ESTIMATION OF EFFECTIVE DENSITY AND FRACTAL – LIKE DIMENSION OF SOOT PARTICLES

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MOTIVATION

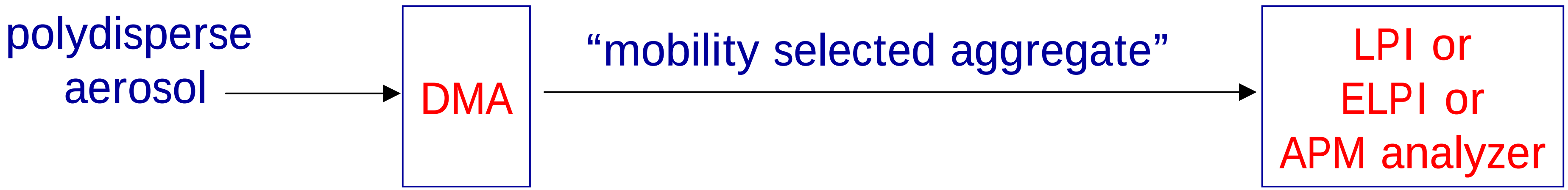
Structure of combustion generated aggregates is important for a number of applications

- Particulate filters (filtration, pressure drop, reactivity of soot cakes)
- Health effects of soot particles (transport, deposition, interaction with tissues)
- Interactions of soot particles with atmospheric constituents
- Performance of carbon black based products

MEASUREMENT METHODS FOR SOOT PARTICLE STRUCTURE

- Tandem mobility-aerodynamic or mobility-mass measurements

  Skillas et al. (1998), Maricq et al. (2000), Park et al. (2003), Van Gulijk et al. (2004), Maricq & Xu (2004)



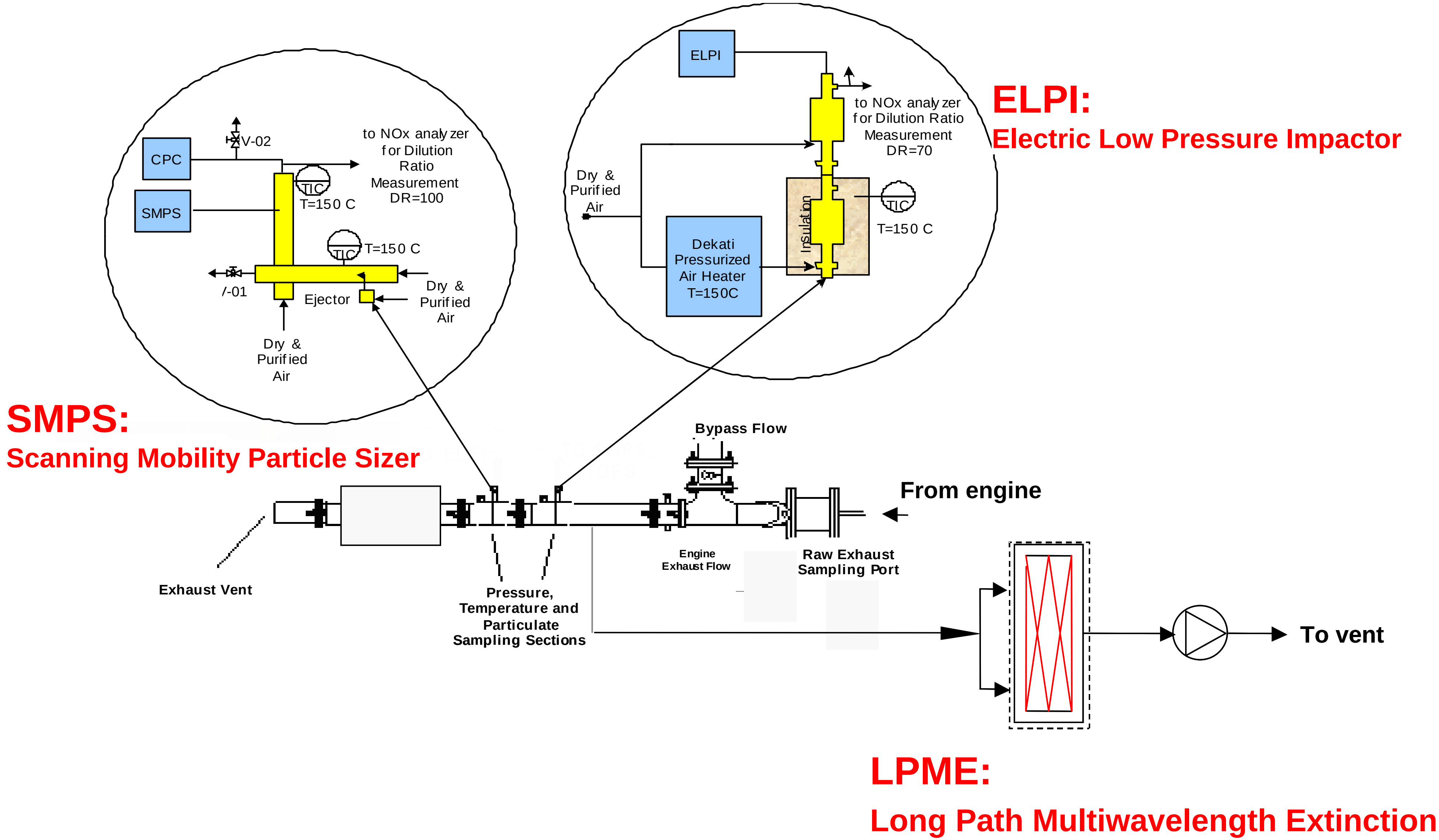
- Parallel application of SMPS and ELPI and matching the two distributions

