Supplemental Information:
Universal relations between soot effective density and primary particle size for common combustion sources

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Jason Olfert and Steven Rogak*

*Corresponding author

1. Effective Density Measurements

Figure S1 provides the effective density measurements presented in the main manuscript, but disaggregated by the original authors and experimental conditions. Legend entries refer to literature given in the main manuscript.

The regression curve on Figure S1 was obtained from simple linear regression of the logarithms of the data, such that the slope and intercept correspond to the prefactor and exponent of the un-transformed variable fit. The two fit parameters were thus the log of ρ_{100} and the log of 3-D_m. The 95% confidence intervals, Δ, in log-space were obtained directly from the Matlab regression function; in untransformed variables the interval is reported as, for example ρ_{100} ±x where

$$x = \frac{\exp(\ln(\rho_{100}) + \Delta) - \exp(\ln(\rho_{100}) - \Delta)}{2}$$

The fits applied equal weights to all data points simply because it is very difficult to assess the accuracy of the measurements from slightly different experimental setups and instrumentation. Because the fit is done in log-space, the regression implicitly assumes that the errors are lognormally distributed about the regression, and therefore the geometric standard deviation of the residuals (about 1.2) is a useful indicator of the spread in the data.

The McKenna burner measurements of Ghazi et al (2013) are highlighted with solid symbols as this was the only premixed laminar flame source included. The mass-mobility exponent is substantially lower (about 2.2) than the other points, which Ghazi et al noted was consistent with earlier studies. Given the equations discussed in the main manuscript, a mass-mobility exponent of 2.2 implies D_{TEM}=0 – that is primary particle does NOT vary with aggregate size. In fact this is what we would expect if all soot experiences the same formation history in the flame. We have included these points in the grand fit but note that the impact on the fit is insignificant (less than 1% change in fits) and that as more measurements become available, it might be possible to develop reliable correlations for more classes of soot sources.

