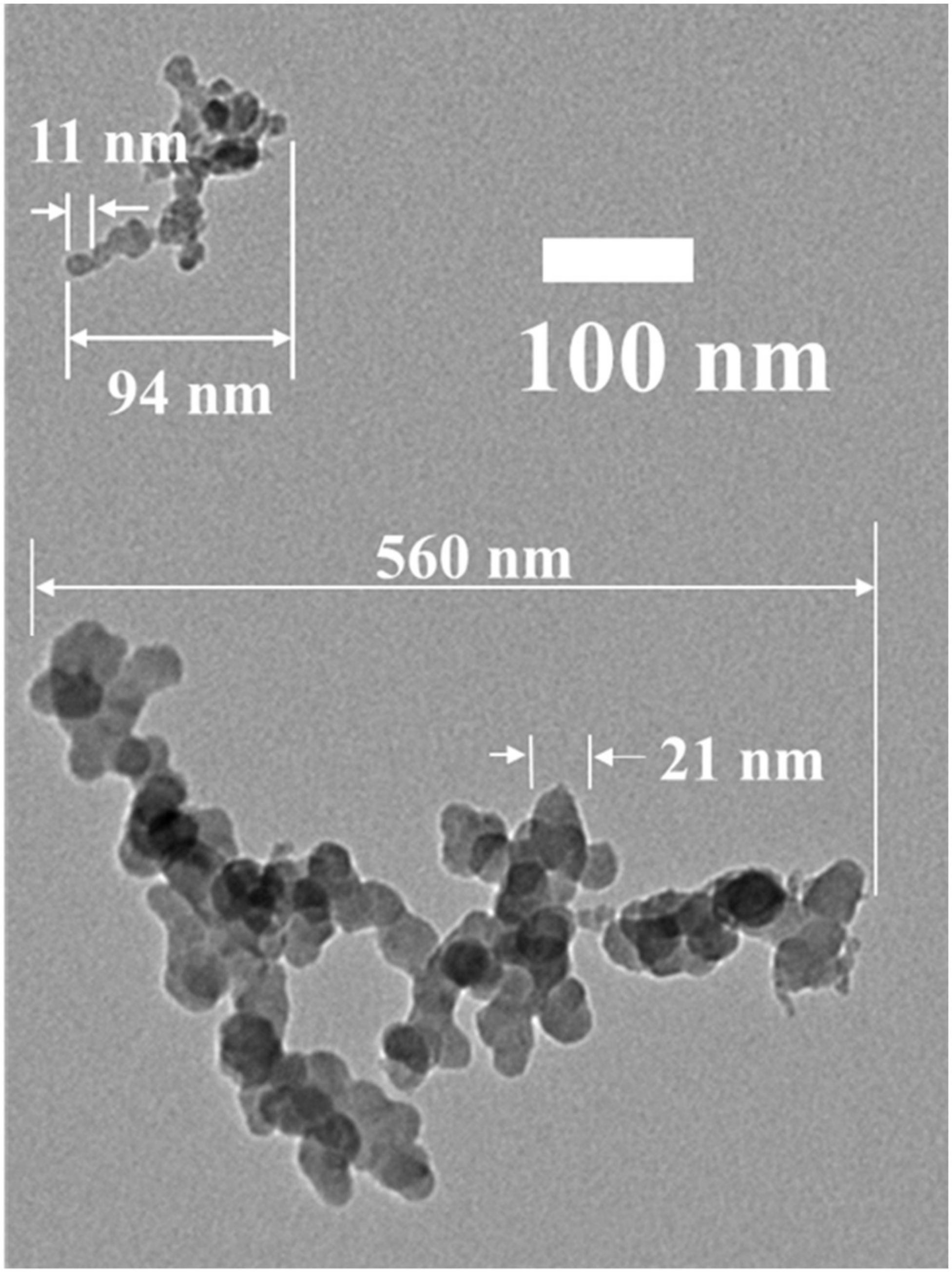
THE EXTERNAL MIXING HYPOTHESIS FOR SOOT STRUCTURE: PART 1. SUPPORTING EVIDENCE PART 2. WHERE IT LEADS

