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Towards Consistent and Reliable ISF Datasets

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Why This is Important

- Soot models are phenomenological – NOT based on first principles; model predictions are essentially calibrated against reported experimental results
- Measurements in sooty flames tend to be more challenging than measurements in soot-free environments (i.e. measurements can have significant errors/uncertainty)
- Soot itself is not a well-defined quantity, either experimentally or in modeling; it is important to have *consistent* and *well-understood* definitions of what we are calling ‘soot’ for purposes of comparing datasets and/or model results
  - Models often define pyrene-dimers as soot, whereas experimentalists distinguish
    - brown vs. black soot
    - organic vs. graphitic carbon
    - 450 nm absorbing vs. 1064 nm absorbing, etc.
    - SMPS signals vs broadband emission/absorption
  - Particle size can mean lots of different things (d_p, R_g, D_{63}, spherical-equiv) – important for calculated soot growth and oxidation rates

Treatment of Data Reliability in ISF To-Date

- One of stated aims of ISF is

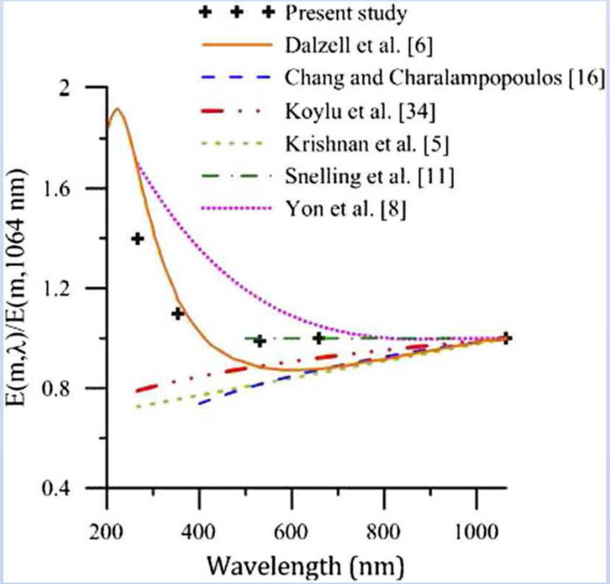
  "To establish an archive of the detailed data sets of target flames *with defined accuracy*"

- There have been periodic discussions amongst ISF Organizing Committee and Scientific Advisory Committee about instituting a consistent assumed K_a/K_e (for example) in reporting soot concentrations in ISF target flames

- ISF-3 program (2016) included presentation on "Soot Data Uncertainty and Standardisation" (Shaddix, Geigle, Gulder, Nathan)
  - f_v measurements: recommend $K_a = 7.5 \pm 0.5$, $K_e = 9.0 \pm 1.0$ (agglomerated, mature soot)
  - LII: primary uncertainty due to calibration (40% error); 15% error shot-to-shot (fluence/trapping); 5% error in most other considerations
  - PIV measurements: uncertainties depend on velocity gradients and soot conc.; typ. ± 1 – 5 m/s
  - CARS measurements: T uncertainty of 5% instantaneous, 2% mean
  - Pyrometry: uncertainties stem from assumed spectral emissivities and signal trapping
  - TLAF

