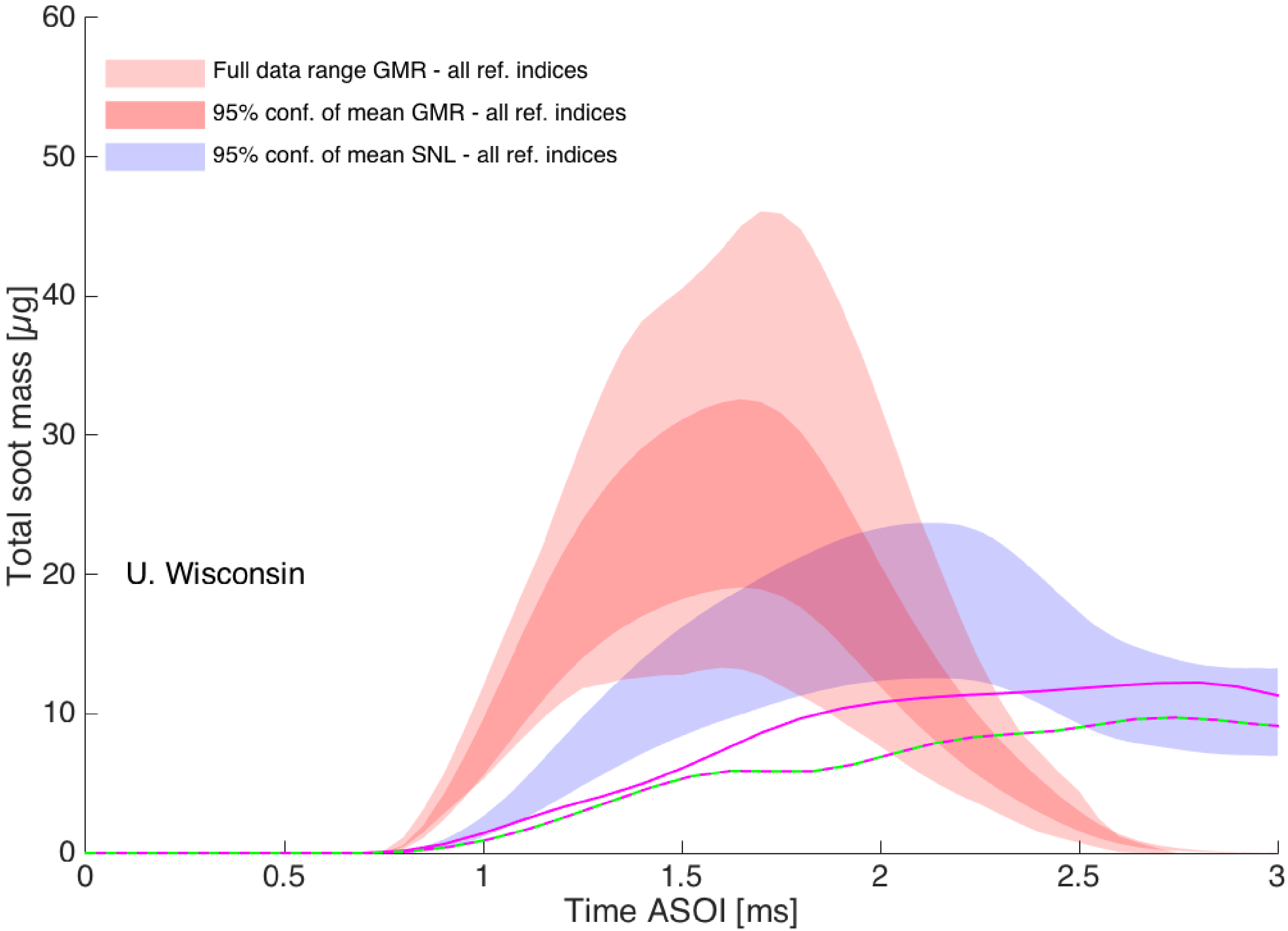
Modeled total soot mass has improved for most institutions since ECN4



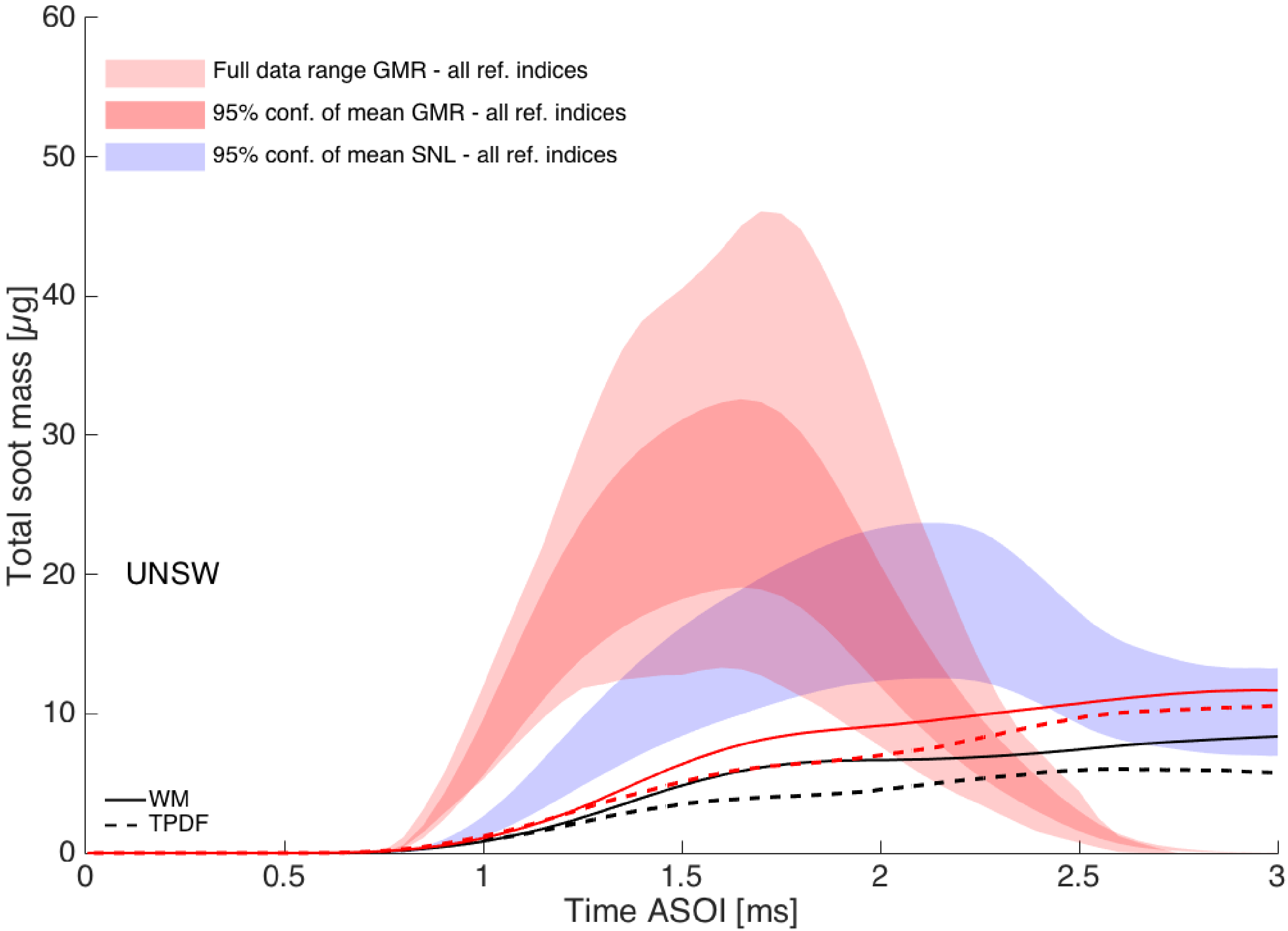
Modeled total soot mass has improved for most institutions since ECN4



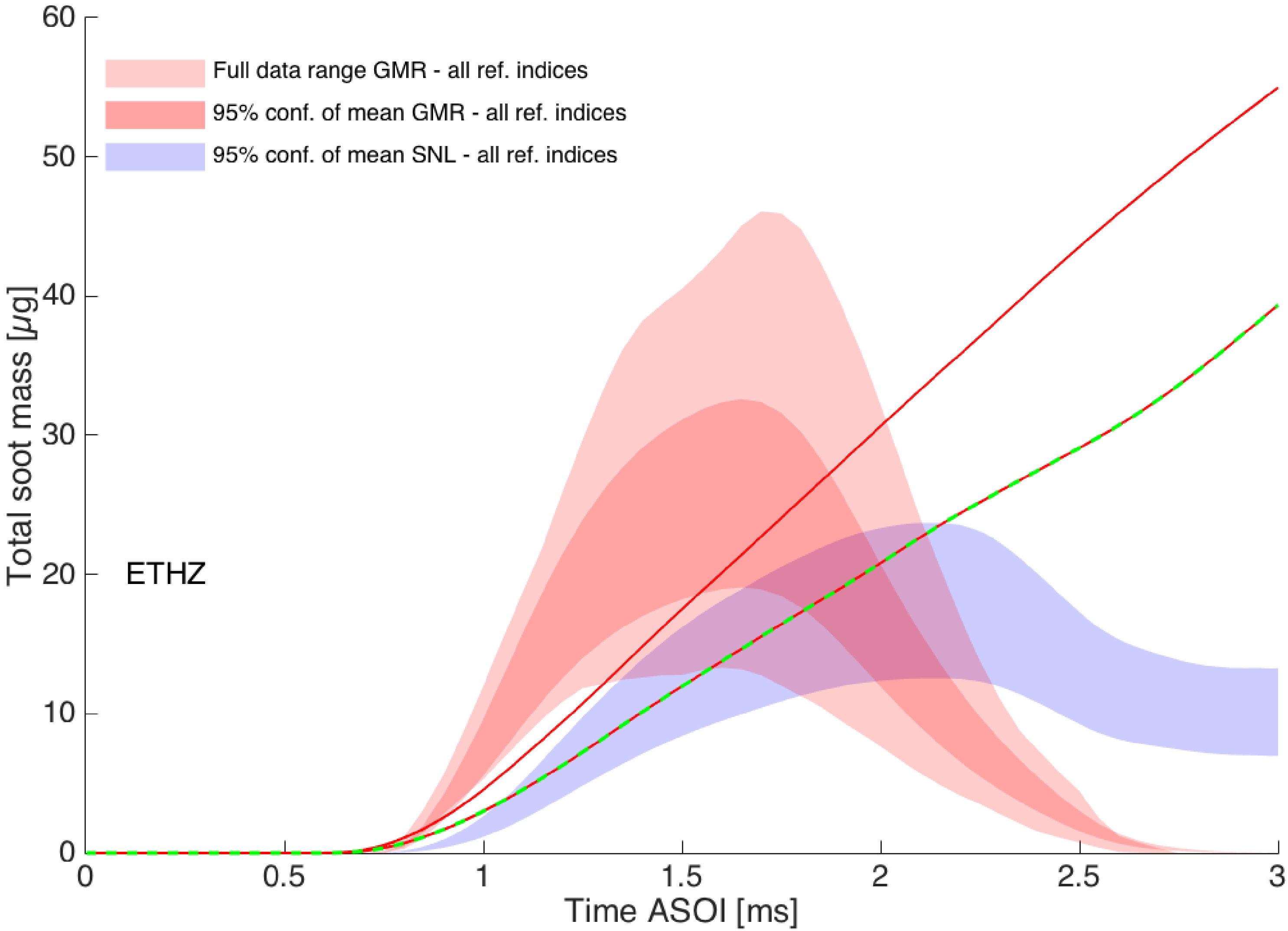
Modeled total soot mass has improved for most institutions since ECN4



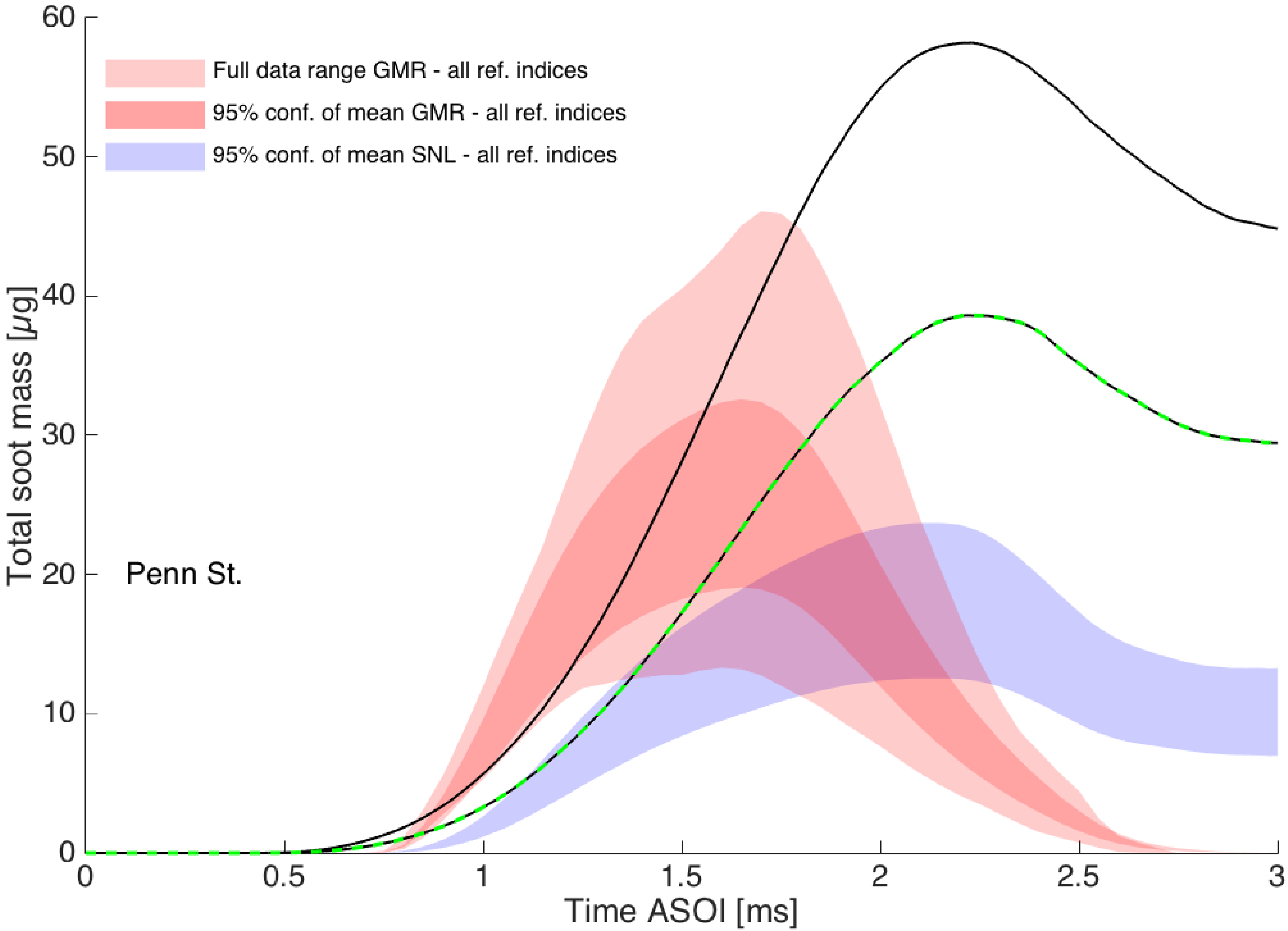
Modeled total soot mass has improved for most institutions since ECN4