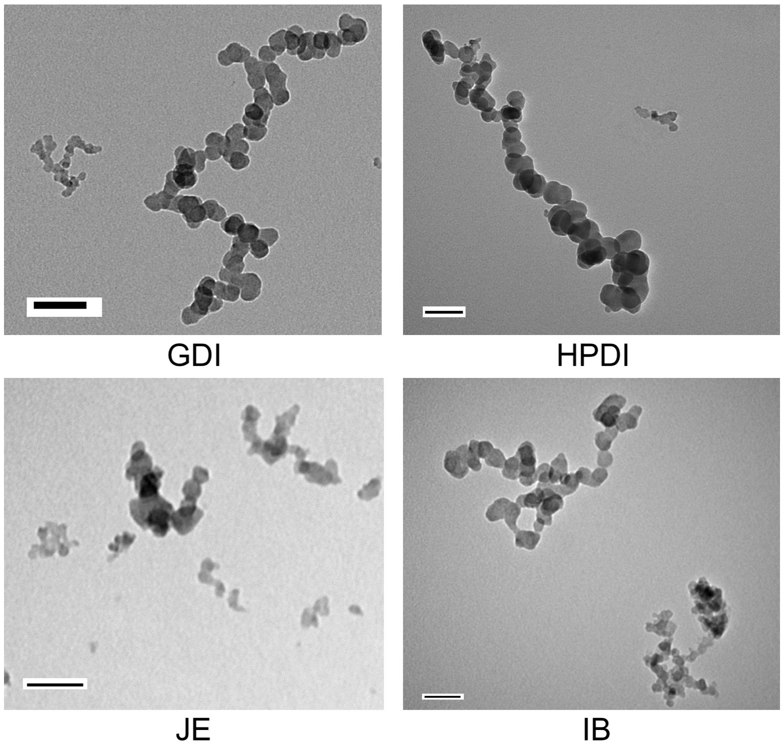
STEVE ROGAK, UNIVERSITY OF BRITISH COLUMBIA

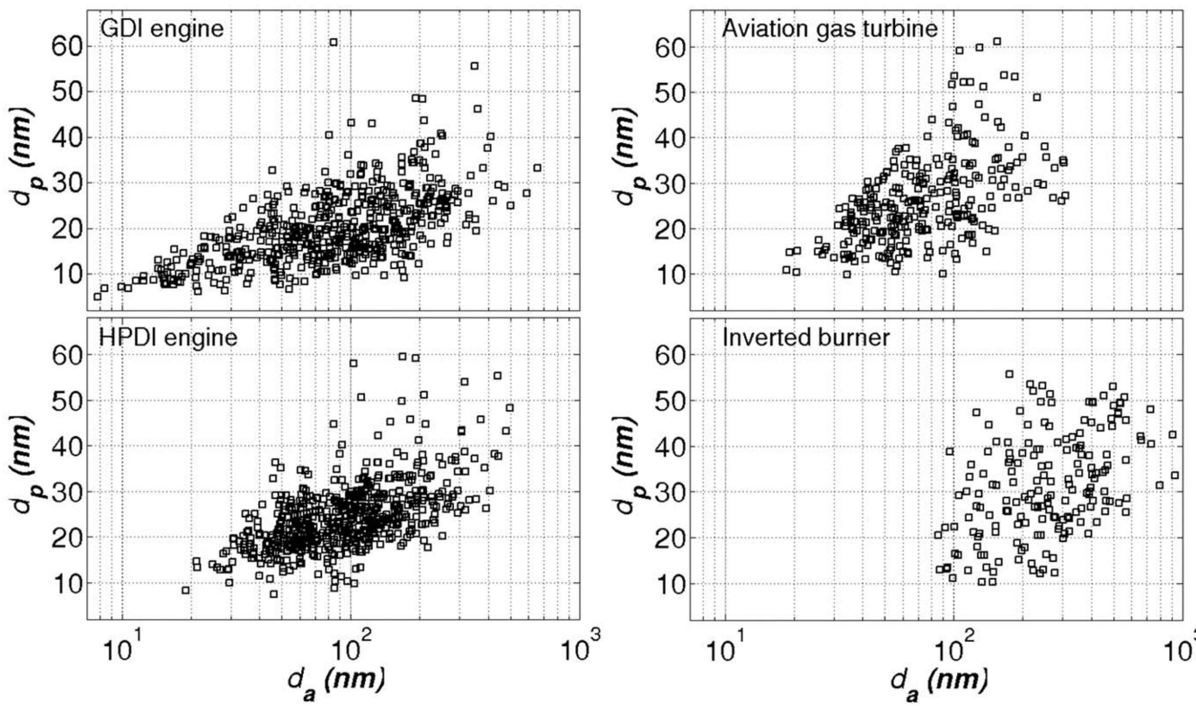
T. SIPKENS, R. DASTANPOUR, A. BALDELLI, J.S. OLFERT, U. TRIVANOVIC, M, KAZEMIMANESH, J. CORBIN, S. GAGNE, A. BOIES AND OTHERS!