Considerations for Soot f_v Quantification

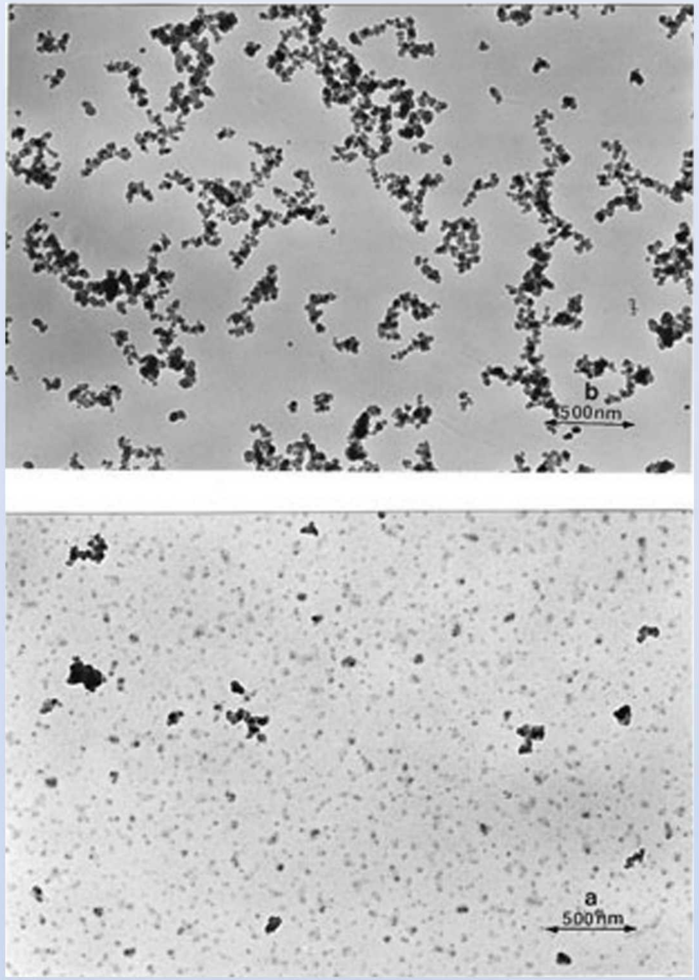
- High variability in *presumed* K_a/K_e values to quantify soot concentration, with older index of refraction measurements yielding low predicted K_a (overpredicting f_v)
- Technical literature has converged on a defensible K_a value for mature soot, for wavelengths from 500 – 1100 nm (0.35-0.40)
- Agglomeration of soot primary particles leads to $K_e > K_a$ (by up to 40%, for highly agglomerated particles)
- For wavelengths < 500 nm, K_a appears to increase, even for mature soot
  - Bejaoui et al. (2014, Lille) spectral study of LII excitation



Key Consideration for Soot f_v Quantification for ISF

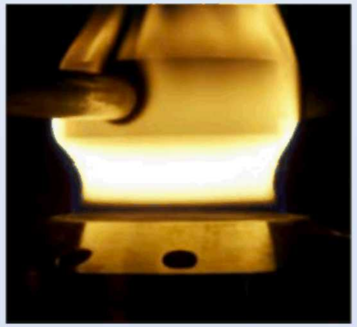
During active particle inception, K_a is initially very low (transparent drops/particles) and then increases to its value for mature soot as particles carbonize

- Implies that not only the soot concentration *magnitude*, but ***the shape of curve for soot formation rate***, is significantly different than presented in existing data
- This effect is particularly sensitive to wavelength of extinction/LII excitation, with long wavelengths taking longer to reach significant absorptivity
  - Long wavelengths are preferred for extinction measurements to minimize contamination from PAH absorption and to minimize contribution of light scattering
  - Long wavelengths preferred for LII to be immune from excitation of PAH/C_2 fluorescence



Dobbins, AST 2007
(from centerline of Santoro flame)

An Example: ISF-4 Laminar Premixed Flame #3



60-mm McKenna Burner, ethylene/air, ϕ = 2.1 at 10 SLPM

- f_v data provided by
  - B. Axelsson, R. Collin, P.-E. Bengtsson, Appl. Opt. 39 (2000) 3683-3690.
    - Extinction (and LII) measurements performed with pulsed 532 nm beam on gated ICCD
  - J. Zerbs, K.P. Geigle, O. Lammel, J. Hader, R. Stirn, R. Hadef, W. Meier, Appl. Phys. B 96 (2009) 683-694.
    - Extinction measurements performed with cw 532 nm and 1064 nm diode lasers and 633 nm cw HeNe laser; 532 nm and 633 nm beams chopped and 1064 nm beam modulated; all colors used lock-in detection on a photodiode.
  - R. Hadef, K.P. Geigle, W. Meier, M. Aigner, Int. J. Thermal Sci. 49 (2010) 1457-1467.
    - 2-D LII measurements with 1064 nm excitation as calibrated by 2-D integrated pulsed extinction at 532 nm on ICCD