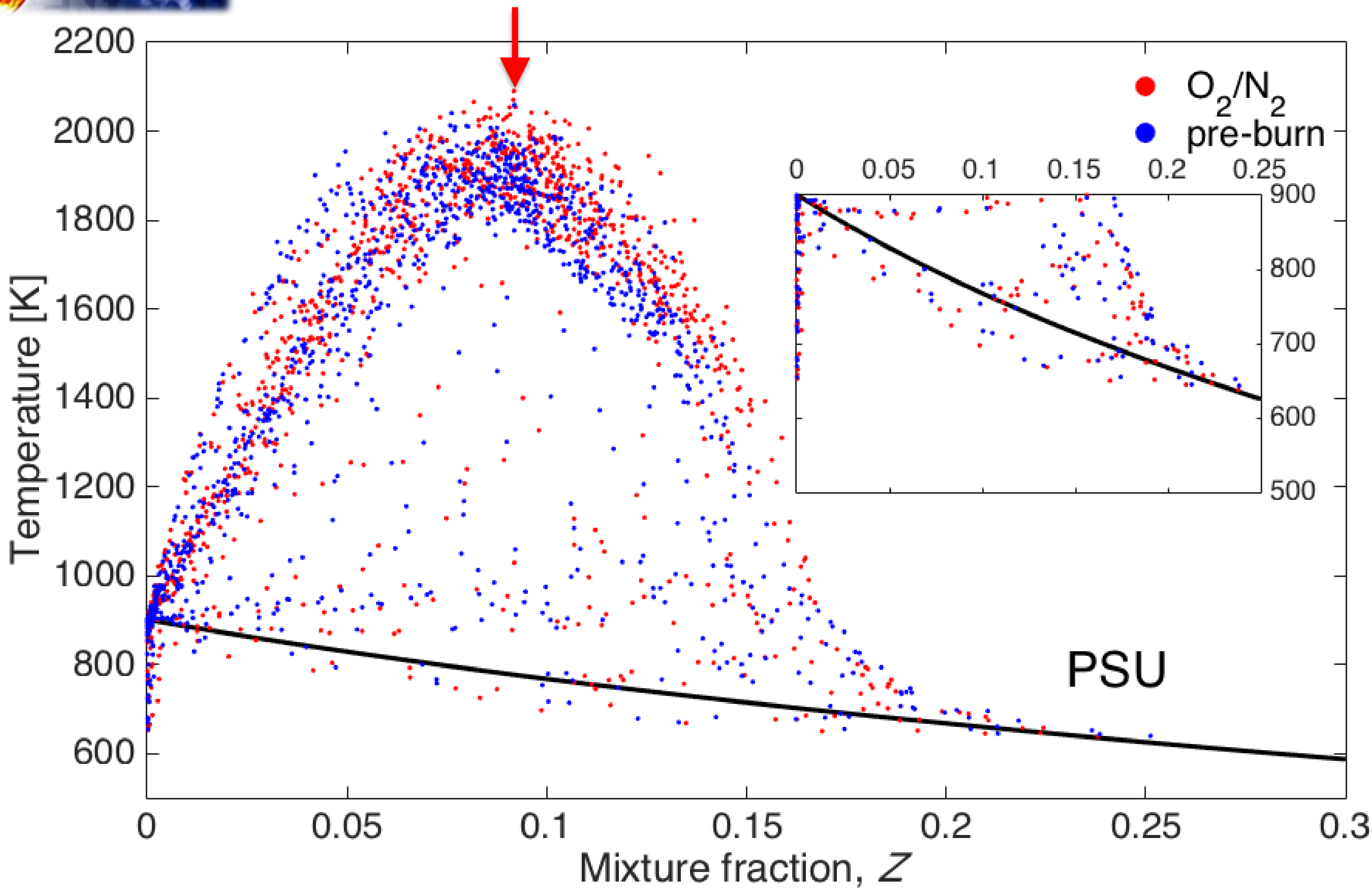
Modeled total soot mass has improved for most institutions since ECN4



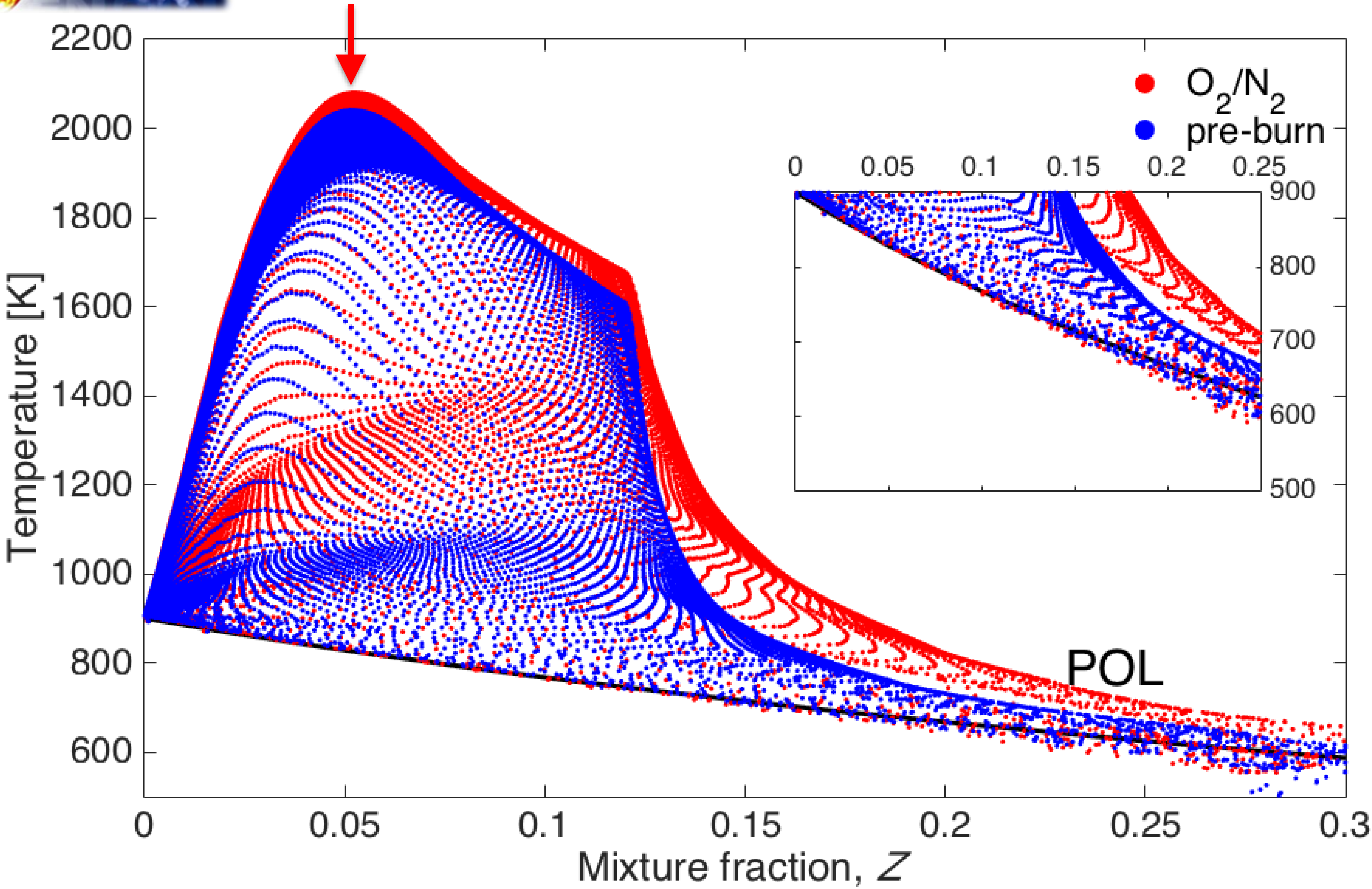
Modeled total soot mass has improved for most institutions since ECN4