PART 1. EXTERNAL MIXING HYPOTHESIS



TEM OBSERVATIONS → EXTERNAL MIXING HYPOTHESIS



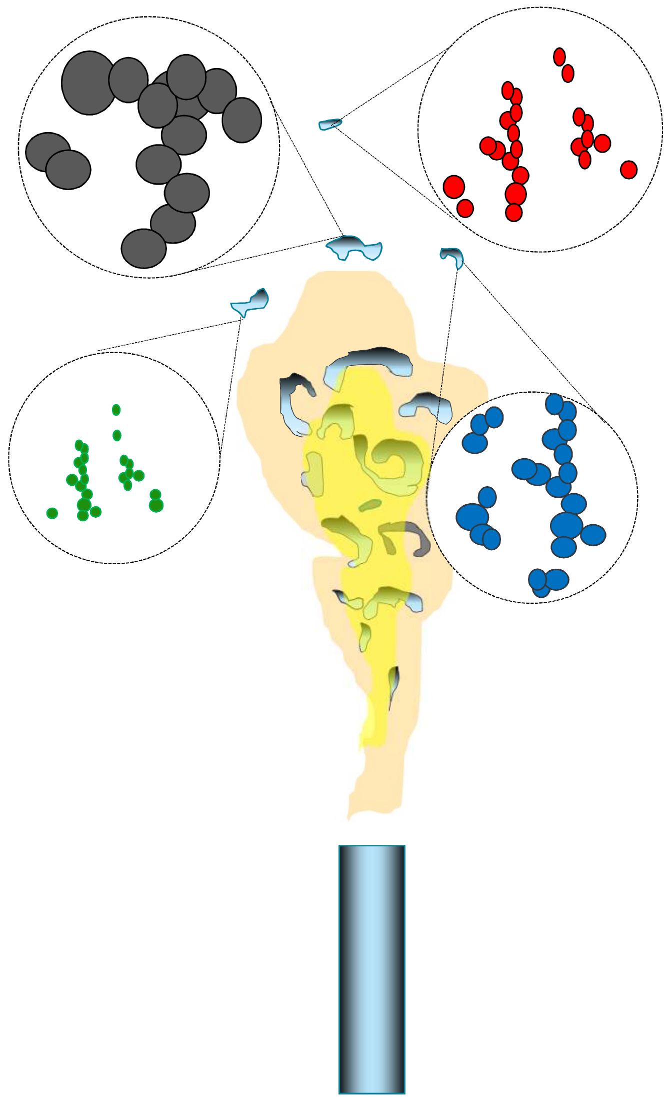


Scale Bars 100nm

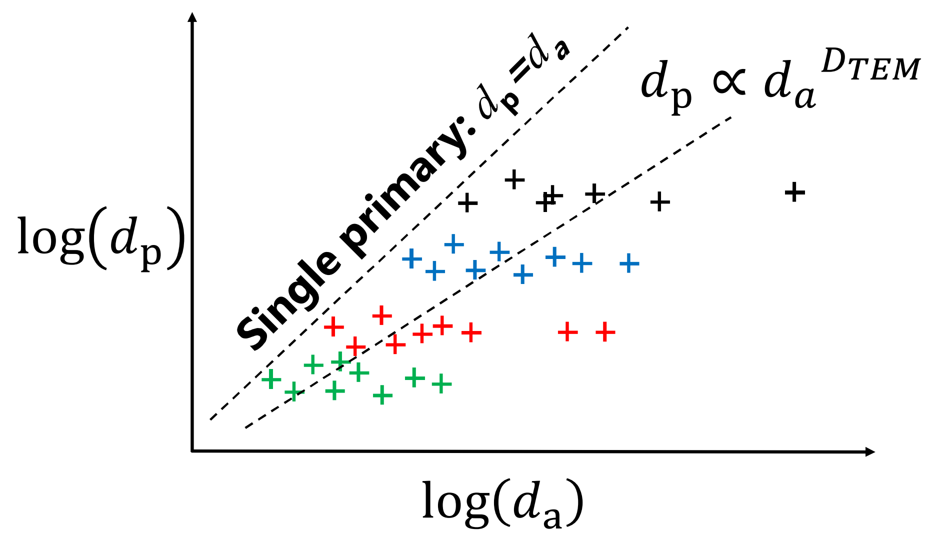
Dastanpour, R. & S. N. Rogak (2014) Observations of a Correlation Between Primary Particle and Aggregate Size for Soot Particles, *Aerosol Science and Technology*, 48:10, 1043-1049