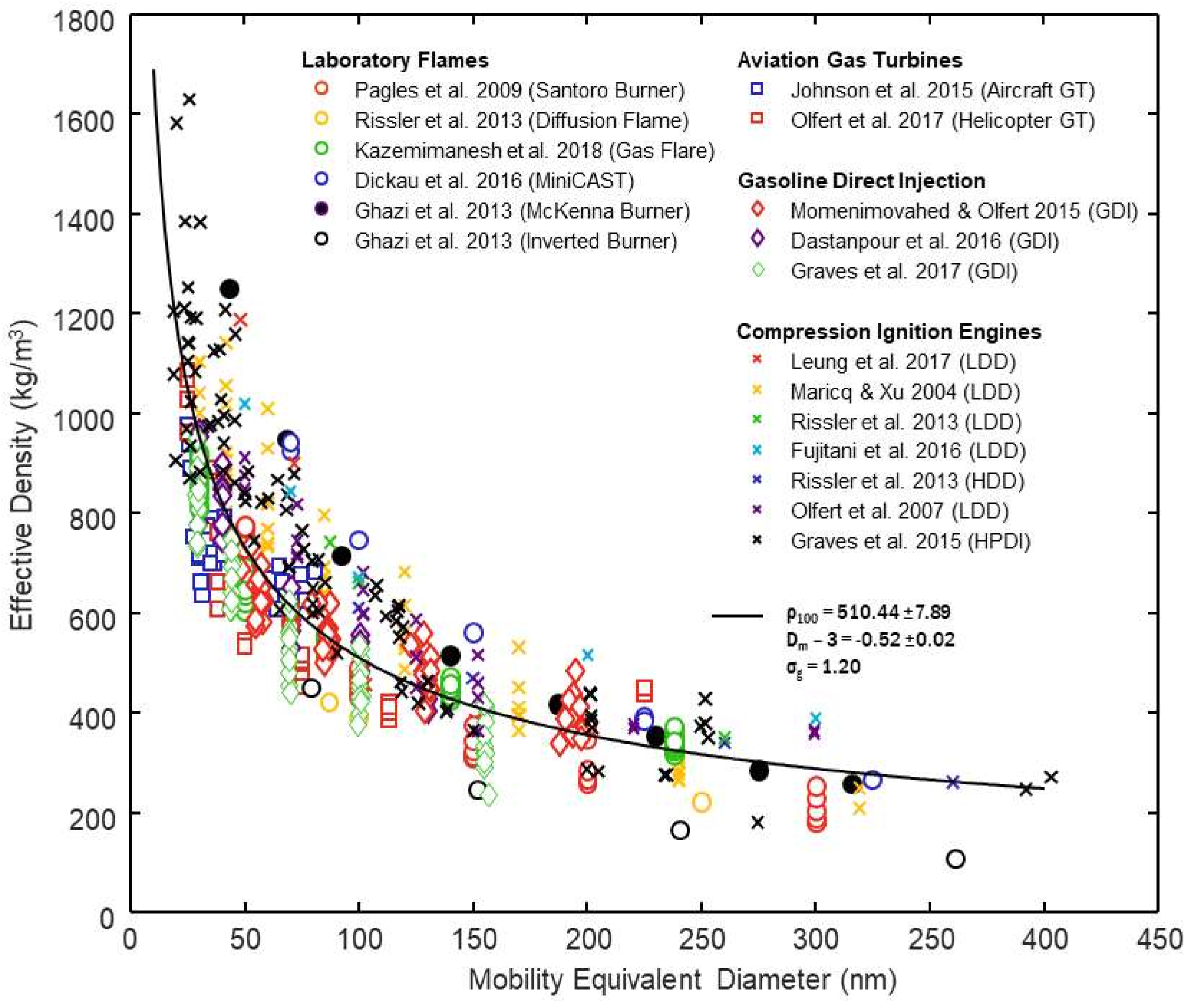

Figure S1 Summary of effective density measurements for fresh soot by source type and study author for light duty diesel engines (LDD), heavy-duty diesel engines (HDD), gasoline direction injection engines (GDI), high-pressure direct-injection methane-fueled compression-ignition engine (HPDI), gas turbines (GT), burners, and flames.

2. Uncertainties in Transmission Electron Microscopy Measurements and Fits

Information on the morphology of individual soot particles can be obtained from TEM analysis of samples that have been thermophoretically or electrostatically deposited on thin films. TEM offers sub-nanometer resolution, but typical images have low contrast so that it is difficult for manual or automated image processing to measure primary particles to better than +/- 1 nanometer, and applying different methods to the same image, sizing errors of 3-10% appear to be typical (Bescond, 2014[1]; Dastanpour, Boone and Rogak 2016[2]; Kook et al, 2016[3]). In determining the projected area-equivalent diameter, d_a, a few errors can arise. Firstly, there is the error (~ 1 nm depending on magnification) in determining the boundary of the image. Secondly, the orientation of the aggregate on the TEM substrate may not be random- for example the aggregate might fall "flat" on the substrate exposing the maximum area in the image. This might bias the TEM-based measurement relative to a true orientation-averaged area. Even if these errors were as large as 10%, there would be an insignificant impact on the appearance of Figure 3 in the main manuscript.

Statistical sampling errors can be important in primary particle sizing because in a single TEM sample, primary particle diameters can vary by nearly an order of magnitude (Dastanpour and Rogak, 2014). Because variations within aggregates are smaller, one must analyze images of a large number of aggregates, not a large number of primaries within a few aggregates.

Only a handful of studies have attempted to characterize the variation of primary particle size with aggregate size, and therefore it is difficult to assess the accuracy of the regression parameters (e.g., D_{TEM}). Table S1 provides data from studies where primary particle size was regressed against projected area diameter. In the earlier studies, the fitting function had the form $d_p = kd_A^{D_{TEM}}$, with diameters in nanometers. This contrasts subtly with the form used in the more recent studies

[1] Bescond, A., J. Yon, F. X. Ouf, D. Ferry, D. Delhaye, D. Gaffie, A. Coppalle, and C. Roze (2014). Automated determination of aggregate primary particle size distribution by TEM image analysis: Application to soot. *Aerosol Sci. Technol.* 48 (8):831–841. doi:10.1080/

[2] Dastanpour, R., J. Boone, and S.N. Rogak (2016). Automated Primary Particle Sizing of Nanoparticle Aggregates by TEM Image Analysis. *Powder Technol.*, 296:218-224.