Penn State Temperature vs. Mixture fraction

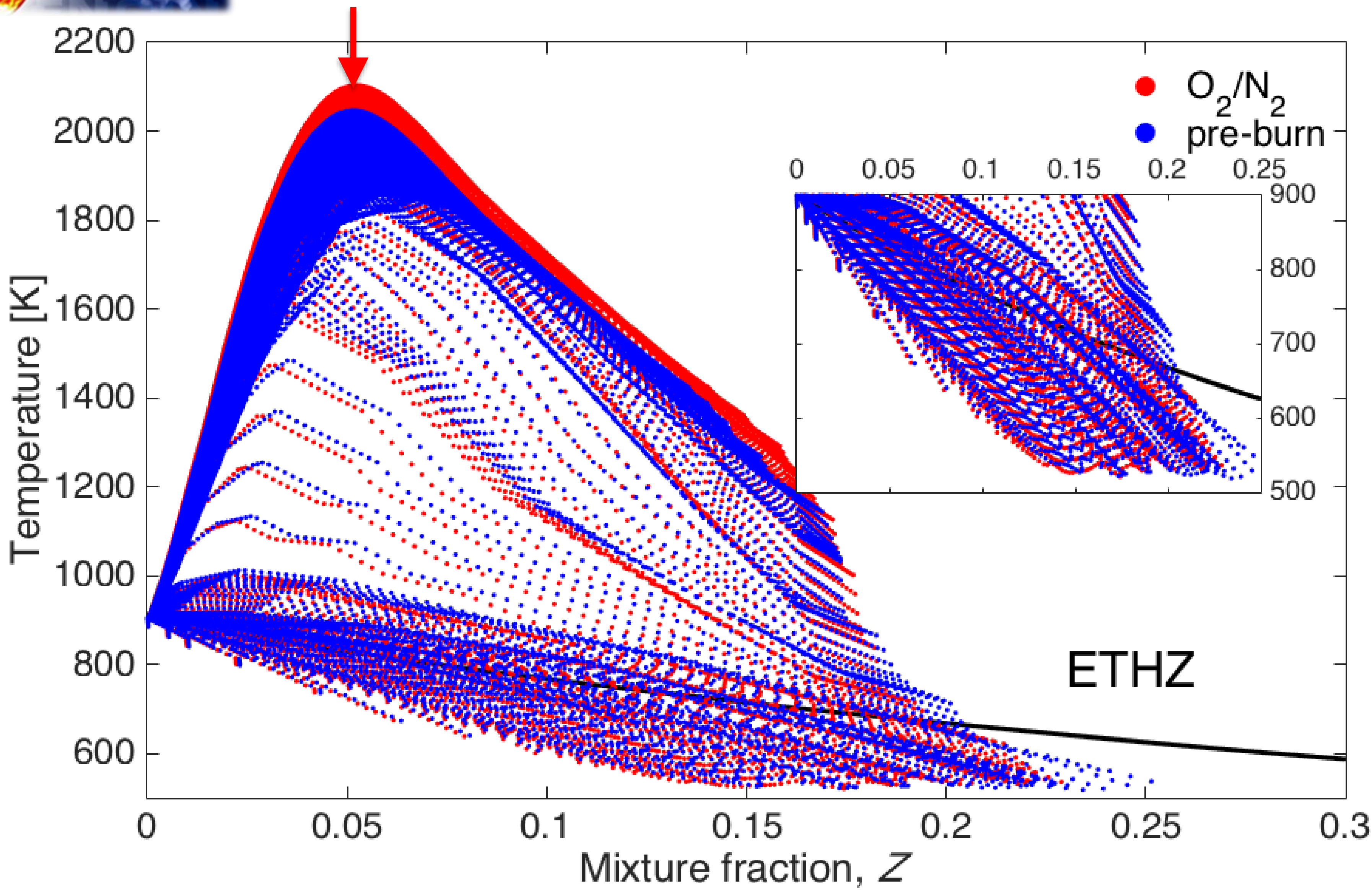


POLIMI T vs. Z



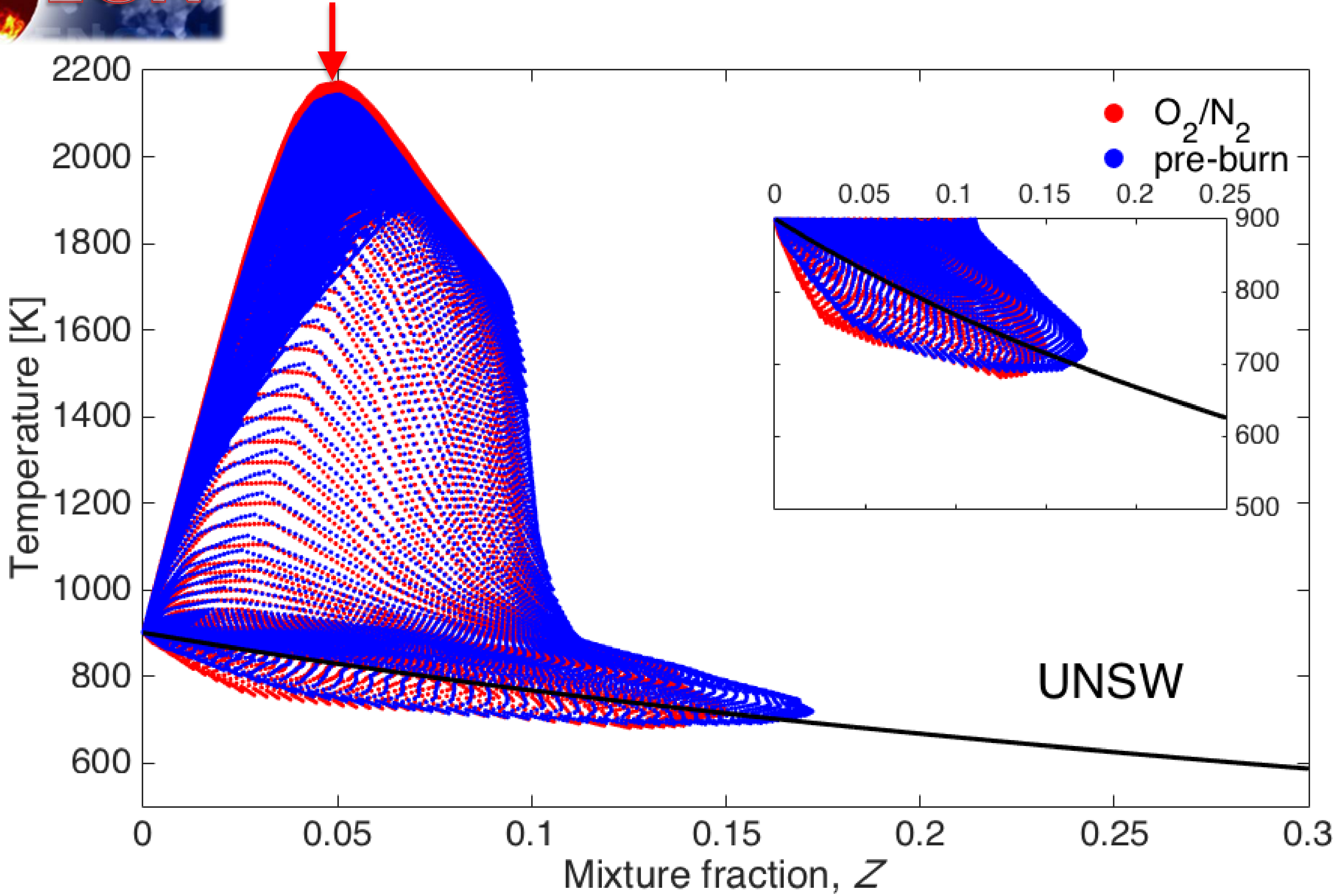
Temperature [K]
2200
2000
1800
1600
1400
1200
1000
800
600
0
0.05
0.1
0.15
0.2
0.25
0.3
Mixture fraction, Z
O_2/N_2
pre-burn
POL
900
800
700
600
500

ETHZ T vs. Z



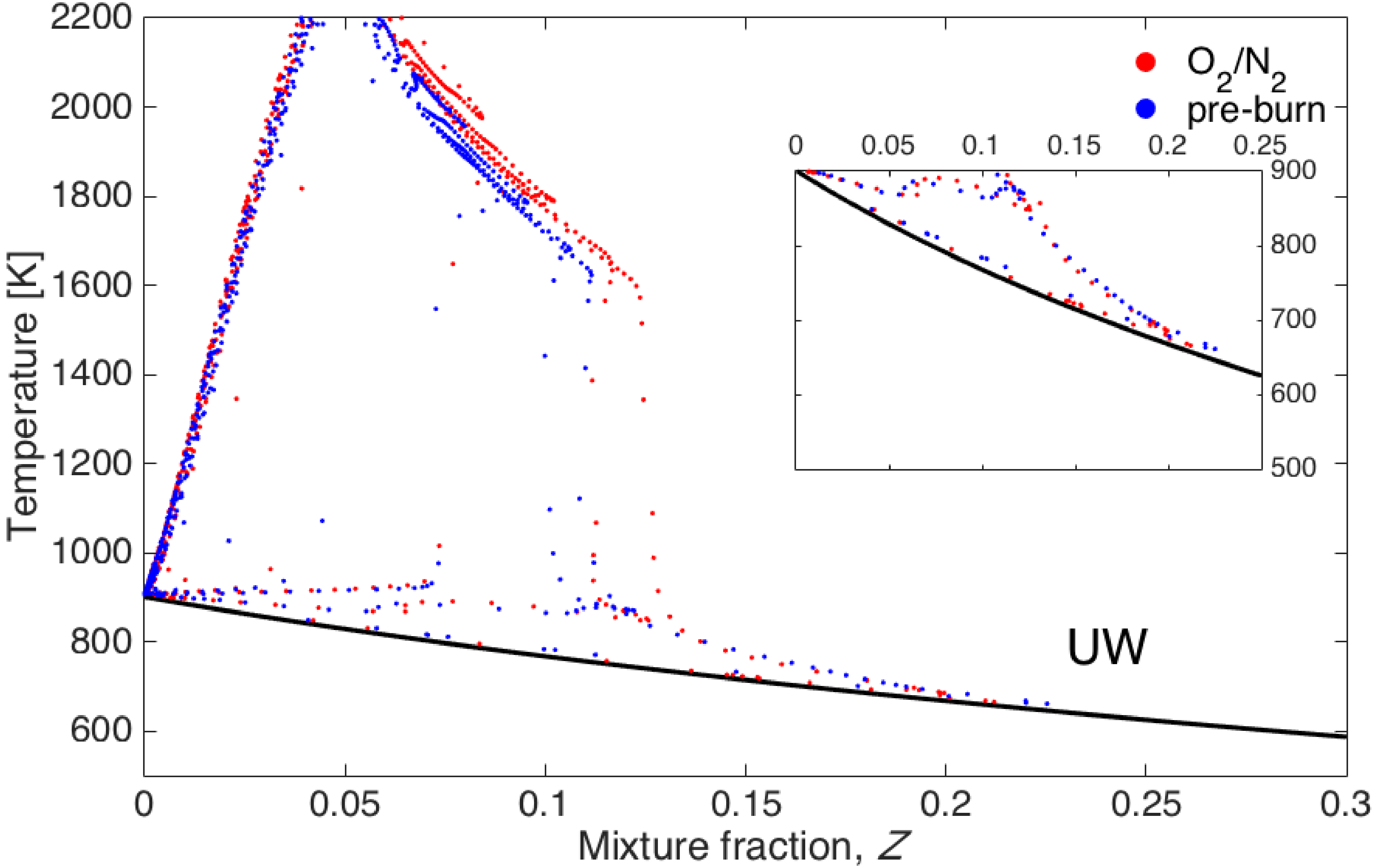
Temperature [K]
2200
2000
1800
1600
1400
1200
1000
800
600
0
0.05
0.1
0.15
0.2
0.25
0.3
Mixture fraction, Z
O2/N2
pre-burn
ETHZ
900
800
700
600
500

UNSW T vs. Z

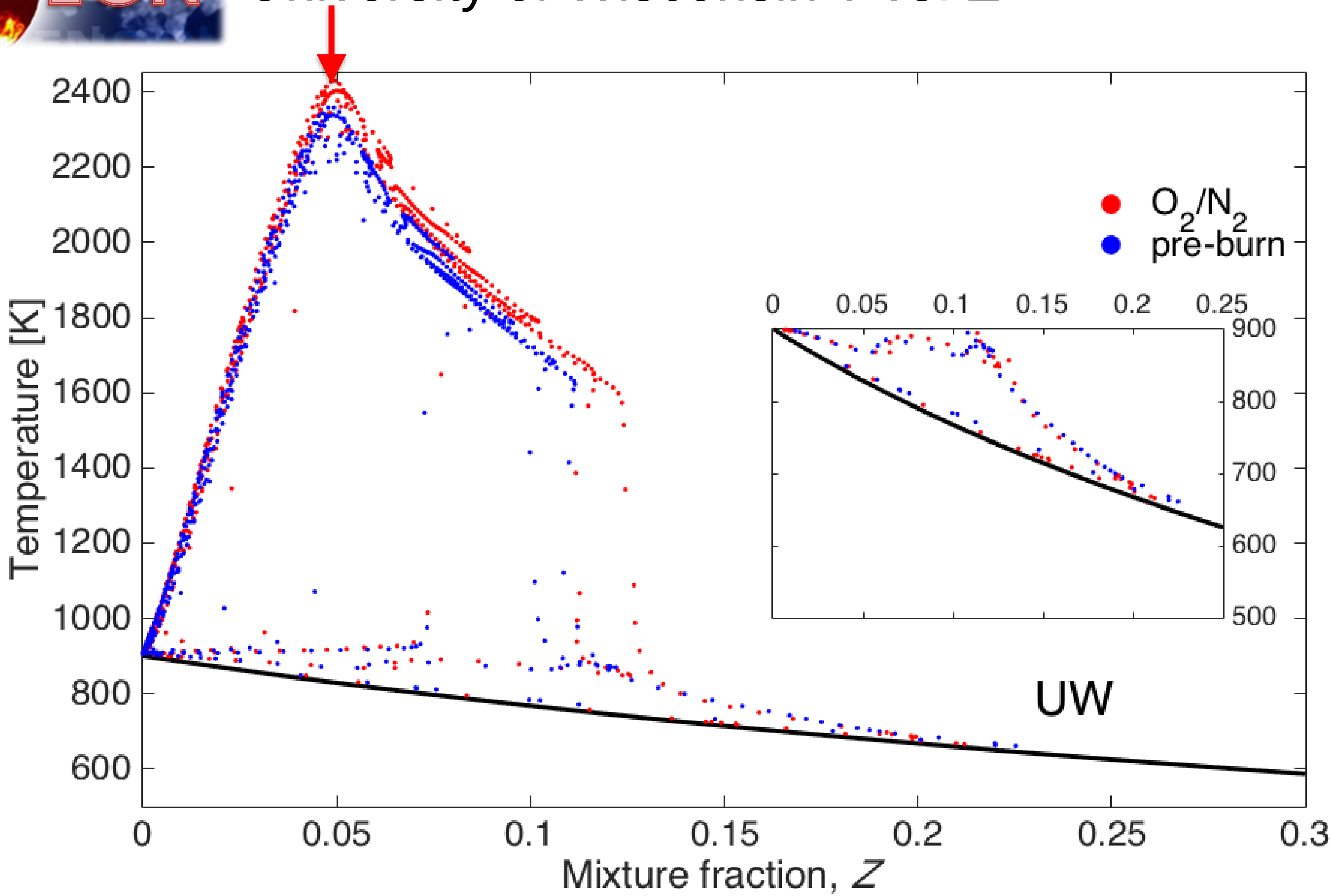


Temperature [K]
Mixture fraction, Z
O_2/N_2
pre-burn
UNSW
2200
2000
1800
1600
1400
1200
1000
800
600
0
0.05
0.1
0.15
0.2
0.25
0.3
900
700
500