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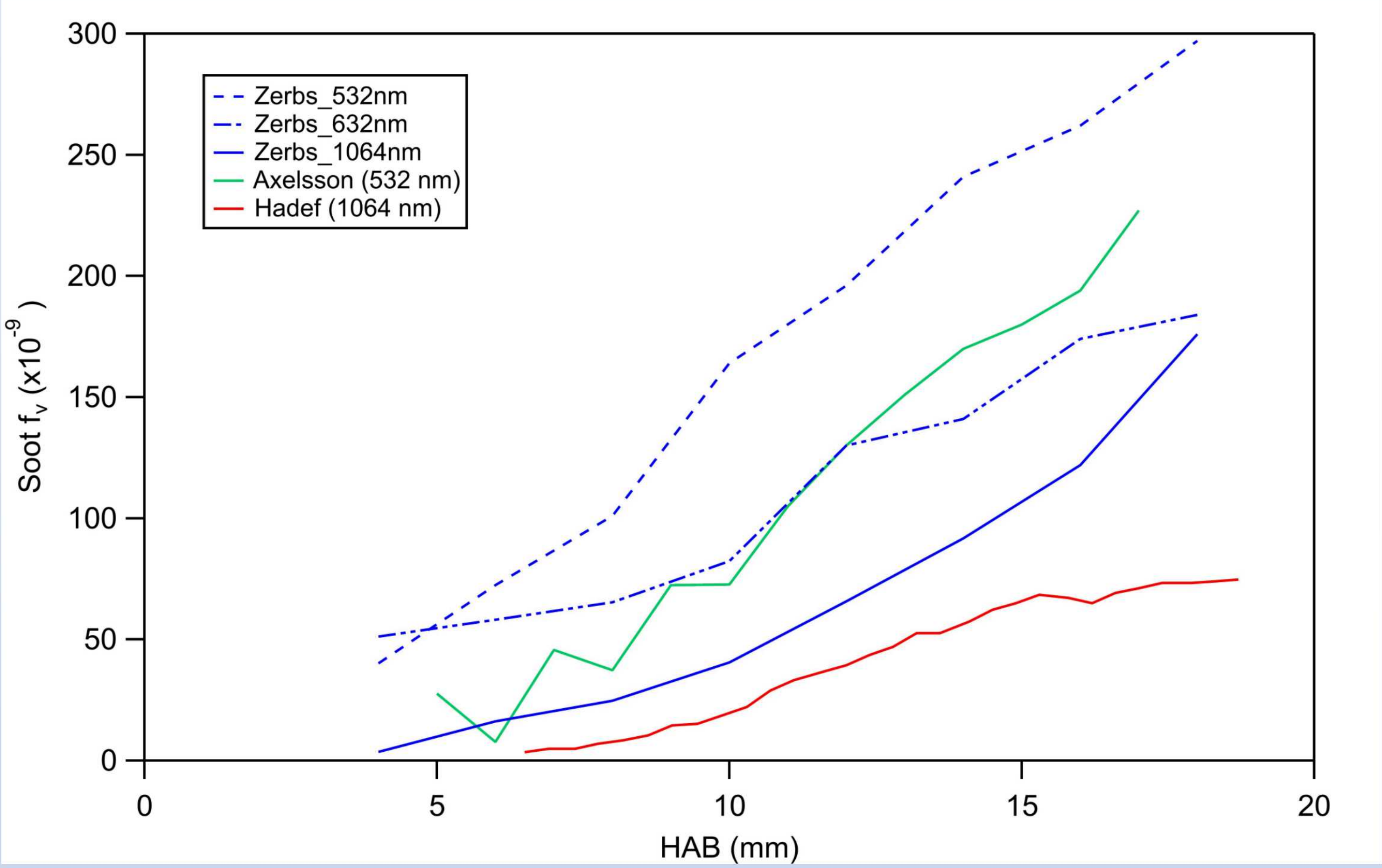