External Mixing Hypothesis – by TEM data

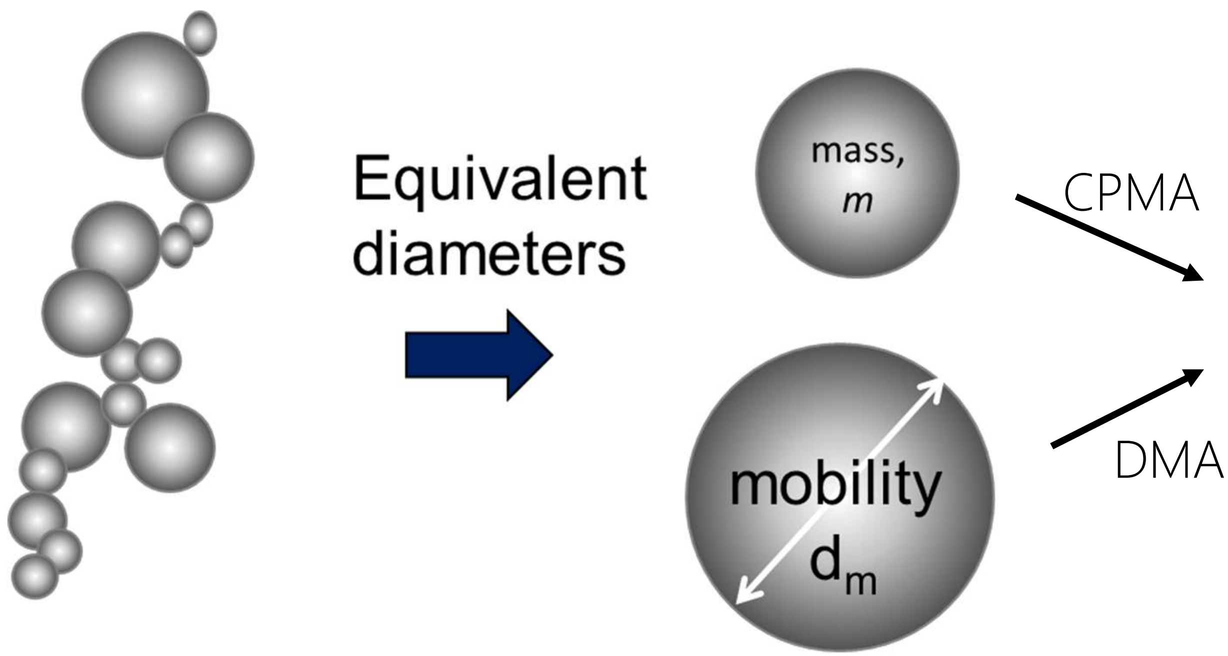
Hypothetical TEM images of soot



Hypothetical post-flame measurements



EFFECTIVE DENSITY



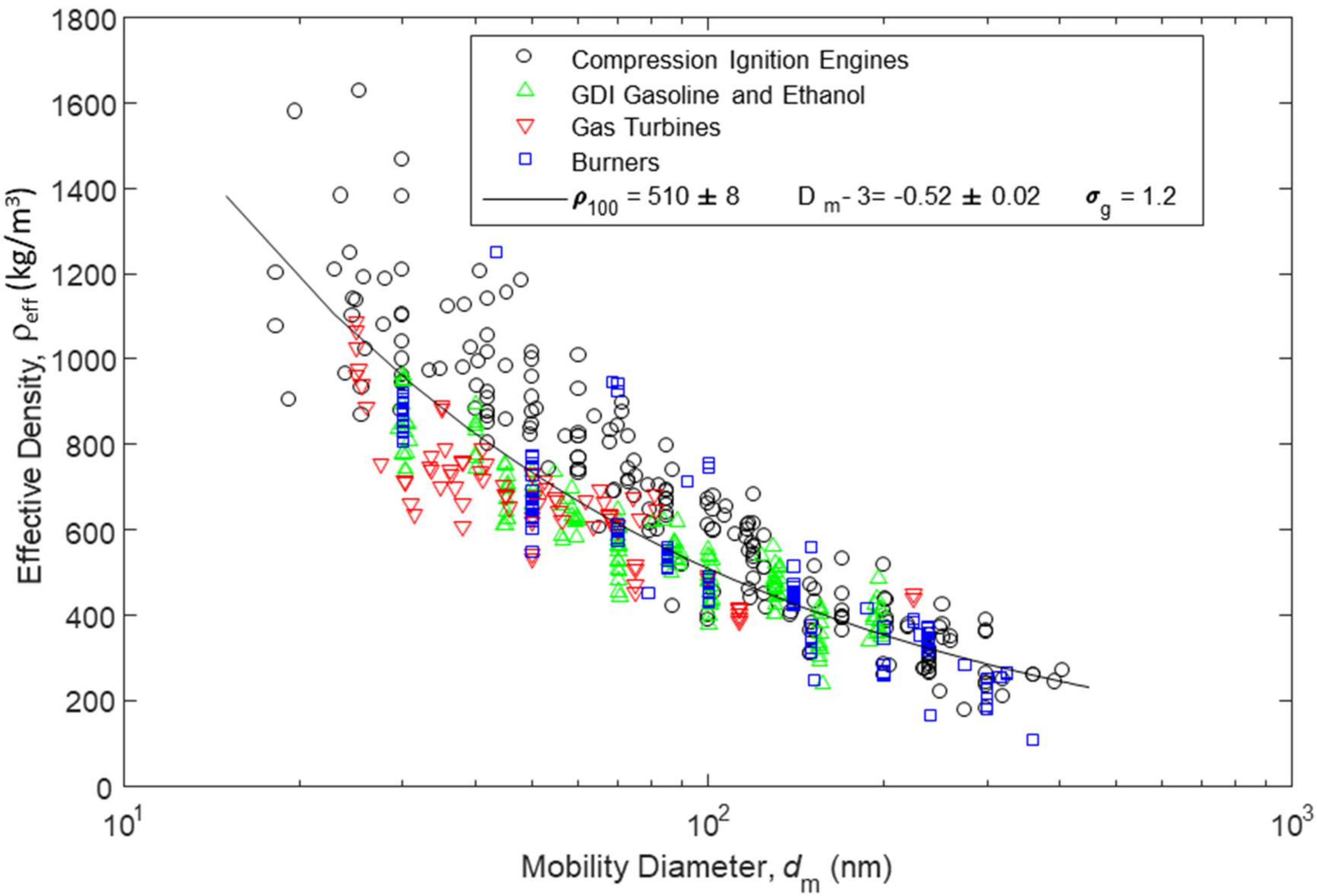
$$\rho_{eff} = \frac{6m}{\pi d_m^3}$$

For fractal aggregates

Mass ~ $d_m^{D_m}$ D_m=mass-mobility exponent

Effective density scales as D_m-3

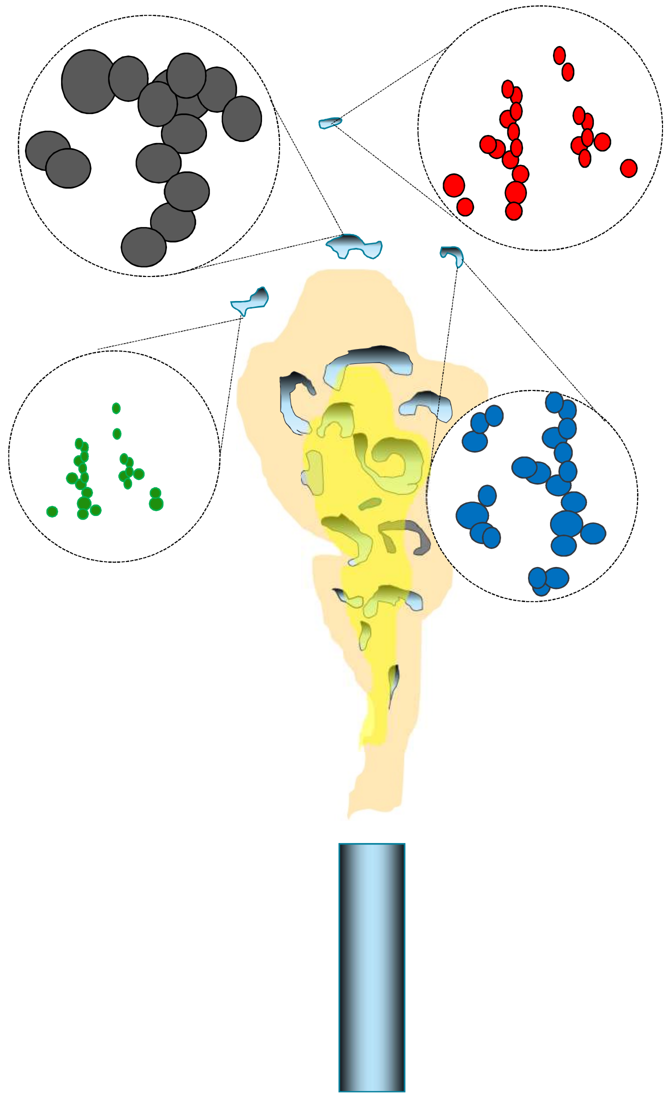
OBSERVATIONS OF EFFECTIVE DENSITY



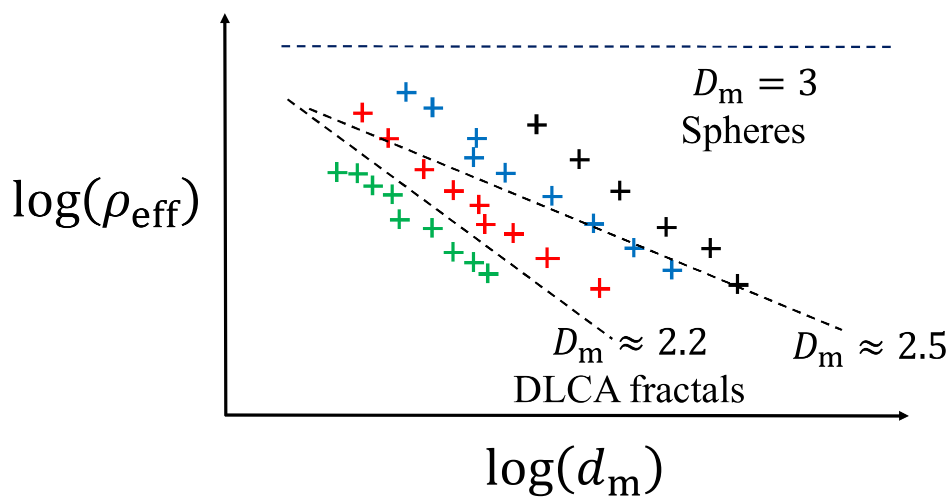
Olfert, J. and Rogak, S.N., "Universal Scaling Relations between Soot Effective Density and Primary Particle for Soot from Common Combustion Sources", *Aerosol Research Letters (Aerosol Sci. and Technol.)*, 2019.

External Mixing Hypothesis – by Effective Density

Hypothetical TEM images of soot



Hypothetical post-flame effective density measurements



LINKING TEM AND EFFECTIVE DENSITY

$$d_{p,100} = (100\ \text{nm}) \left(\frac{\rho_{eff,100}}{k_a \rho} \right)^{\frac{1}{3-2D_\alpha}}$$

$$D_{TEM} = \frac{D_m - 2D_\alpha}{3 - 2D_\alpha}$$

$d_{p,100}$ primary particle size for aggregates with 100 nm mobility diameter

Effective density measurements provide D_m and ρ_{eff}.