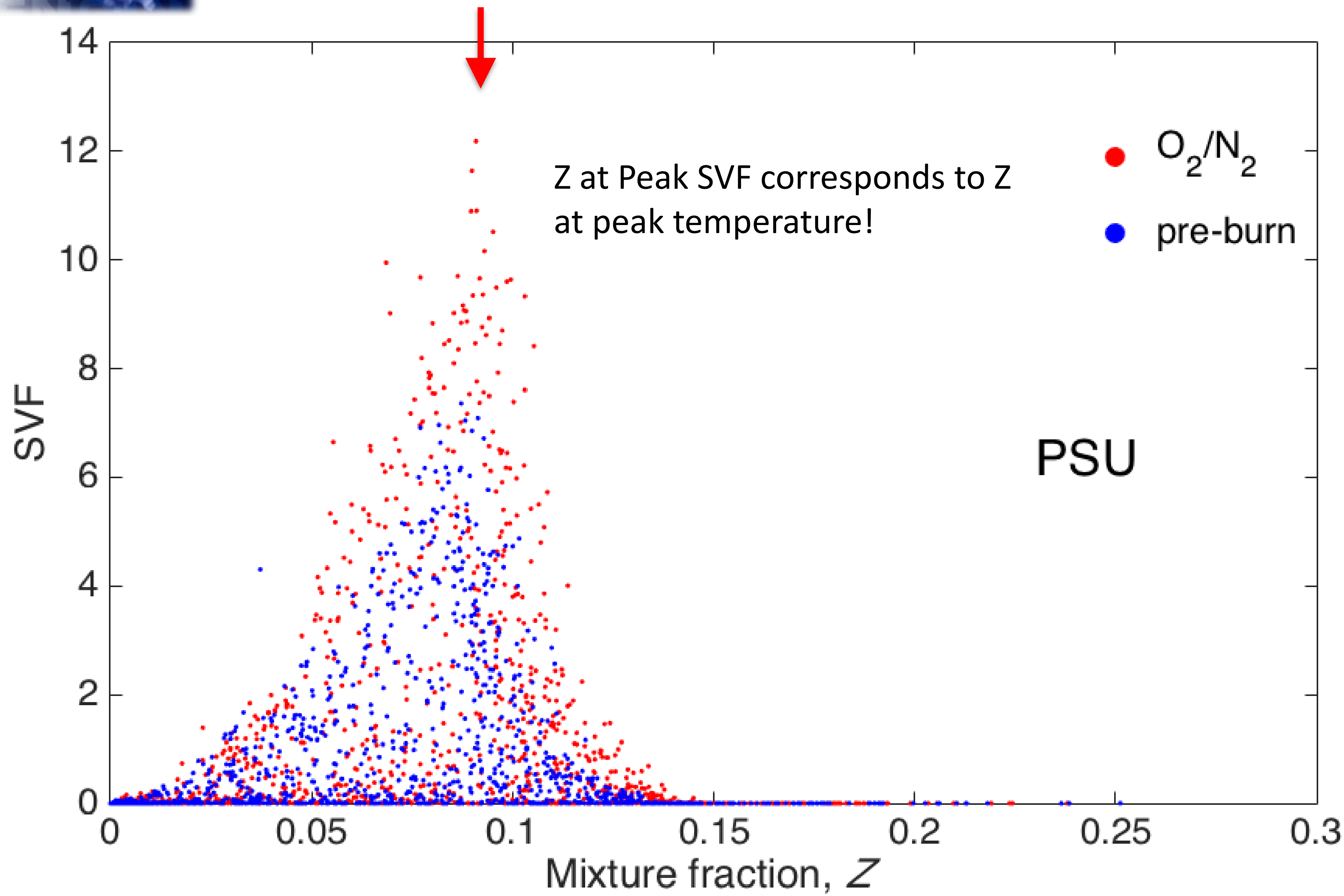
University of Wisconsin T vs. Z



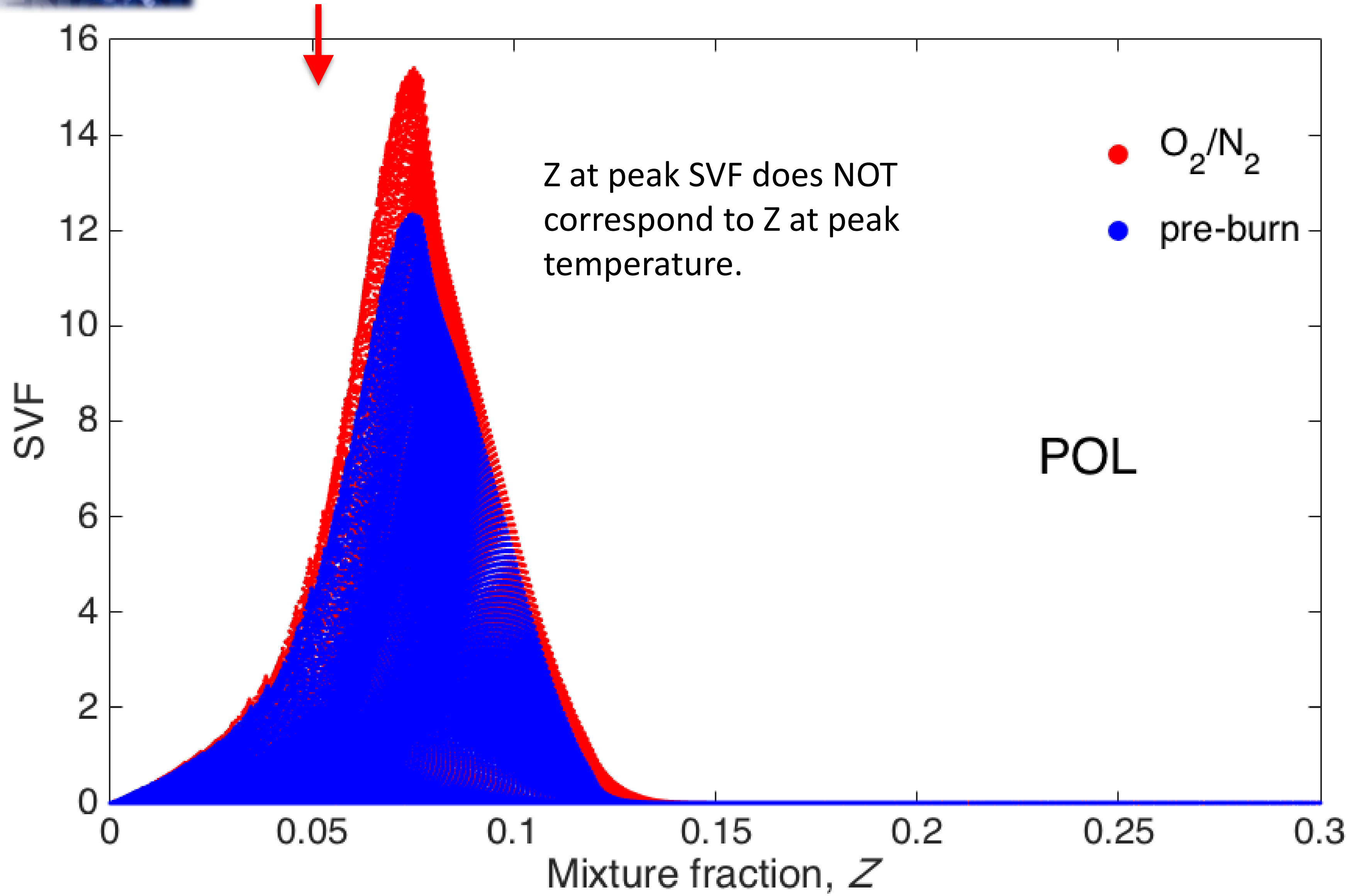
University of Wisconsin T vs. Z



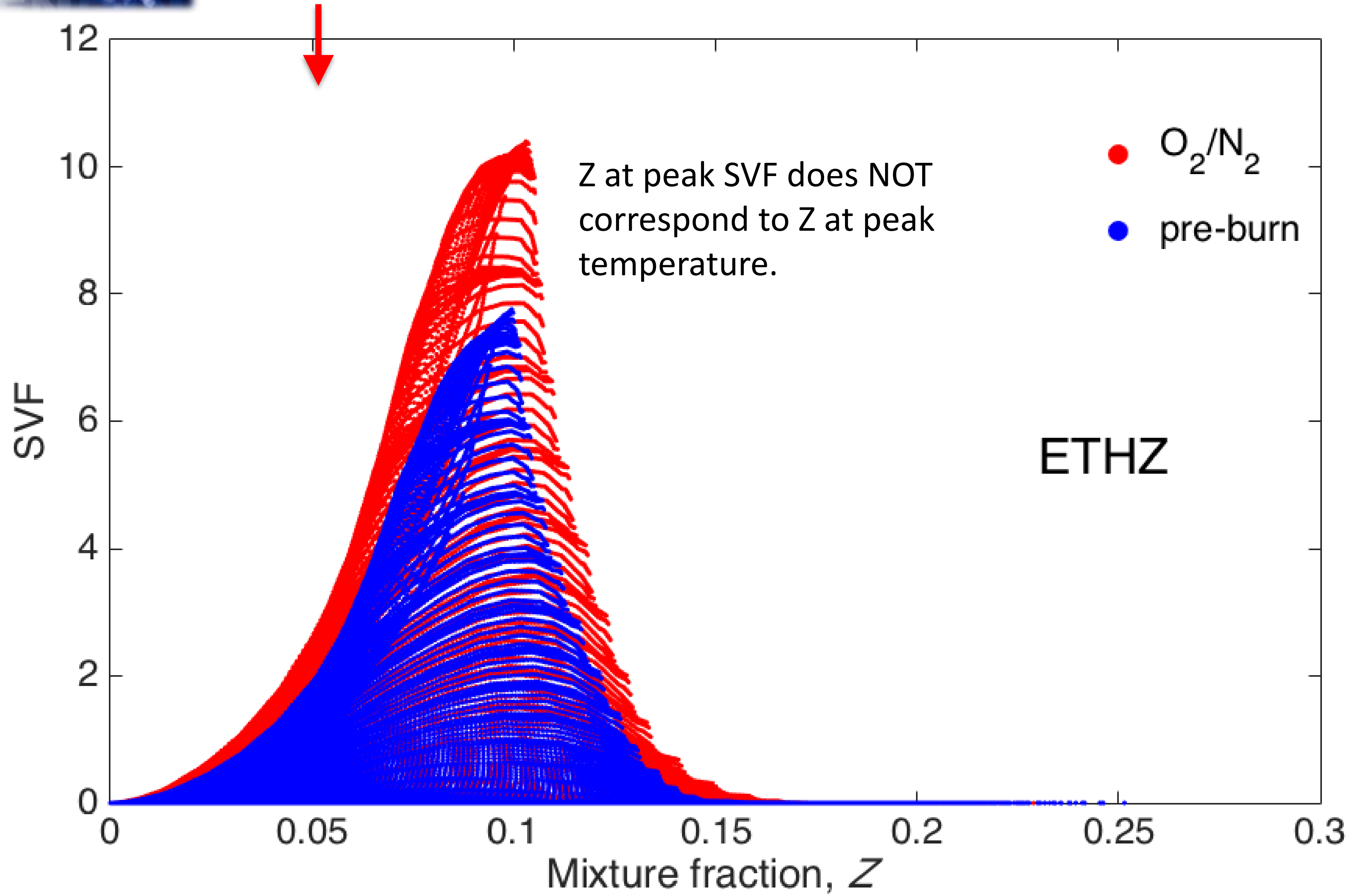
Penn State SVF vs. Mixture fraction



POLIMI SVF vs. Z

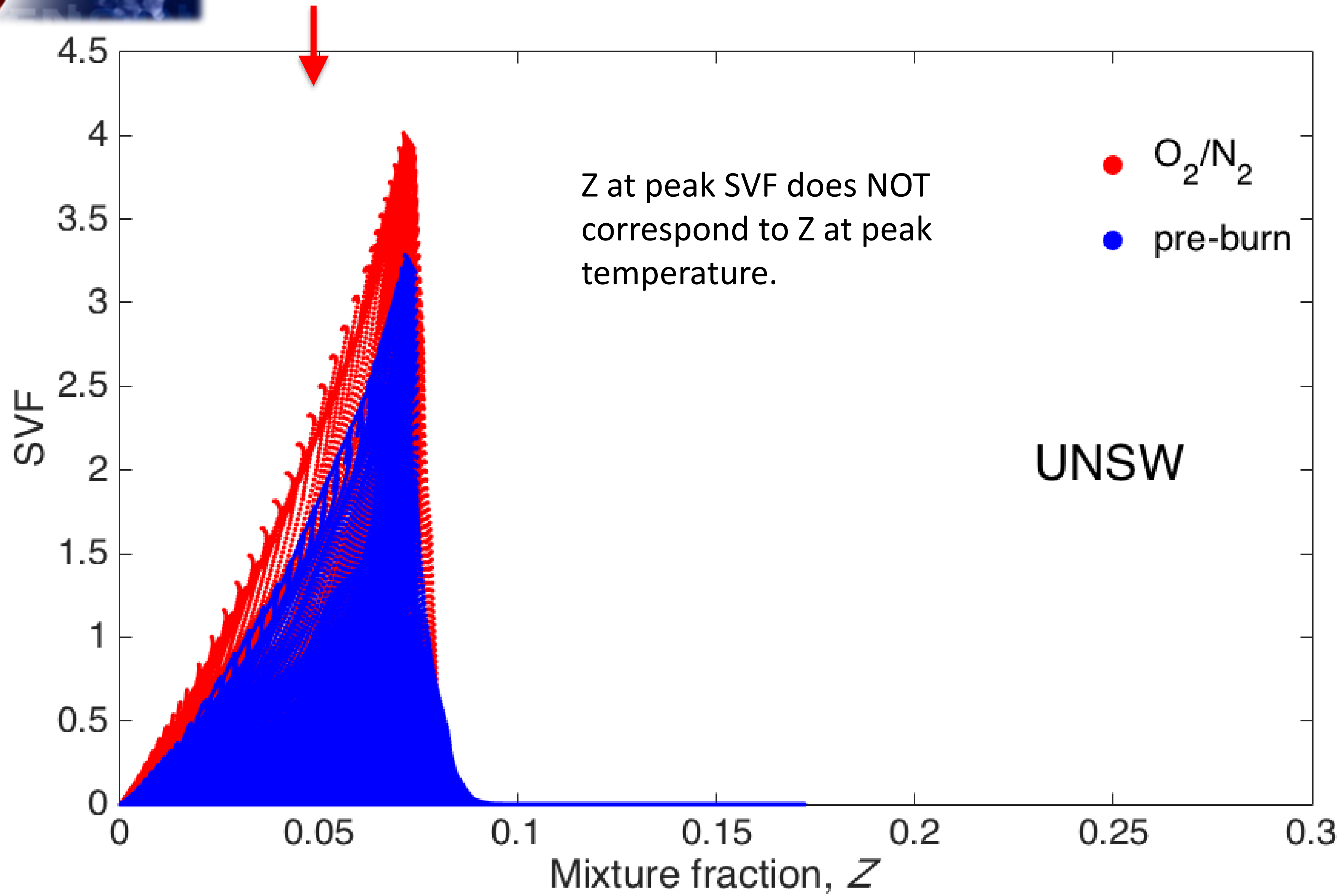


ETHZ T vs. Z



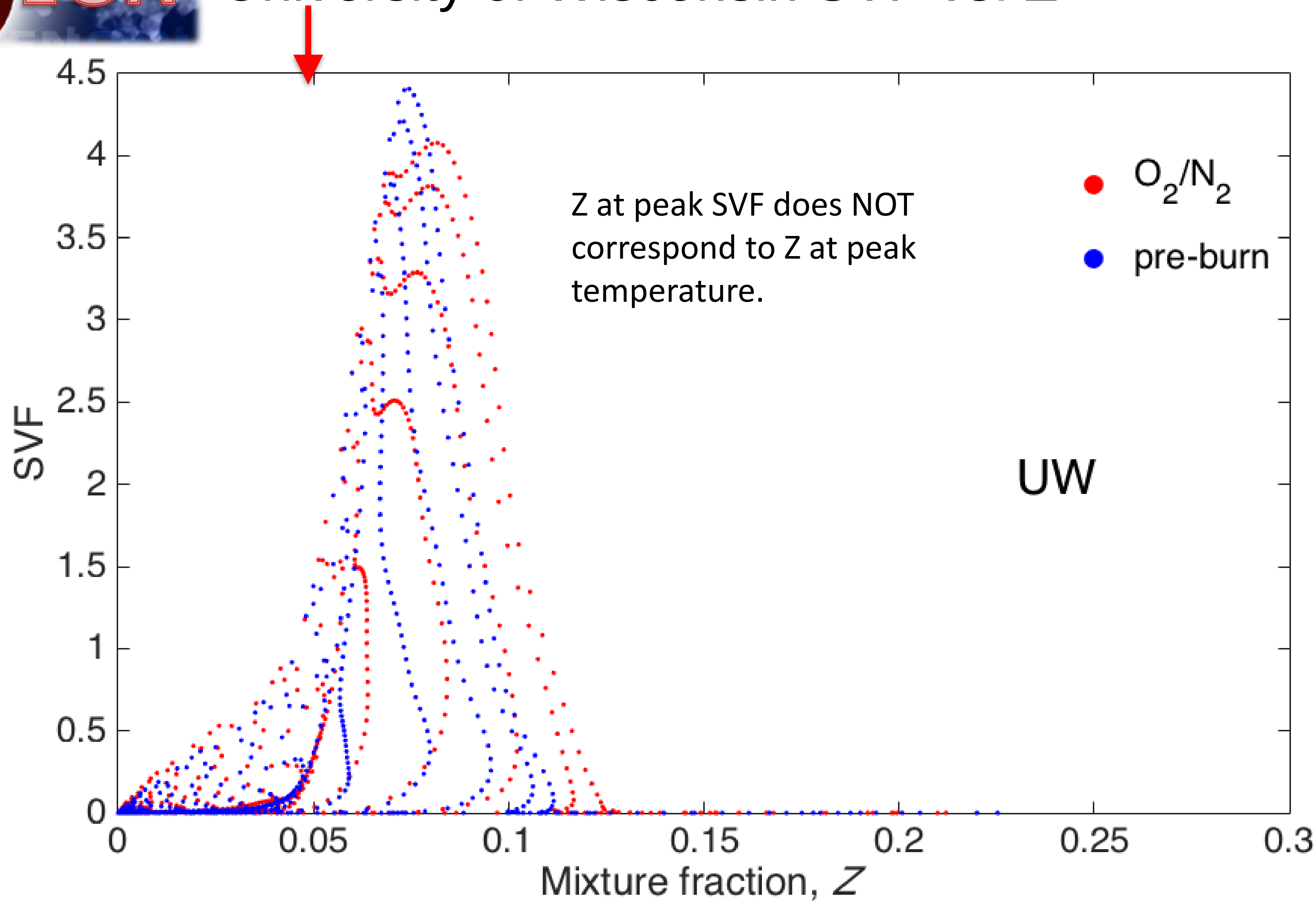
Z at peak SVF does NOT correspond to Z at peak temperature.
O_2/N_2
pre-burn
ETHZ
SVF
Mixture fraction, Z
0
2
4
6
8
10
12
0
0.05
0.1
0.15
0.2
0.25
0.3

UNSW SVF vs. Z



Z at peak SVF does NOT correspond to Z at peak temperature.