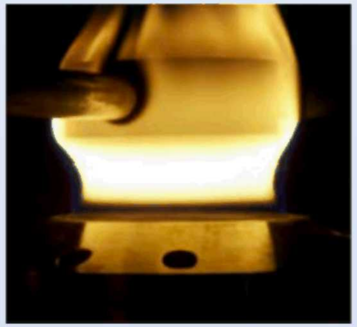
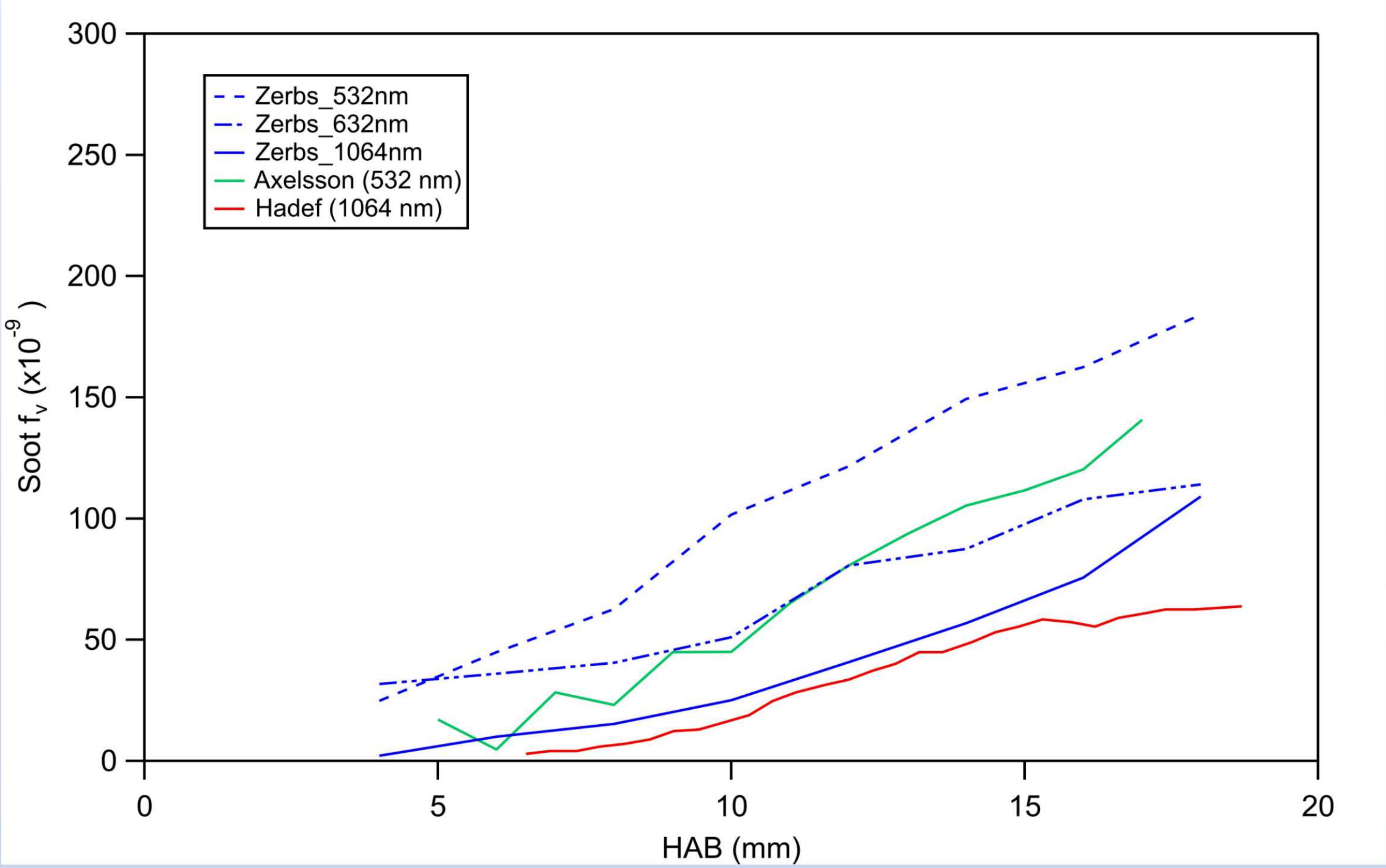
As-reported data