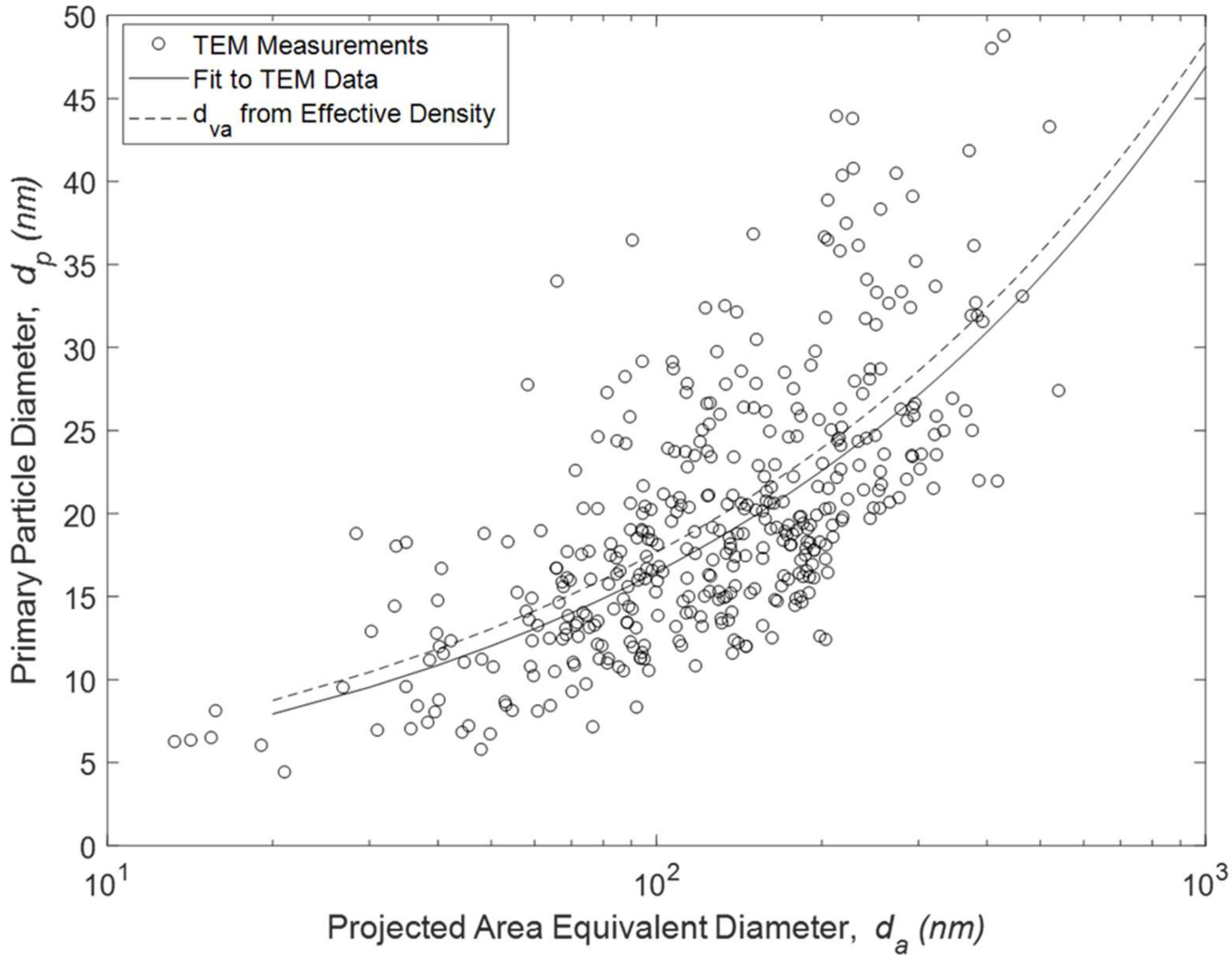
ρ is material density of carbon ~2 g/cc

Prior work on the drag of fractal aggregates provides k_a and D_α=1.1 →

$$D_{TEM} = \frac{D_m - 2.2}{0.8} \quad \sim 0.38\ for\ D_m = 2.5$$

Dastanpour, R., Rogak, S.N., Graves, B., Olfert, J., Eggersdorfer, M.L. & Boies, A.M. "Improved sizing of soot primary particles using mass-mobility measurements", Aerosol Sci. and Technol., 50:101-109, 2016.

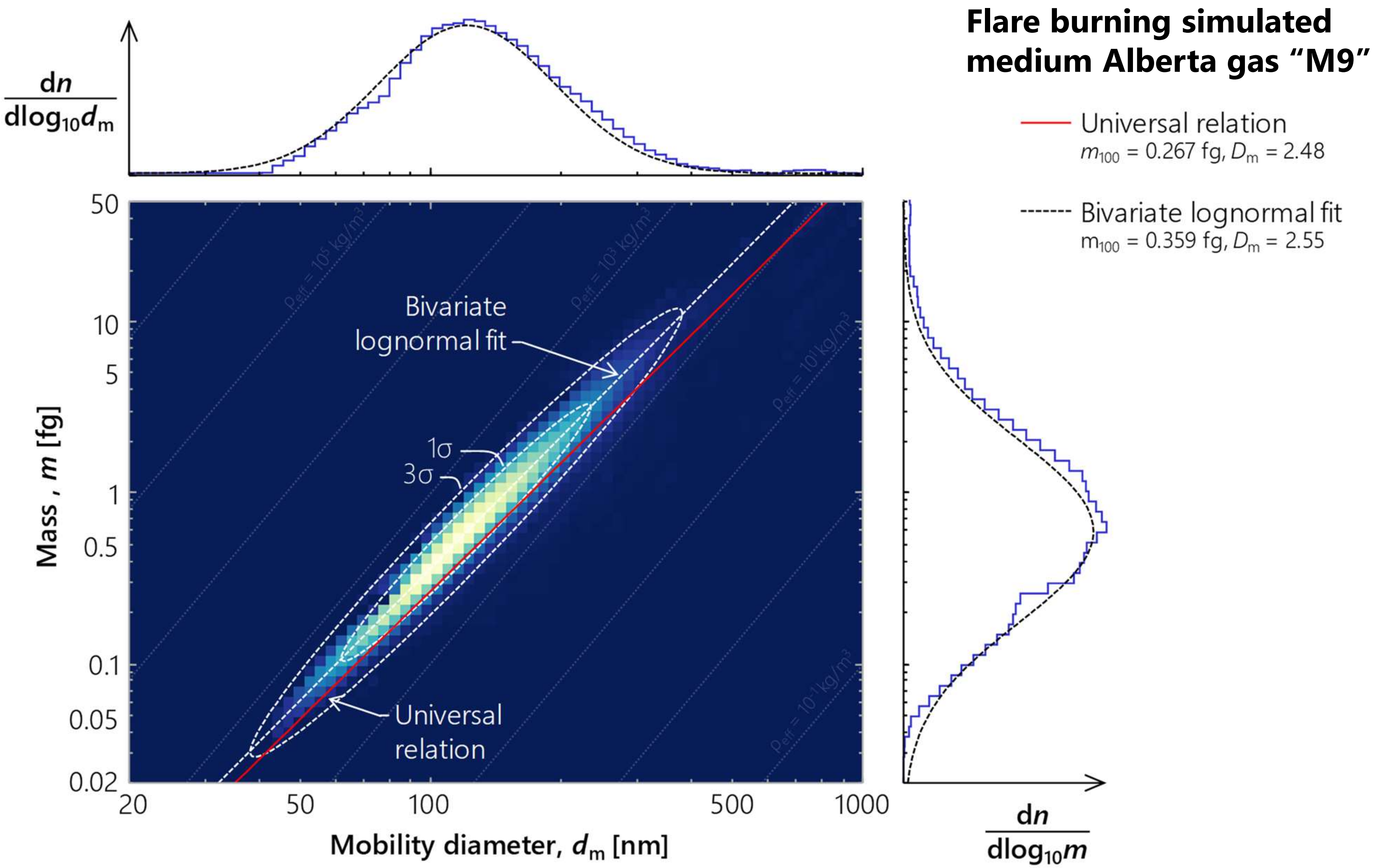
TEM AND EFFECTIVE DENSITY AGREE



Kazemimanesh, M., Dastanpour, R., Baldelli, A, Moallemi, A., Thomson, K.A., Jefferson, M.A., Johnson, M.R., Rogak, S.N., and Olfert, J.S., " Size, effective density, morphology, and nano-structure of soot particles generated from buoyant turbulent diffusion flames", *J. Aerosol Sci.*, 20 132:22-31, 2019.