University of Wisconsin SVF vs. Z



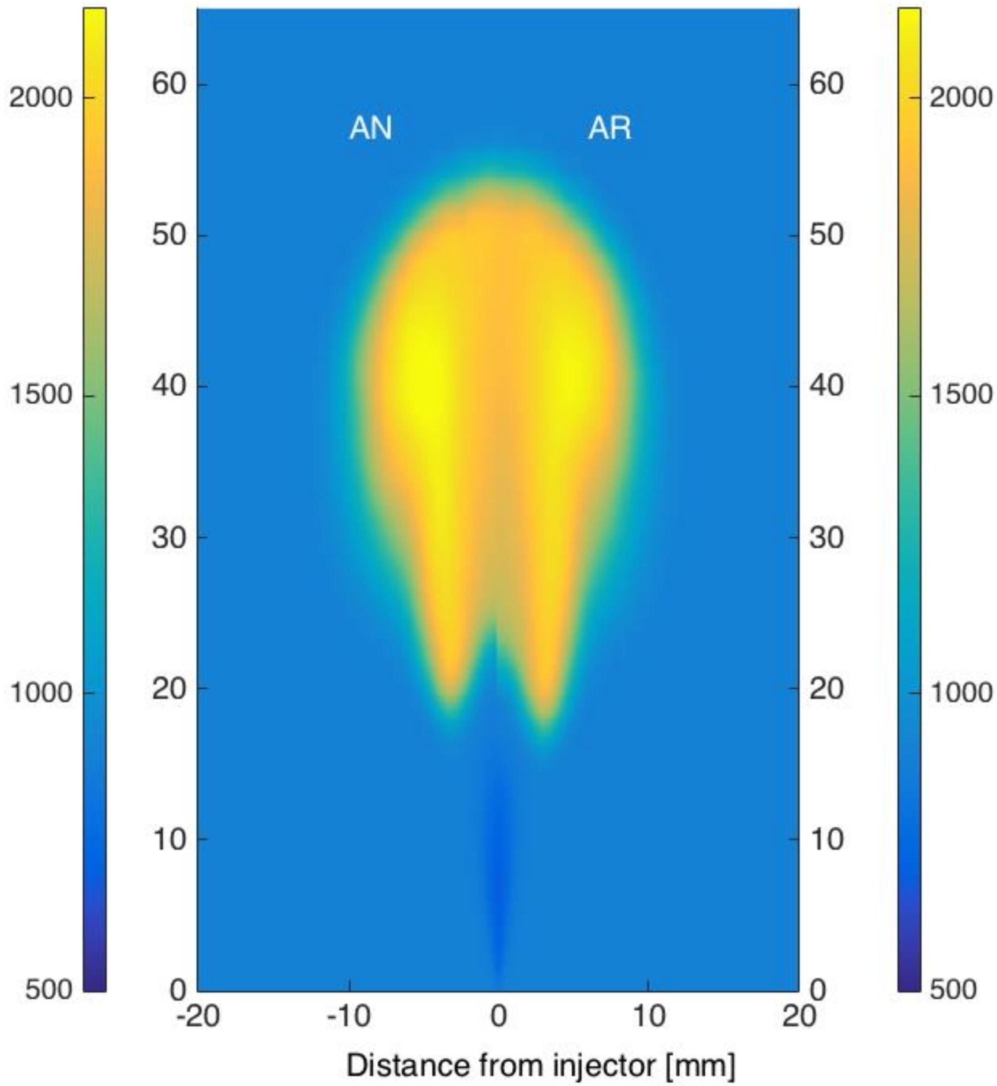
What can we learn by comparing T, OH, C_2H_2, SVF in AN and AR cases?

- Expectation is that the presence of CO_2 and H_2O should increase the OH concentration due to a slowing of the CO and H_2 oxidation reactions:
  - $CO + OH \leftrightarrow CO_2 + H$
  - $H_2 + OH \leftrightarrow H_2O + H$
  - i.e., *AR* should have higher OH than *AN* case

- Let's look at 2D maps of T, OH, C_2H_2, SVF, ΔOH, ΔT, ΔC_2H_2, etc…

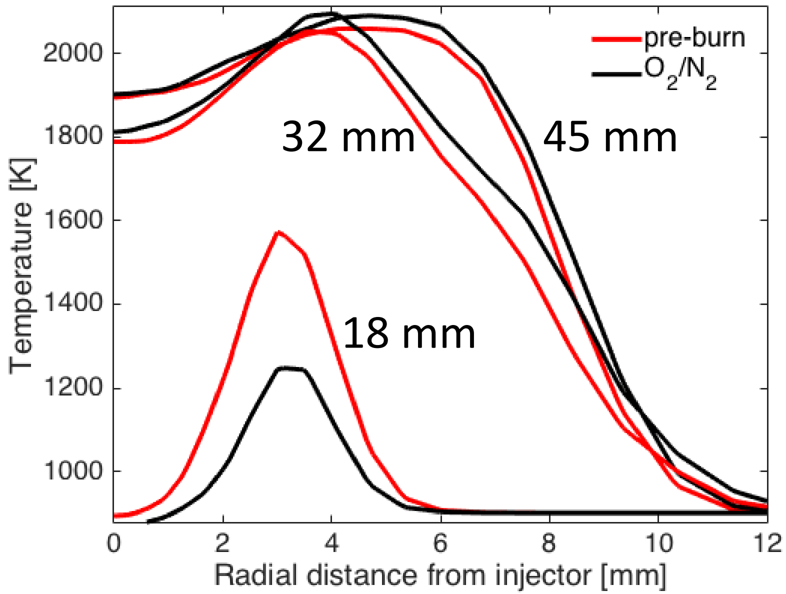
What can we learn by comparing the AN and AR cases?

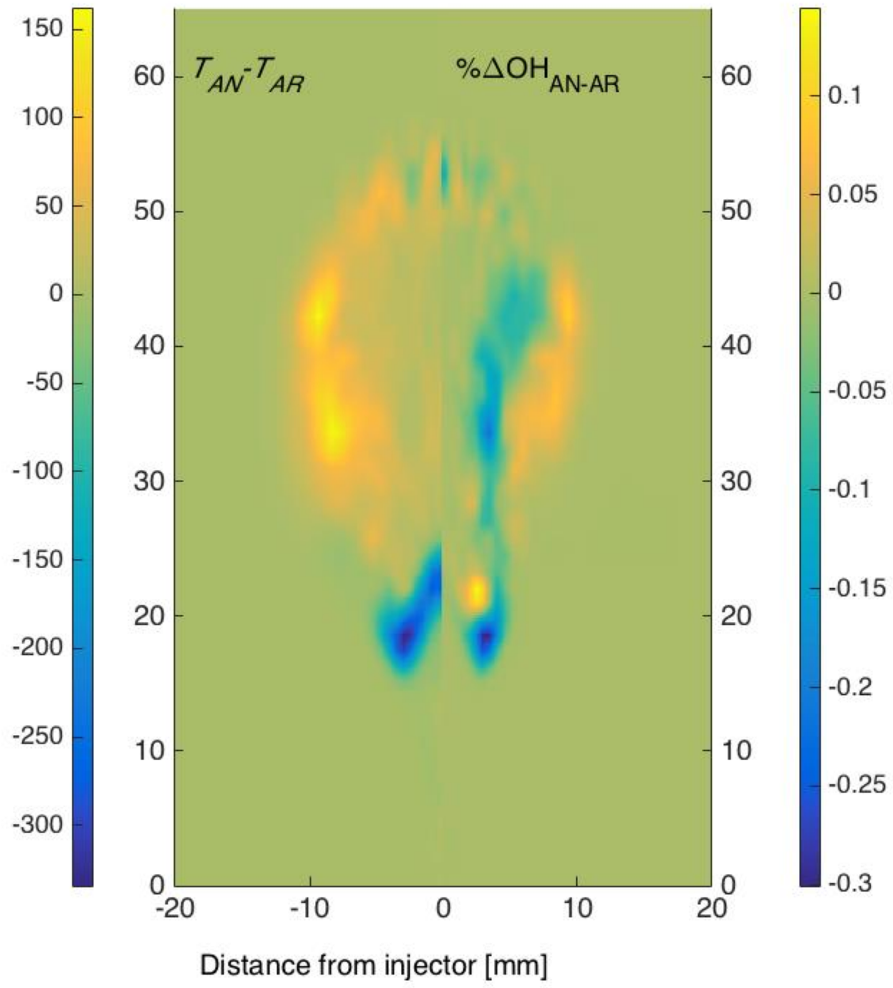
- UNSW
  - Comparing T maps, AR case may be expected to have slightly shorter lift-off length based on temperature map
    - LOL
      - AR: 16.1 mm
      - AN: 15.7 mm
  - Otherwise, differences are difficult to distinguish unless we look at ΔT



UNSW tPDF results show some larger temperature differences but less in OH

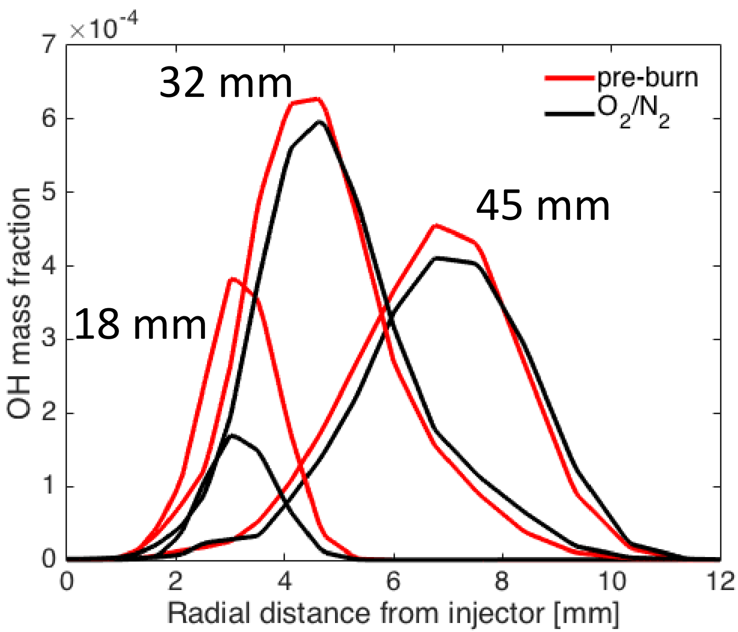
- UNSW-tPDF
  - AN and AR case show significant differences in lift-off region
  - Are impacts observed in global analysis of LOL?
  - Does higher temperature in lift-off region lead to higher soot? NO!
  - Do OH increases follow temperature?

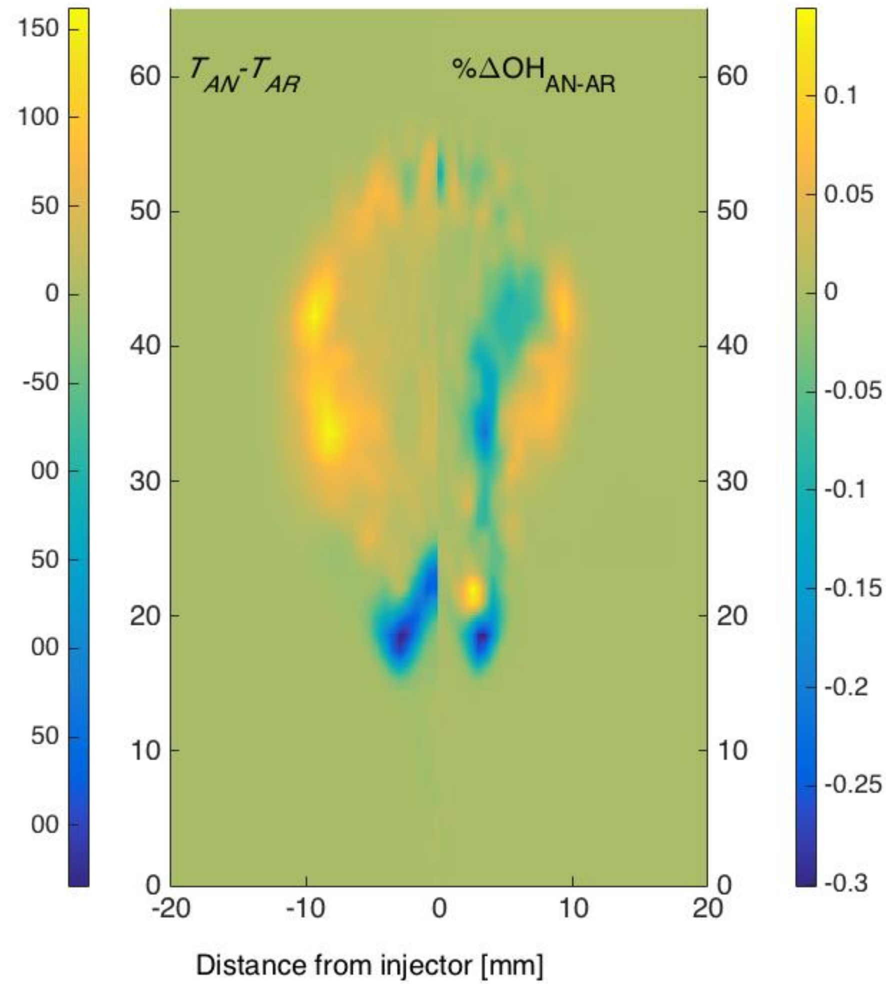




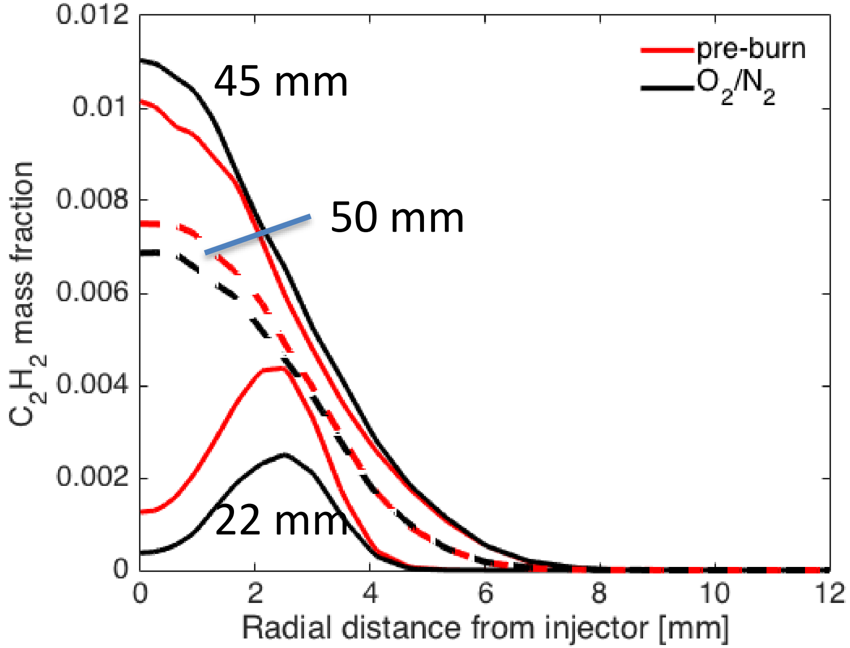
UNSW tPDF results show some larger temperature differences but less in OH