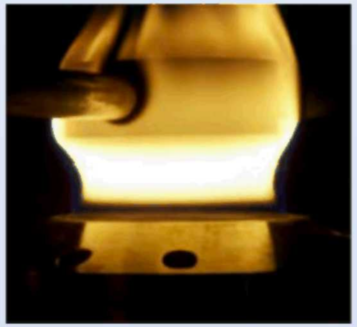




An Example: ISF-4 Laminar Premixed Flame #3

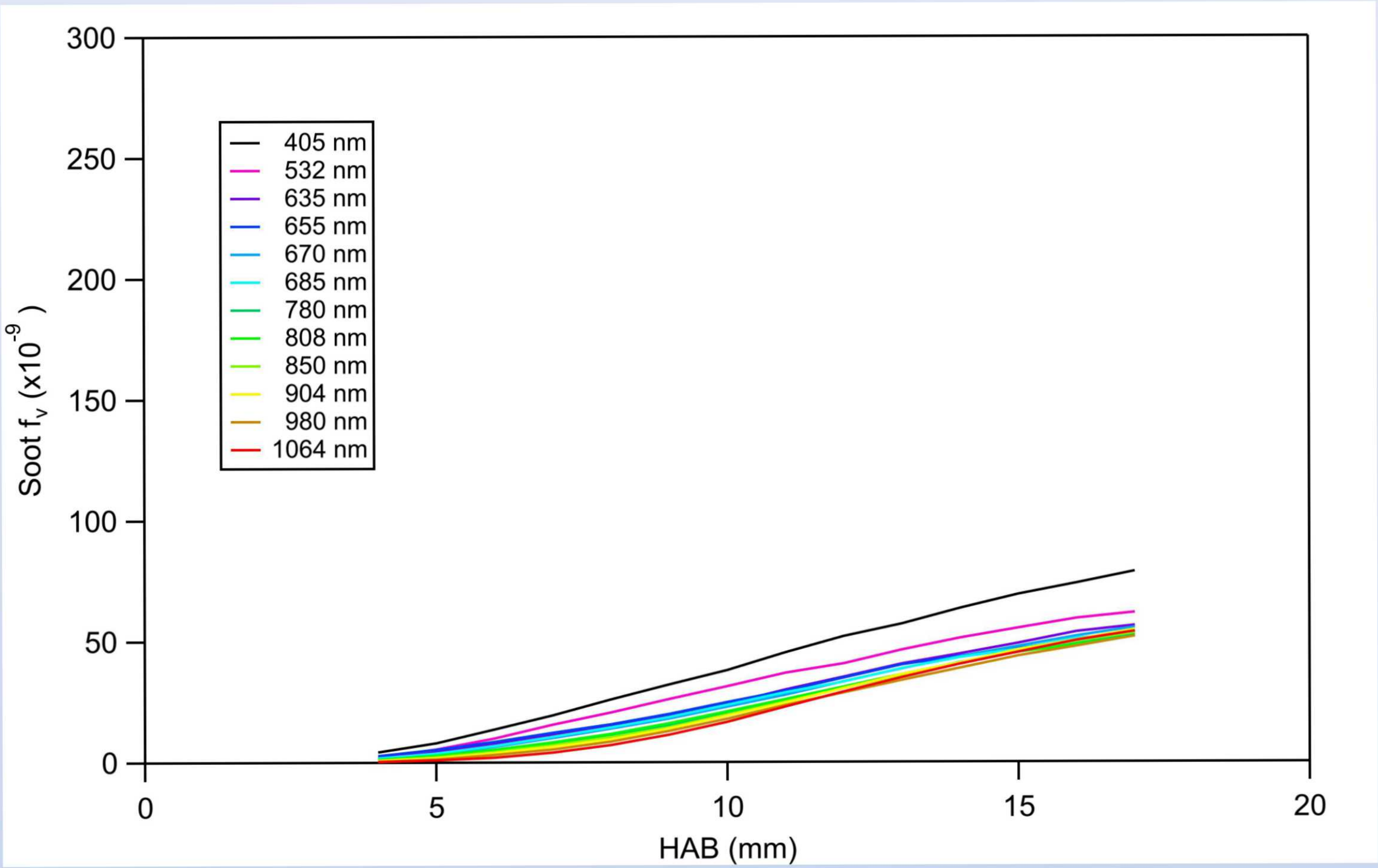
Standardised data: E(m) = 0.35