  Ristimaaki et al (2002), **Present work**

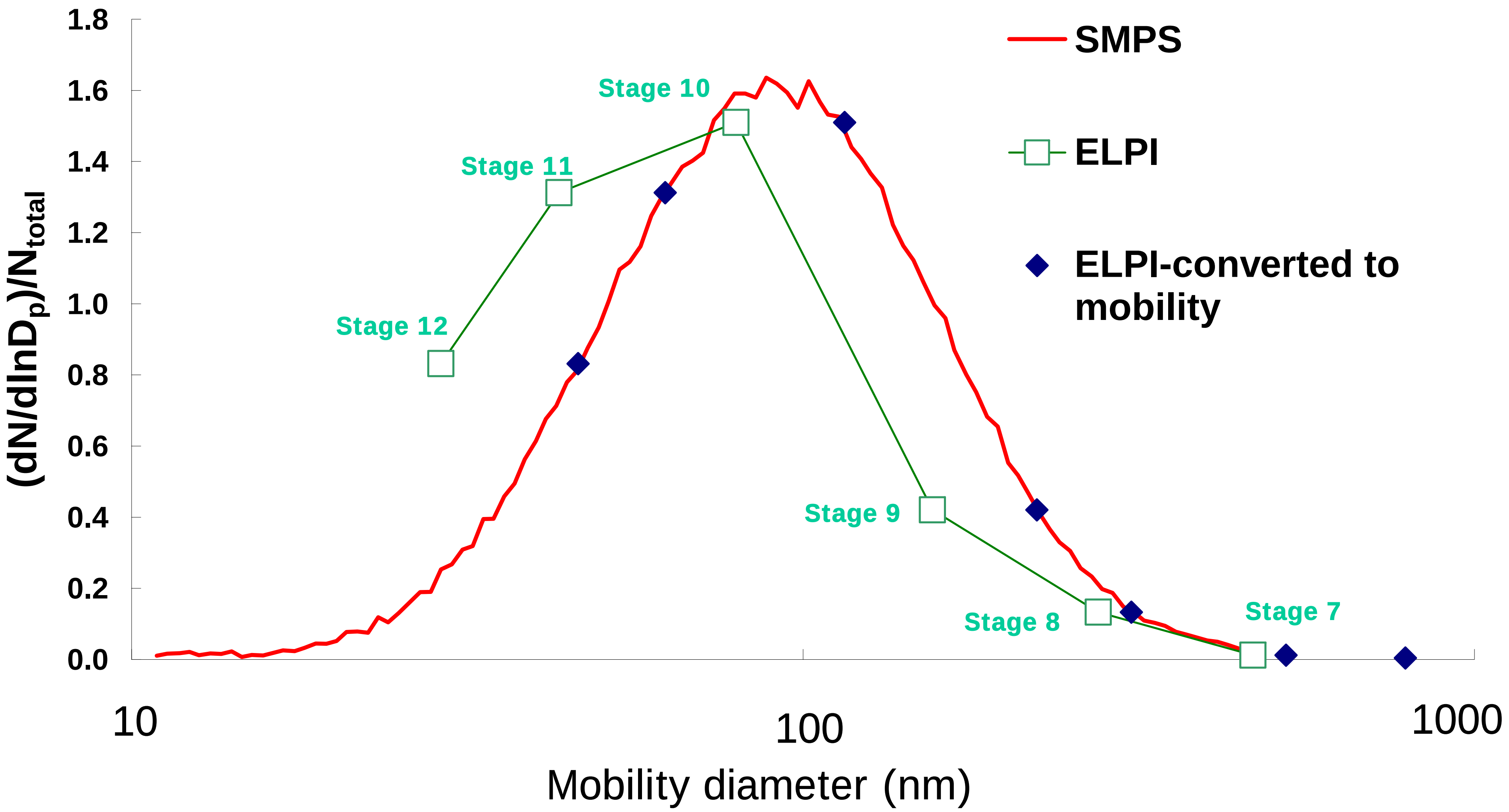
- Optical methods and microscopy

  Sorensen and co-workers (1995-2004), di Stasio et al. (1999-2004)