PART 2 SOME NEW DIRECTIONS OF RESEARCH

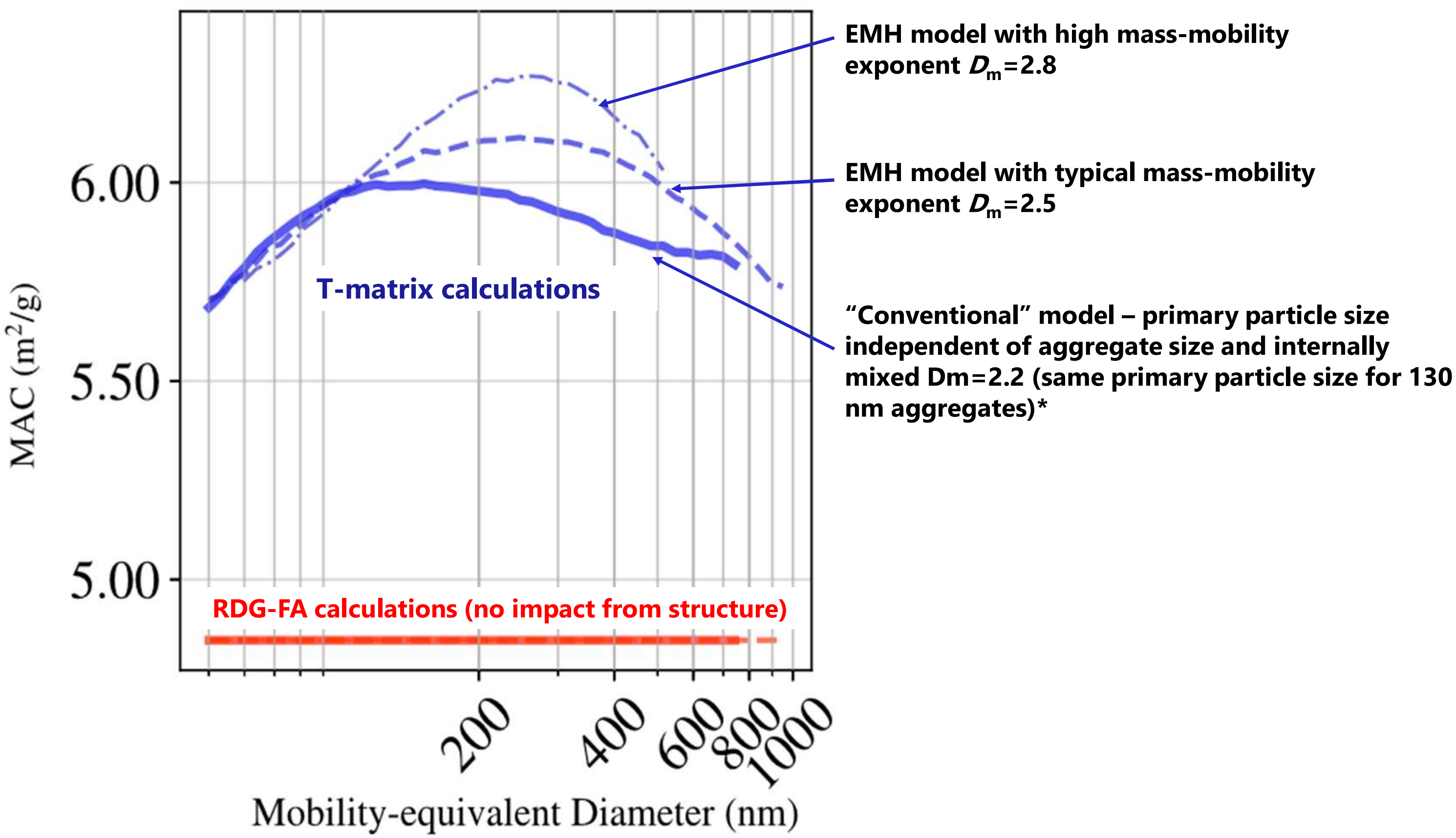
2-D MASS-MOBILITY DISTRIBUTIONS



Trivanovic, U., T.A. Sipkens, M. Kazemimaneshb, A. Baldelli, A. M. Jefferson, B.M. Conrad, M.R. Johnson, J. C. Corbin, J. S. Olfert, S. N. Rogak, "Morphology and size of soot from gas flares as a function of fuel and water addition", accepted for publication in Fuel, June 2020