[3] Kook, S., R. Zhang, Q-N Chan, T. Aizawa, K. Kondo, L.M. Pickett, E. Cenker, G. Brueaux, O. Anderson, J. Pagels and E. Nordin (2016). Automated Detection of Primary Particles from Transmission Electron Microscope (TEM) Images of Soot Aggregates in Diesel Engine Environments., *SAE International Journal of Engines*, 9(1):279-296.

(and present manuscript), $d_{\mathrm{p}} = d_{\mathrm{p10}} \left(\frac{d_{\mathrm{a}}}{100\ \mathrm{nm}}\right)^{D_{TEM}}$. The physical meaning of D_{TEM} is the same in both cases, and the 95% confidence intervals on this parameter can be compared, for either form of the fit function. However, $d_{\mathrm{p\,100}}$ has a more tangible meaning than k, and confidence intervals on these two parameters cannot be prepared because k is highly correlated with D_{TEM}, while $\mathrm{d}_{\mathrm{p100}}$ is not highly correlated with D_{TEM} because the mean aggregate size of most soot datasets is not far from 100 nm. While it is straightforward to compute d_{p100} given k and $\mathrm{D}_{\mathrm{TEM}}$, it is impossible to compute the confidence interval on this without going back to the original raw data- one reason that many entries in Table S1 are missing.

Table S1 also reports, where available, the geometric standard deviation of the residuals from the regression. These are typically far larger than the errors expected in measuring primary particle size, and therefore represent the distribution of primary particles sizes for a given projected area diameter.

Table S1. Summary of prior TEM measurements of primary particle d_p size including scaling relations d_a. Literature references are given in the main manuscript except Trivanovic et al[4].

Source type	d_{p100} (nm)	95% CI (nm)	D_{TEM}	95% CI	σ_g
Kazimemanesh et al (2018) Large laboratory flame					
Light fuel mixture	16.5	0.5	0.45	0.04	1.33
Medium fuel mixture	17.1	0.9	0.54	0.07	1.40
Heavy fuel mixture	17.9	1.0	0.52	0.07	1.34
Dastanpour et al (2017) inverted burner					
"N_2 diluted" fuel	17.8		0.38		
"High EC" condition	17.7		0.34		
Ghazi et al (2013) Inverted burner equivalence ratio 0.57	14.5		0.33		
Trivanovic et al (2019) dual fuel marine engine					
Diesel, idle	21.9	2.8	0.41	0.13	1.43
Diesel, 25% load	23.0	3.1	0.38	0.12	1.40
Diesel, 50% load	20.9	2.3	0.46	0.11	1.38
Diesel, 75% load	28.3	6.0	0.26	0.17	1.33
LNG+pilot, idle	23.0	3.6	0.35	0.14	1.35
LNG+pilot, low load	29.5	2.7	0.29	0.1	1.28
LNG+pilot, medium load	26.3	1.5	0.3	0.06	1.34
LNG+pilot, high load	29.4	2.4	0.27	0.08	1.33
Graves et al (2015) HPDI Compression Ignition Engine					
B75 20% EGR	27.1		0.13	0.05	
B75 0% EGR	21.9		0.13	0.08	
B50 20% EGR	17.2		0.2	0.08	
B37 20% EGR	18.4		0.39	0.11	
B25 20% EGR	25.7		0.45	0.14	
A63 80% premixed	22.5		0.31	0.07	
Dastanpour et al (2016) Gasoline Direct Injection Engine					
E0 Highway	22.9		0.18	0.15	
E0 Speed	18.8		0.16	0.16	
E10 Highway	20.0		0.27	0.1	
E10 Speed	16.4		0.45	0.12	
E30 Highway	18.7		0.21	0.12	
E30 Speed	17.2		0.26	0.7	

[4] Trivanovic, U., J.C. Corbin, P. Kirchen, W. Peng, J. Yang, W. Miller, S. Gagne and S.N. Rogak (2019). "Characteristics of particulate emissions from a dual-fuel marine engine: understanding mobility distributions through microscopy", *J. Aerosol Sci.* in preparation.