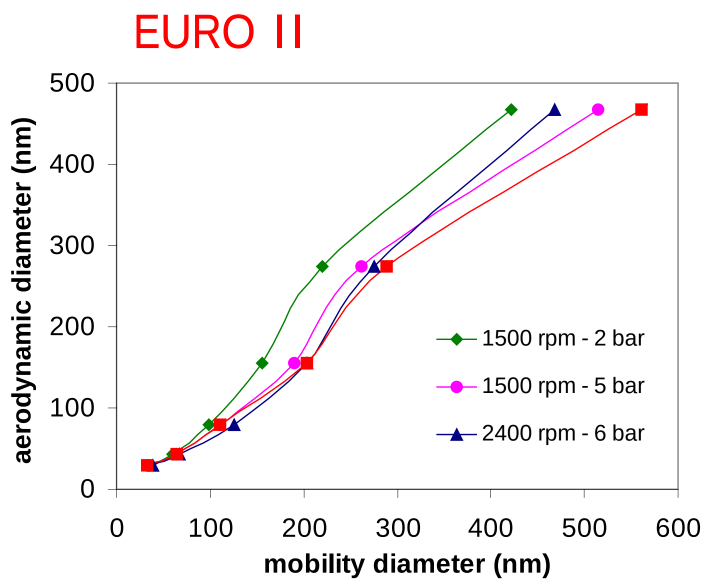
PARTICLE MEASUREMENT SETUP

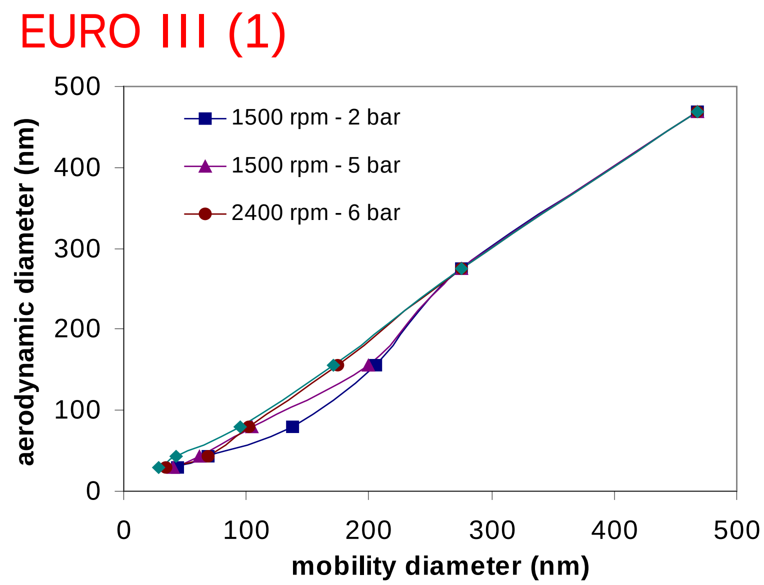


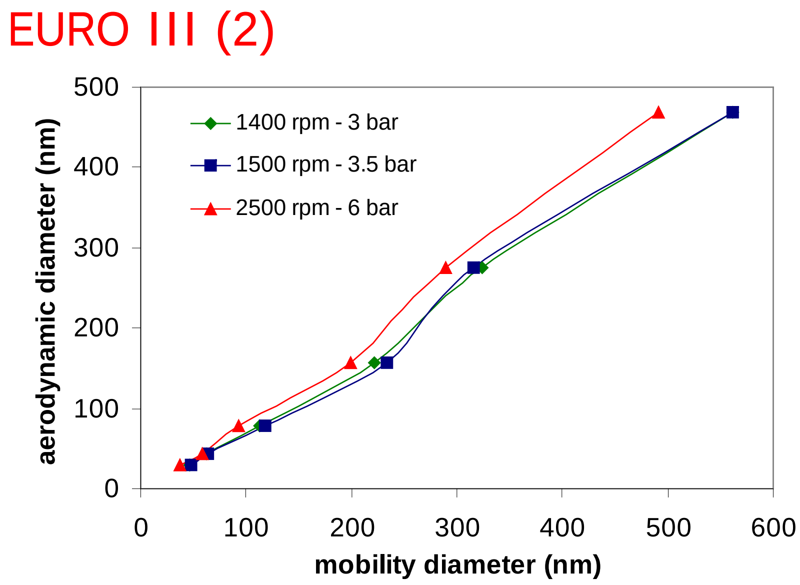
AERODYNAMIC vs. MOBILITY DIAMETER



AERODYNAMIC vs. MOBILITY DIAMETER
FOR 3 PASSENGER CAR DIESEL ENGINES





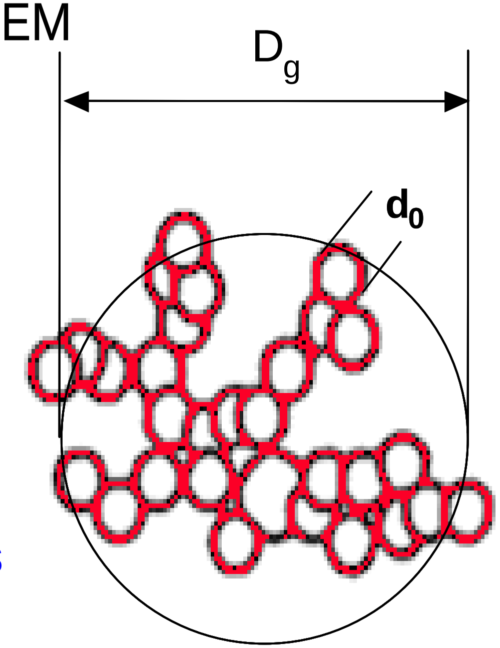


DIESEL SOOT FRACTAL AGGREGATES: Definitions

Density and size of primary particles

$\rho_0 \approx 2150\ \mathrm{kg/m^3}$ (CV 20%) based on gravimetry vs. LPME

$d_0 \approx 32\ \mathrm{nm}$ (CV 20%) based on soot cake permeability and TEM



Number of primary particles per aggregate

$$N_A = k_g \left[\frac{D_g}{d_0}\right]^{D_f} \qquad k_g = \frac{1}{f}\left[\frac{D_f}{D_f+2}\right]^{-\frac{D_f}{2}}$$

volume filling factor, Naumman (2003) $f \approx 1.43$

$D_f \approx 2.4$ on the average   Kittelson & McMurry (2002) and others

Geometric diameter

$$\frac{D_{geo}}{d_0} = [fN_A]^{\frac{1}{D_f}}$$

Diameter of gyration