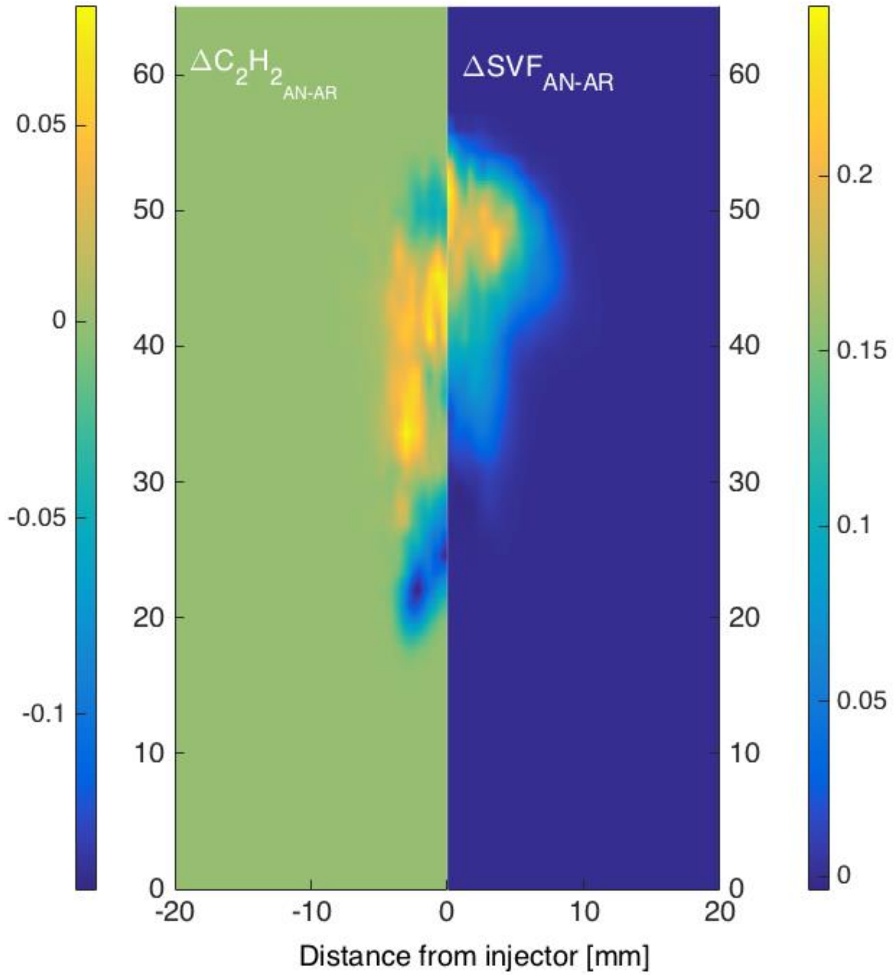
- UNSW-tPDF
  - OH profiles do not show large differences except in lift-off region
  - Higher OH in AR case expected based on higher H_2O in ambient…
  - H_2+OH <-> H_2O+H (H_2O drives reverse rxn)
  - CO+OH <-> CO_2+H (CO_2 drives reverse rxn)
  - OH differences not directly associated with T…





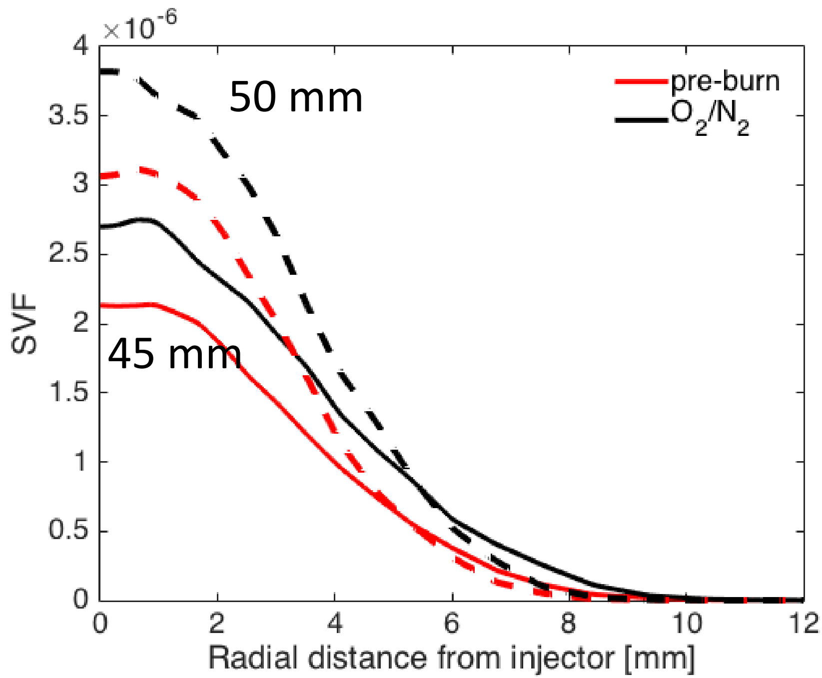
UNSW AN case shows ~5% increase in C_2H_2 and 20% bump in SVF

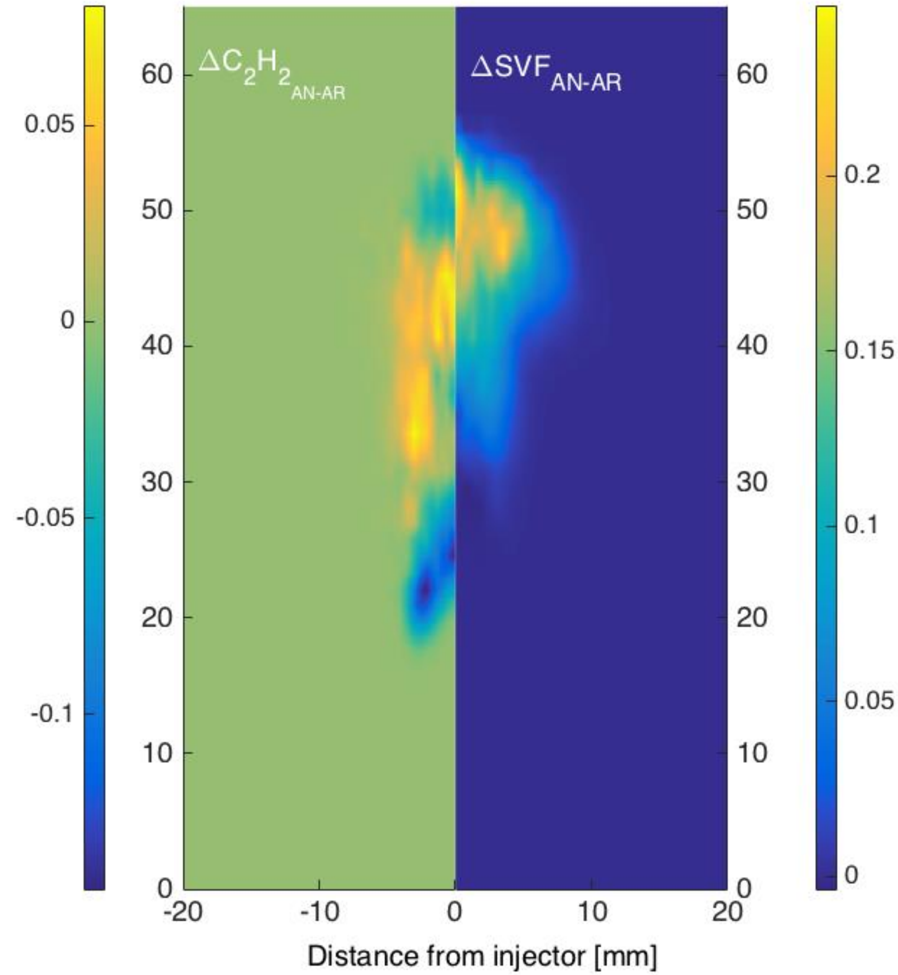




…we saw more OH in the AR case upstream, but we're also seeing more acetylene! Even so, the SVF is lower for AR!

UNSW AN case shows ~5% increase in C_2H_2 and 20% bump in SVF

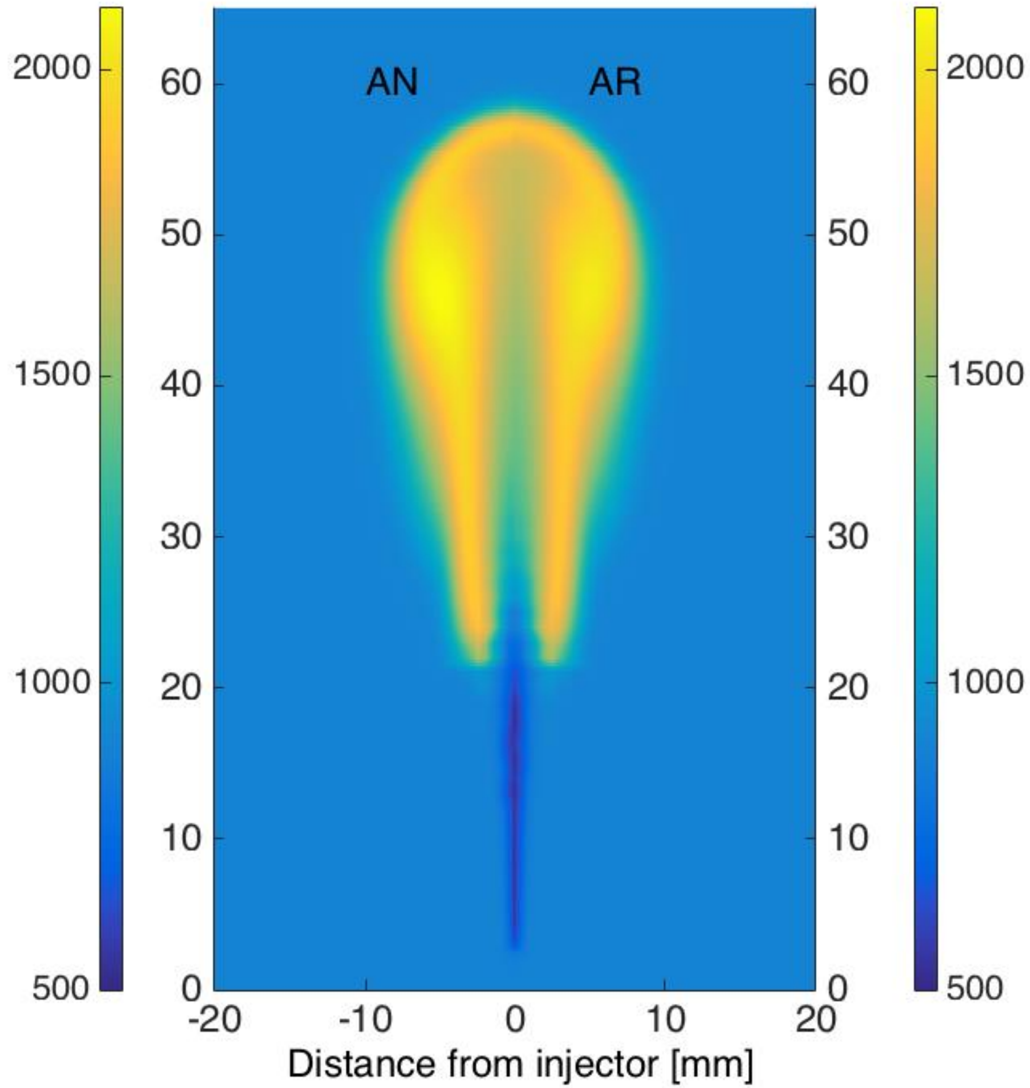




…At 45 mm we saw more acetylene in the AN case and we're also seeing more SVF. However, at 50 mm we see less acetylene in the AN case but still more SVF…