IMPACT OF AGGREGATE STRUCTURE ON OPTICAL PROPERTIES

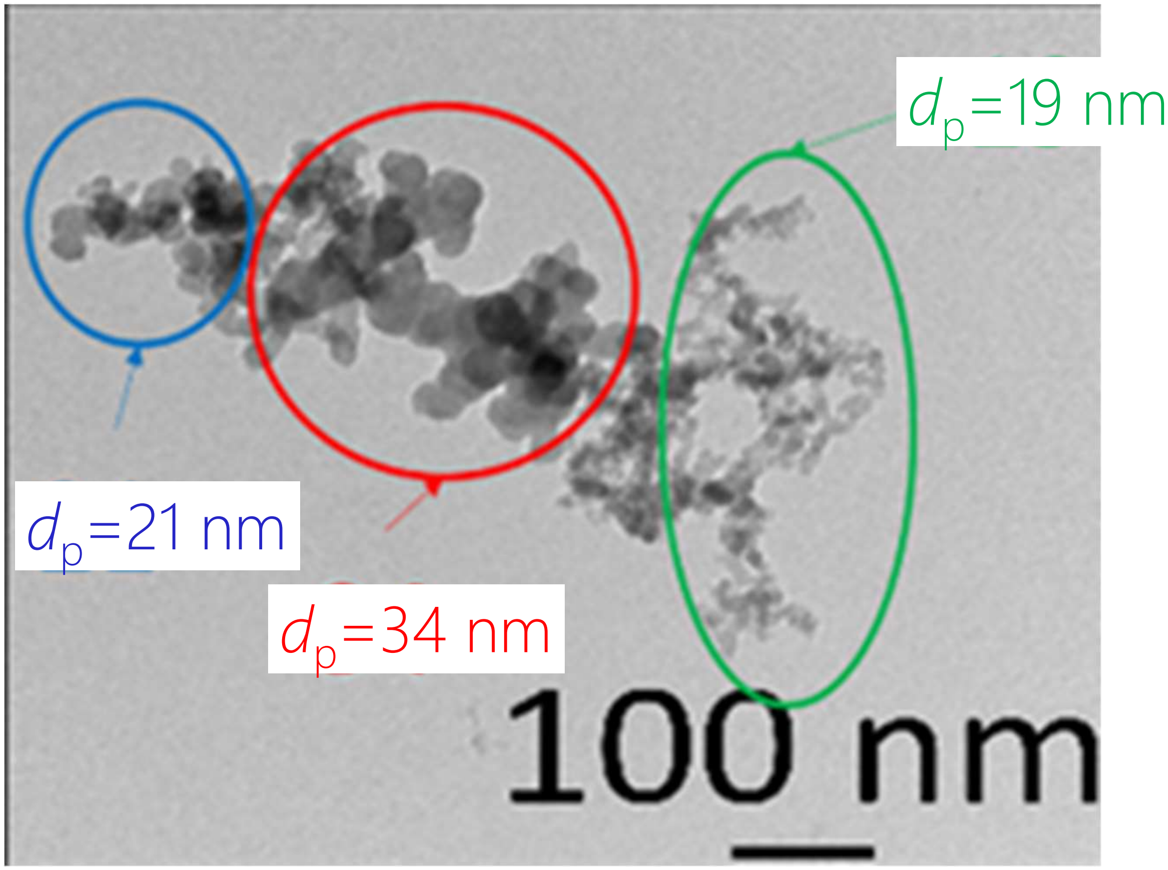
Mass-specific absorption coefficient (MAC) at 550 nm m_λ=1.95+0.79j
Computations with simulated aggregates



Keyhan Babaee MASc Thesis, University of British Columbia, April 2020

T-matrix calculations from Liu et al Atmos. Chem. Phys., 18, 6259–6273, 2018

THE EXTERNAL MIXING HYPOTHESIS BREAKS DOWN



An indication of post-flame dilution history?

Baldelli, A., Trivanovic, U., Corbin, J.C., Lobo, P., Gagné, S., Miller, J.W., Kirchen, P. and Rogak, S. (2020). Typical and Atypical Morphology of Non-volatile Particles from a Diesel and Natural Gas Marine Engine. Aerosol Air Qual. Res. 20: 730-740. https://doi.org/10.4209/aaqr.2020.01.0006

CONCLUSIONS

- Primary particle size variations are “externally mixed” in soot aggregates
- This gives broad 2-d mass mobility size distributions, characterized through new inversion methods
- The way the primary particles are distributed among the aggregates affects the optical properties – important for modelling polydisperse aerosols
- EMH describes “normal” soot, but the breakdown of the EMH could tell us more about soot emission processes

ACKNOWLEDGEMENT

NSERC CRSNG

NSERC flarenet

COMPARING EFFECTIVE DENSITY AND TEM

Source Type	From Effective Density		From TEM	
	D_{TEM}	$\mathbf{d_{va,100}}$ **(nm)**	D_{TEM}	$\mathbf{d_{p,100}}$ **(nm)**
Flare (Kazemimanesh *et al.*, 2019)	0.45	**17.4**	0.50	**17.1**
Inverted Burner				
N_2 diluted (Dastanpour *et al.* 2017)	0.39	**18.5**	0.38	**17.8**
High EC (Dastanpour *et al.*, 2017)	0.33	**23.9**	0.34	**17.7**
EQR 0.57 (Gazi *et al.*, 2013)	0.11	**15.7**	0.33	**14.5**
HPDI Compression Ignition Engine				
B75 20% EGR (Graves *et al.*, 2015)	0.19	**19.2**	0.13	**27.1**
B75 0% EGR(Graves *et al.*, 2015)	0.38	**19.8**	0.13	**21.9**
B50 20% EGR(Graves *et al.*, 2015)	0.29	**19.2**	0.20	**17.2**
B37 20% EGR(Graves *et al.*, 2015)	0.49	**23.7**	0.39	**18.4**
B25 20% EGR(Graves *et al.*, 2015)	0.53	**26.1**	0.45	**25.7**
Average GDI (Dastanpour *et al.*, 2016)	0.41	**17.7**	0.26	**19.0**
Aircraft Engine (Johnson *et al.*, 2015)	0.73	**21.3**	0.39	**30.2**
Average	**0.39**	**20.2**	**0.32**	**20.6**

Olfert, J. and Rogak, S.N., "Universal Scaling Relations between Soot Effective Density and Primary Particle for Soot from Common Combustion Sources", *Aerosol Research Letters (Aerosol Sci. and Technol.)*, 2019.