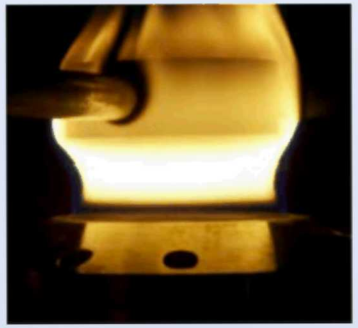




An Example: ISF-4 Laminar Premixed Flame #3

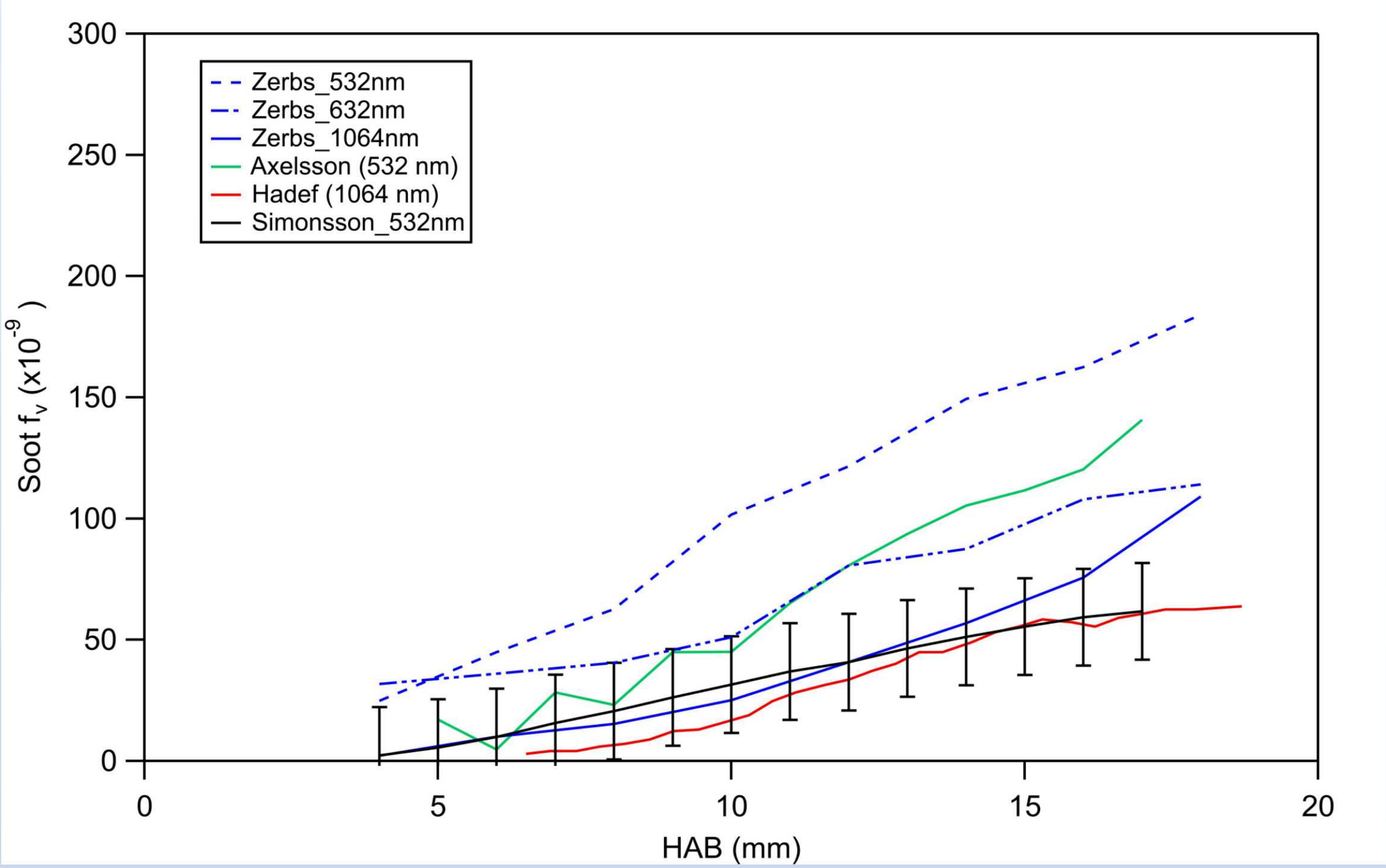
'New' data: Simonsson et al., Appl. Phys. B 2015, E(m) = 0.35