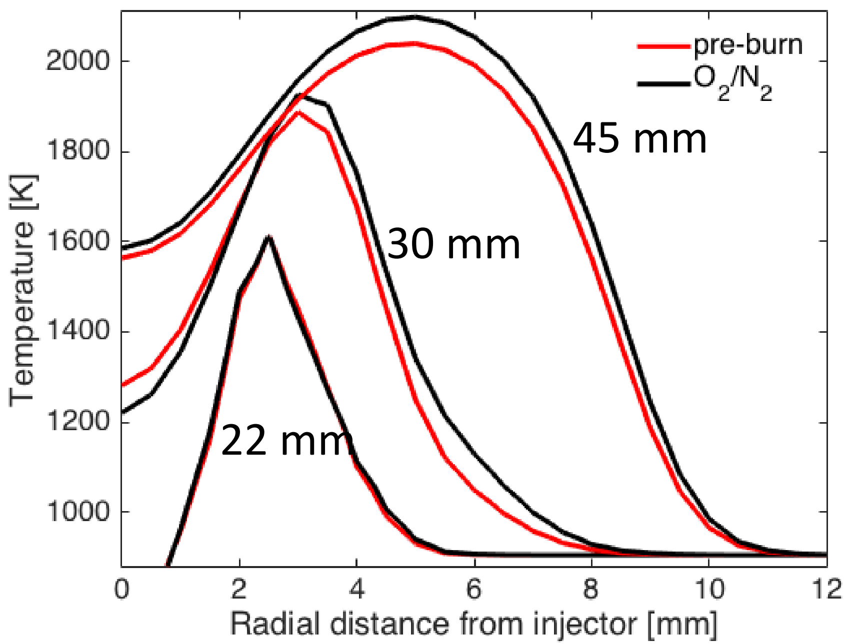
ETHZ results are contrary to those of UNSW…

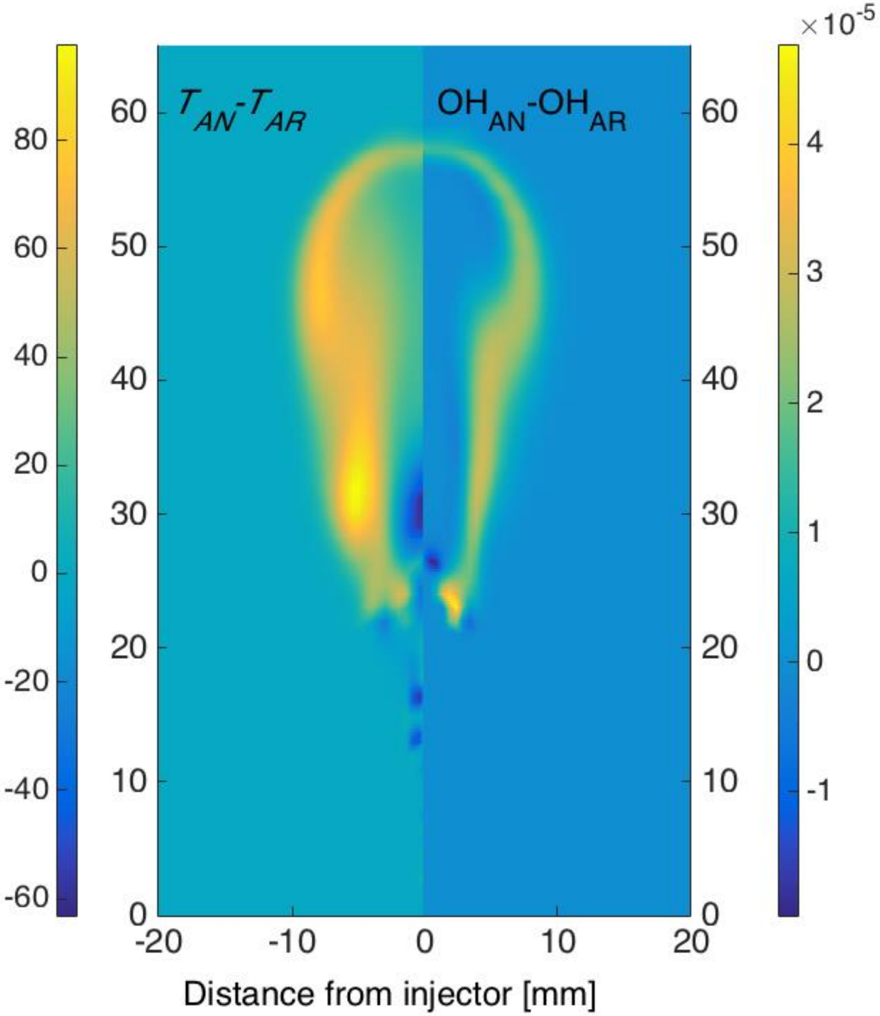
- ETHZ
  - Contrary to UNSW result
    - temperature in lift-off region appears similar for AR and AN cases
    - Liquid and vapor region are colder than UNSW results
  - Similar to UNSW result
    - AN case shows some HIGHER temperatures downstream



ETHZ T and OH results are contrary to those of UNSW…

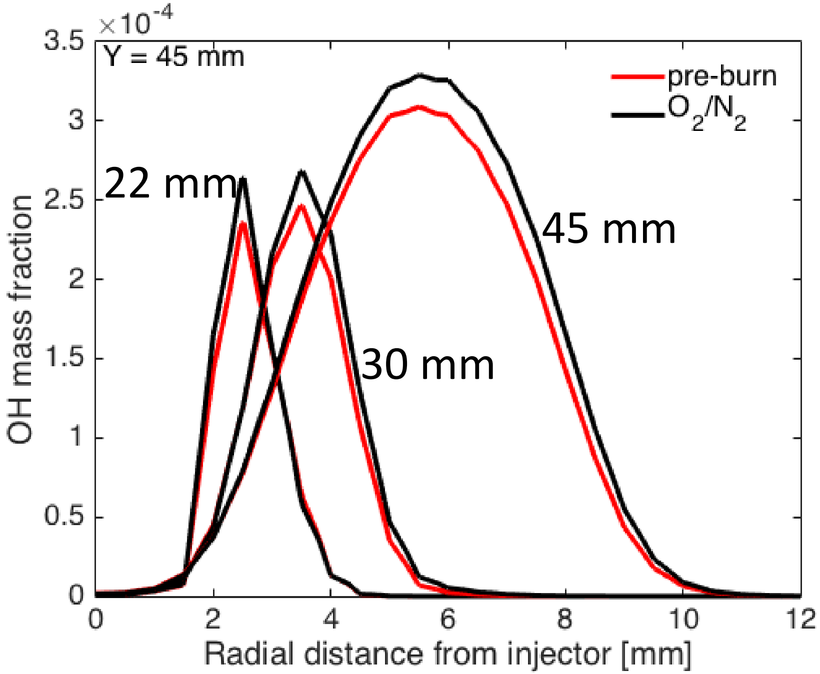
- ETHZ
  - AN case shows up to 100 K higher temperatures 1.5 ms ASOI
  - <u>AN case has regions with higher OH than AR case</u>
    - H_2+OH <--> H_2O+H ?!?

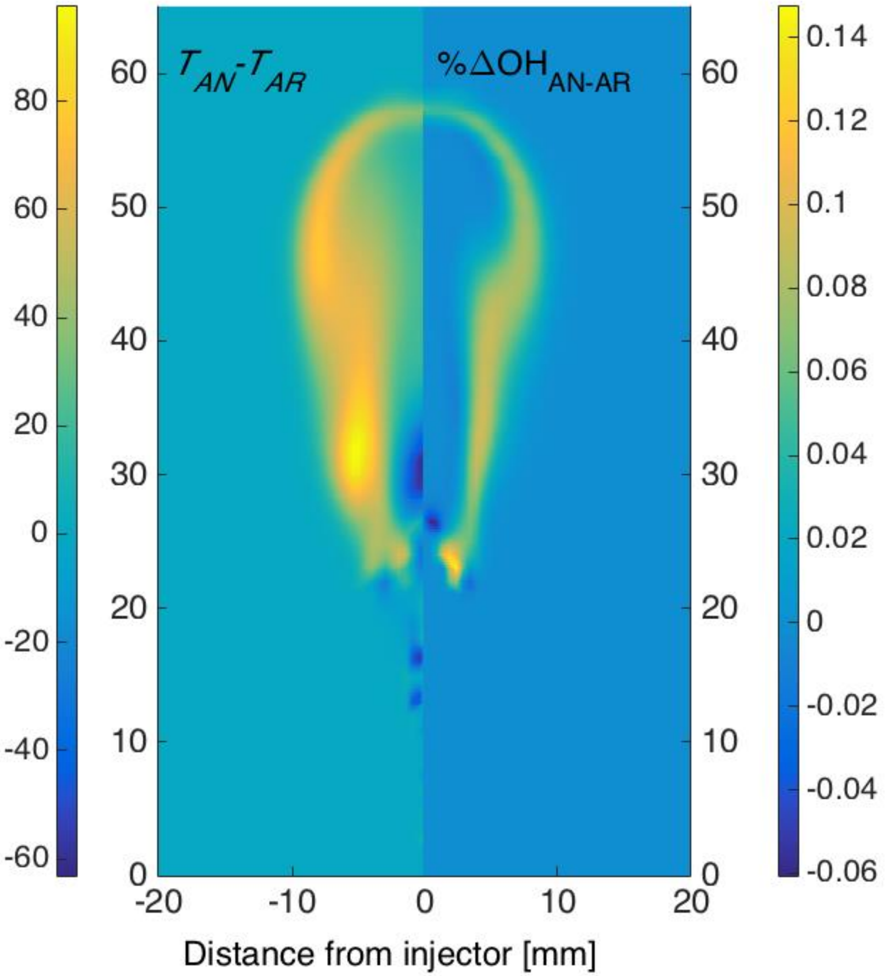




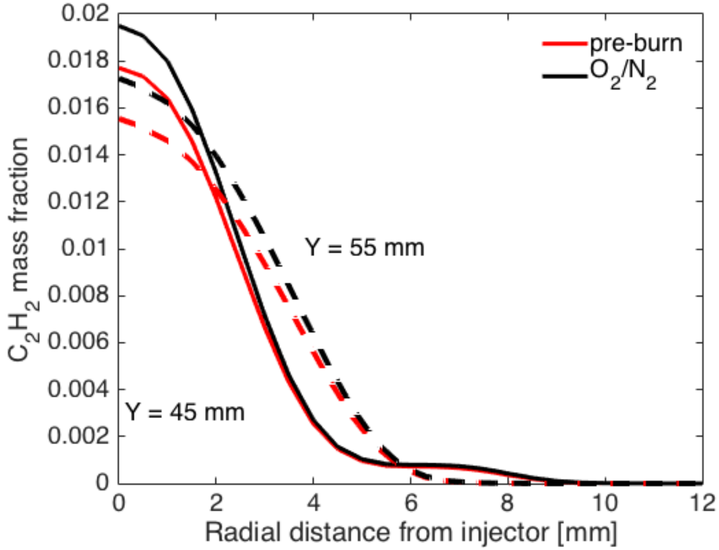
ETHZ OH and T results are contrary to those of UNSW…

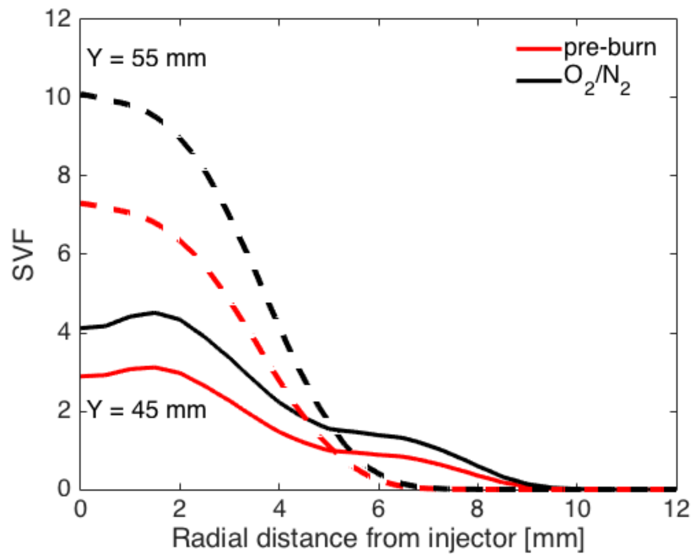
- ETHZ
  - Differences in OH are are quite small, but follow region of higher AN temperature
  - Perhaps H,C,O chemistry or thermo needs to be corrected…