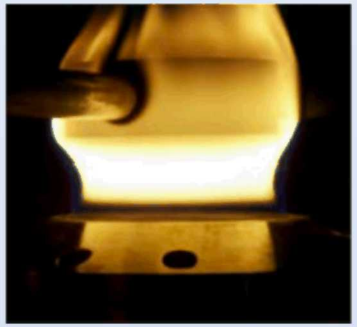




An Example: ISF-4 Laminar Premixed Flame #3

My best recommendation, for E(m) = 0.35





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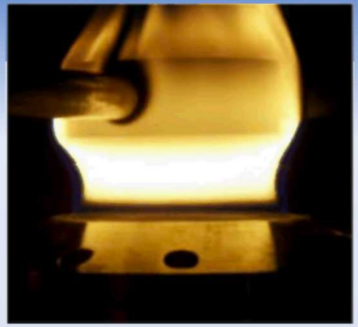

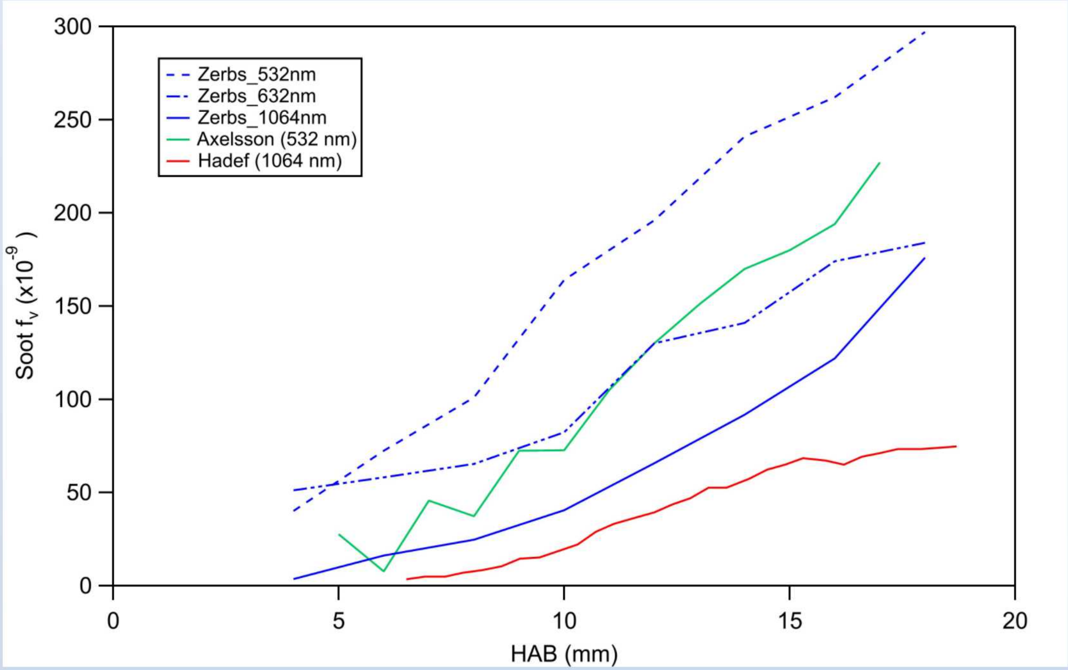

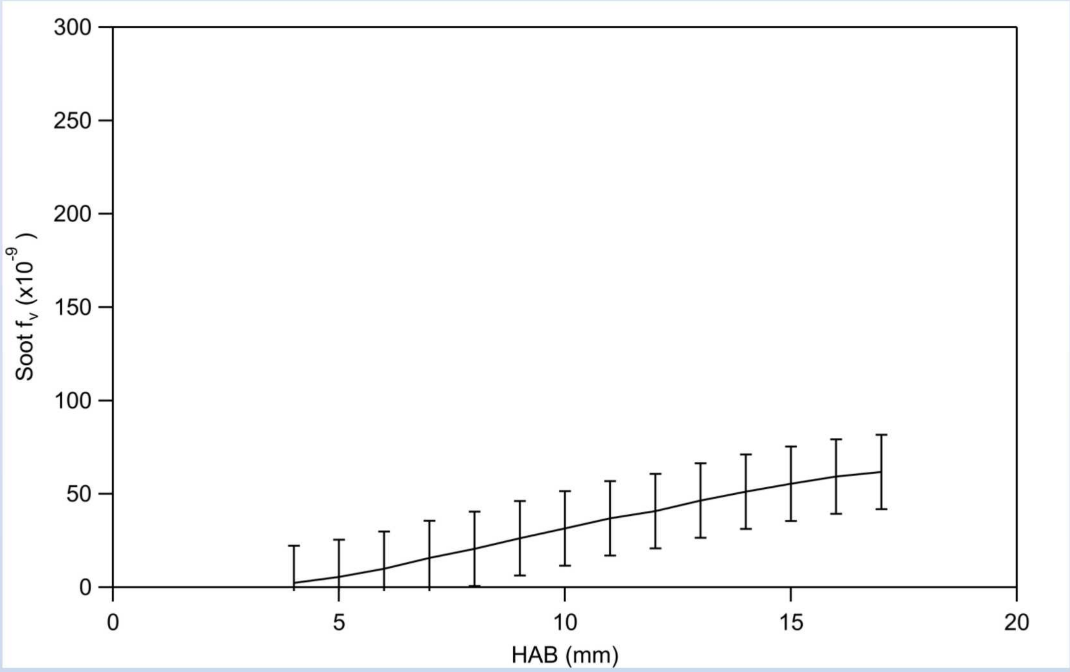

E(m) of mature soot

- Kempema et al. Appl. Phys. B 2016

Table 2 Soot absorption functions from in-flame measurements in laminar coflow diffusion flames

	$E(m)$	λ (nm)	Diagnostic	Burner	Fuel
Snelling et al. [25]	0.4	532	LII/LII modeling	Gülder burner	Ethylene
Snelling et al. [44]	0.45 ± 0.04	465	Modulated LII	Gülder burner	Ethylene
	0.45 ± 0.03	577			
	0.42 ± 0.02	865			
This work					
2 cm HAB	0.38 ± 0.05	532	TSPD/spec-LOSA	Yale burner	Ethylene
3.5 cm HAB	0.43 ± 0.05	532			
6 cm HAB	0.42 ± 0.05	532			
Williams et al. [19]	0.41–0.44	635	In-flame GSLE	Similar to Santoro burner	Ethylene
Williams et al. [19]	0.40–0.44	635	In-flame GSLE	Similar to Santoro burner	Kerosene
Williams et al. [19]	0.34	635	In-flame GSLE	Similar to Santoro burner	Methane
This work					
2 cm HAB	0.36 ± 0.05	635	TSPD/spec-LOSA	Yale burner	Ethylene
3.5 cm HAB	0.42 ± 0.05	635			
6 cm HAB	0.40 ± 0.05	635			