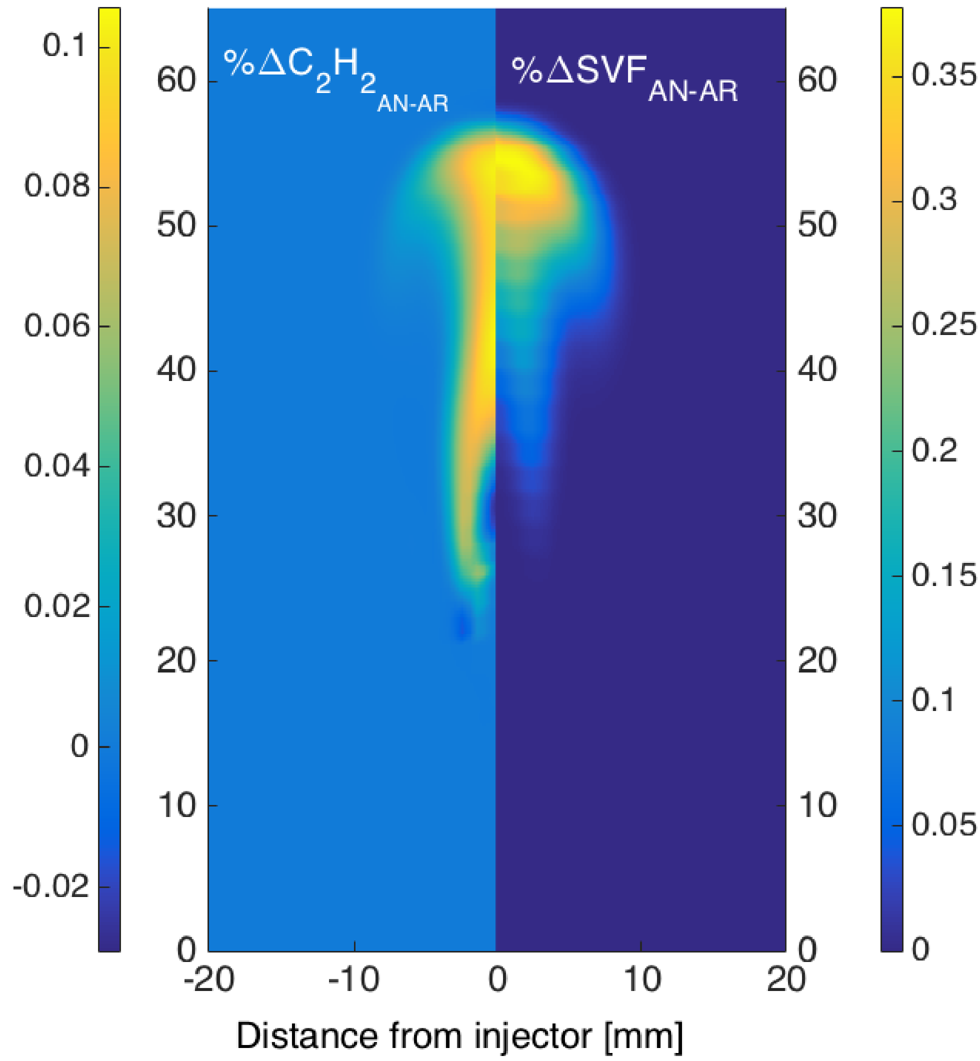




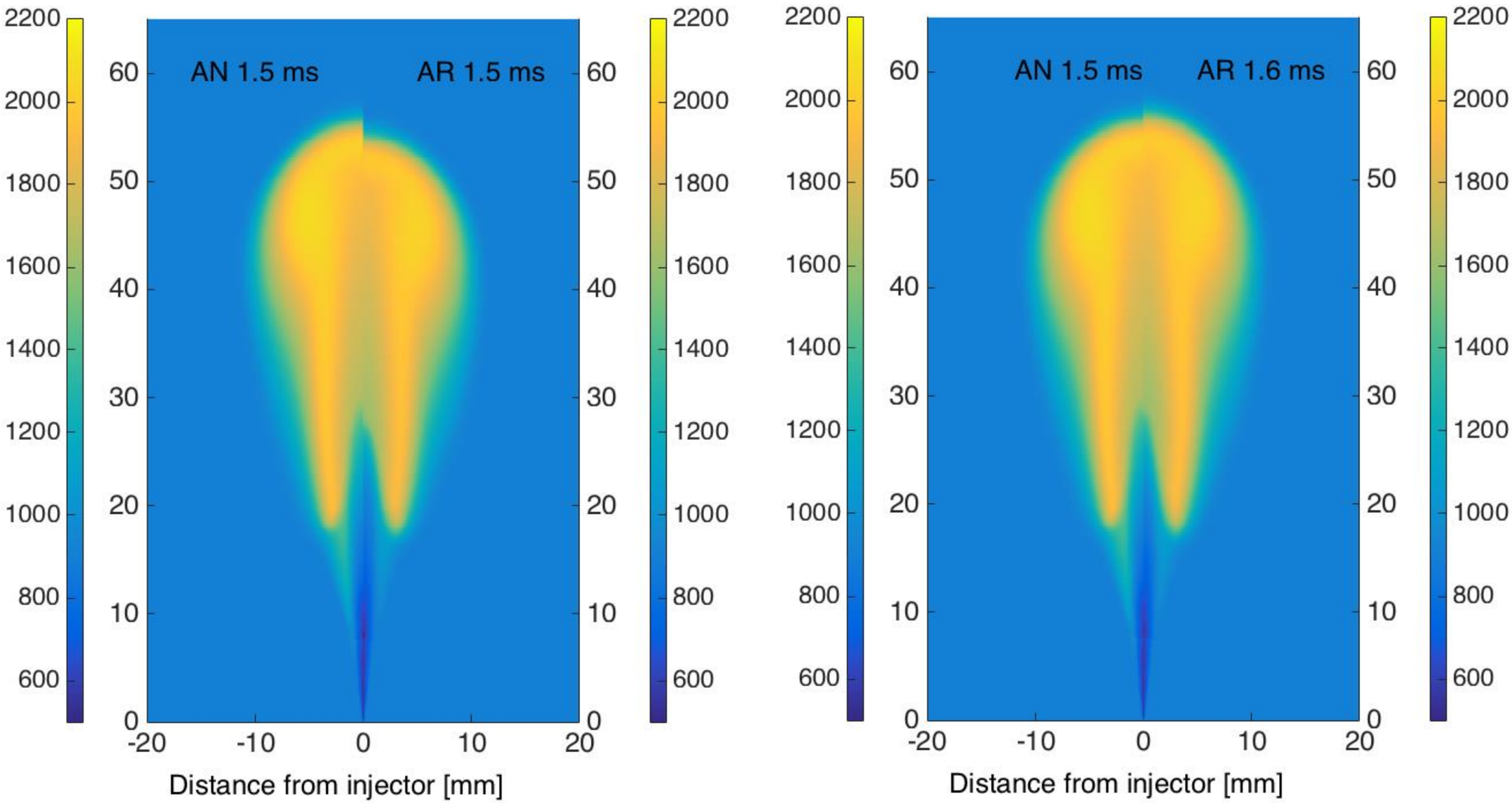
ETHZ “*AN*” case shows higher C_2H_2 and SVF—larger % increase in SVF





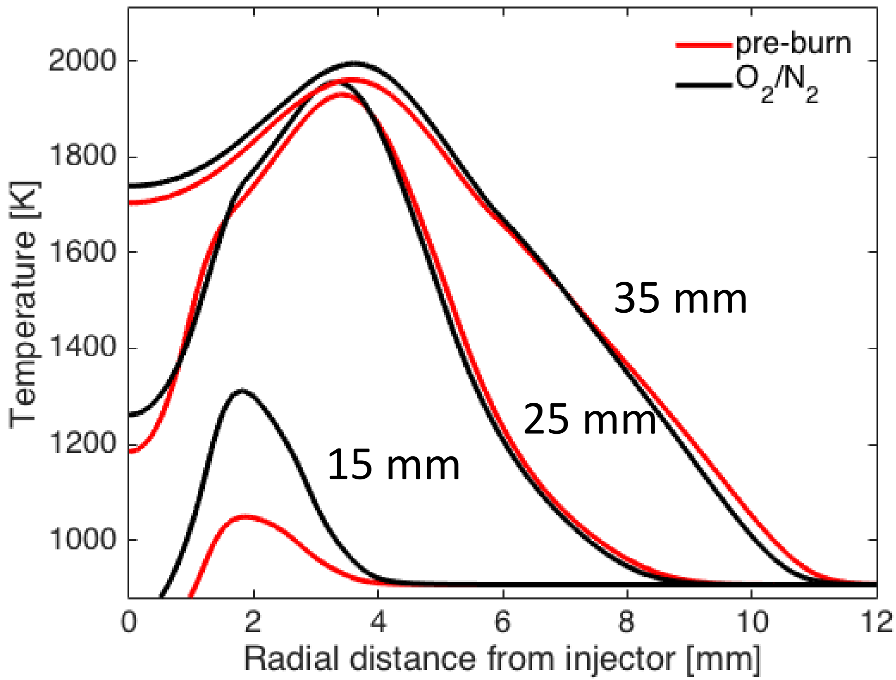


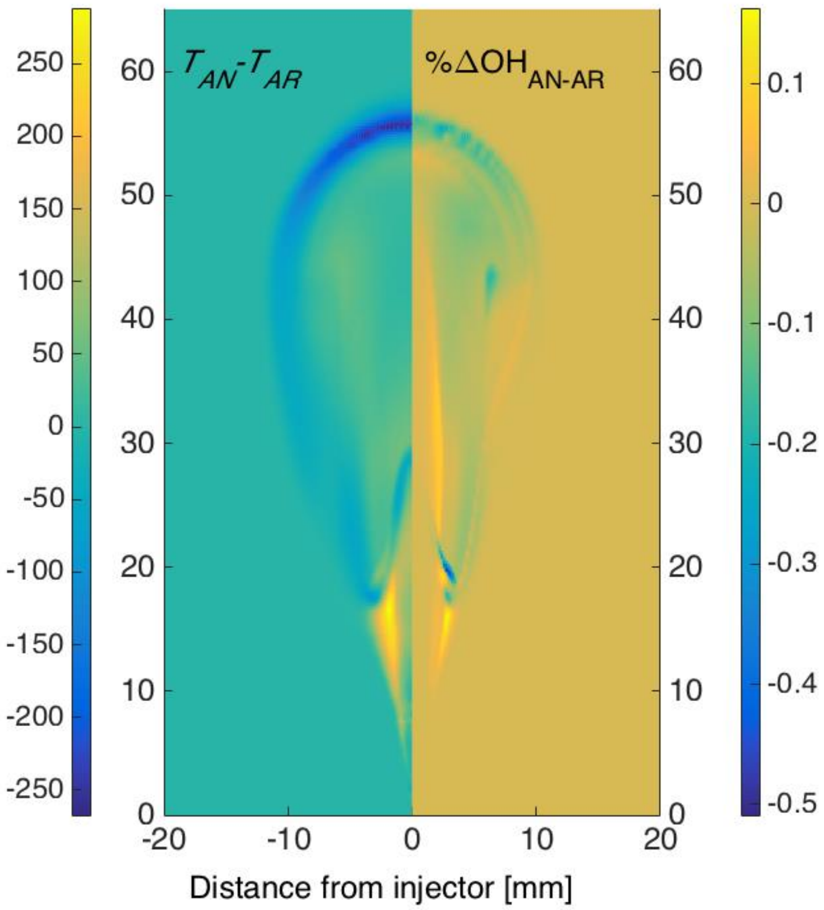
POLIMI T results indicate different penetration for AR and AN cases…



POLIMI AN case yields higher temperatures—esp. in lift-off region

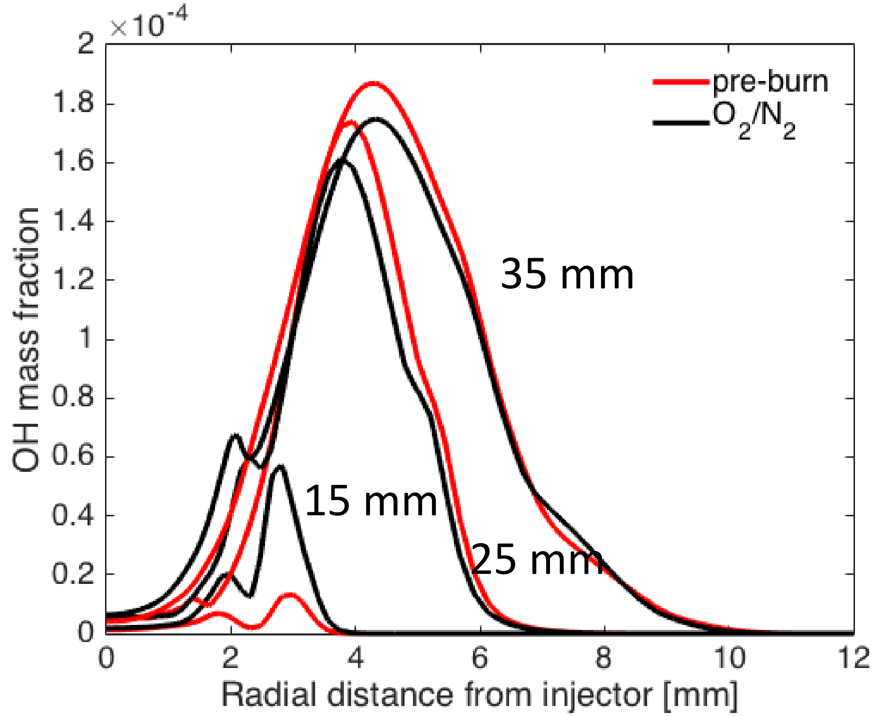
- POLIMI
  - Neglecting head region due to different penetration, AN and AR case have similar temperature fields 1.5 ms with biggest differences in lift-off region

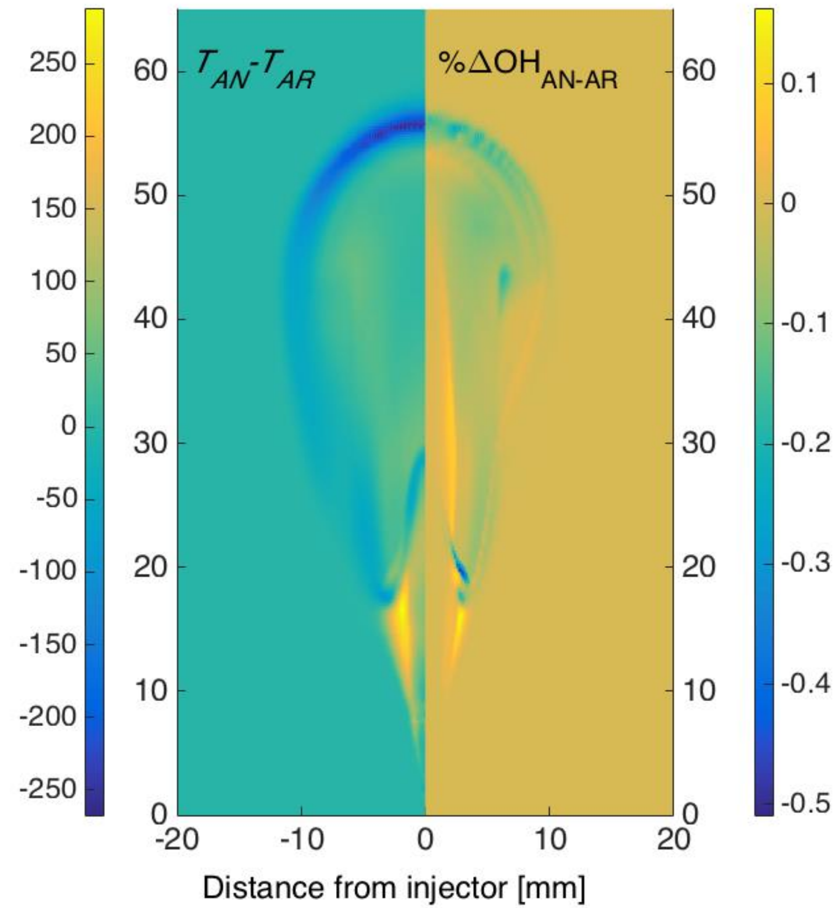




OH higher in pre-burn case, except in lift-off region…

- POLIMI
  - AR (pre-burn) shows slightly higher OH except in lift-off region
  - Lift-off region differs from UNSW result – higher T and OH in O_2/N_2 (AN) case…





Outlook toward ECN6

- We have a lot to consider and need to get started earlier…
- Paper plans:
  - Joint IFPEN/CAT/SNL Spray C/D Experimental paper
  - Potential Spray A pre-burn vs. O2/N2 paper with modeling
    - Perhaps need to do some work on spray models and mixing…
    - What is Penn State doing right that's capturing appropriate total soot mass ramp up?
- Can we spend the time and close the book on Spray A soot modeling and begin C/D soot modeling?
- New Experimental ID, LOL, and Soot Data:
  - N-heptane with n370 injector
    - How much does the soot model matter?
  - Certification diesel fuel and four CRC surrogates
- Develop a long term vision for the Soot Topic
- New ECN Graphic/Logo…