$$D_g = \left[\frac{D_f}{D_f+2}\right]^{\frac{1}{2}} D_{geo}$$

Mass equivalent diameter

$$\frac{D_{geo}}{d_0} = \left[[f(\frac{D_{mass}}{d_0})^3]^{1/D_f} \right]$$

MOBILITY DIAMETER OF FRACTAL AGGREGATES

$$D_{me} = h_{KR} D_{geo} = (-0.06483 D_f^2 + 0.6353 D_f - 0.4898) D_{geo}$$

h_{KR} : **Kirkwood – Riseman ratio accounting for shielding effects and hydrodynamic interactions**

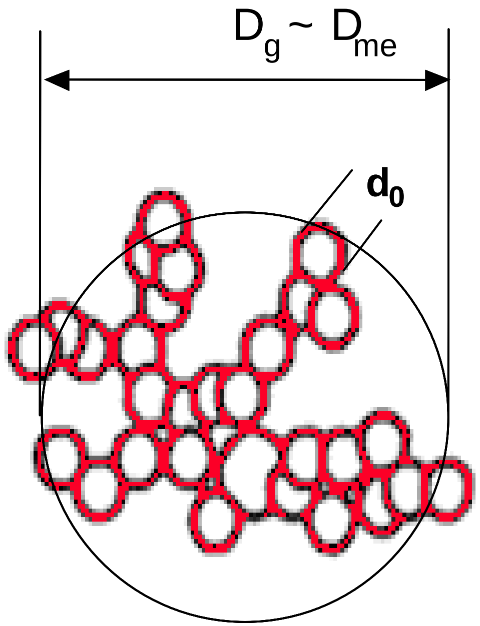
Naumman (2003)

Gyration diameter ~ Mobility diameter

$$D_g = \left[\frac{D_f}{D_f + 2} \right]^{\frac{1}{2}} \frac{D_{me}}{h_{KR}}$$

Fractal scaling based on Mobility diameter

$$N_A = k_g \left[\frac{D_g}{d_0} \right]^{D_f} = k_m \left[\frac{D_{me}}{d_0} \right]^{D_f}$$



EFFECTIVE DENSITY OF AGGREGATES

$$\rho_{\text{eff}} D_{me}^2 C_c(D_{me}) = \rho_1 D_{ae}^2 C_c(D_{ae})$$

ρ_{eff} : effective density

D_{me} : mobility diameter

ρ_1 : unit density (1 g/cm^3)

D_{ae} : aerodynamic diameter

C_c: Stokes-Cunningham Factor

Basic equations of analysis

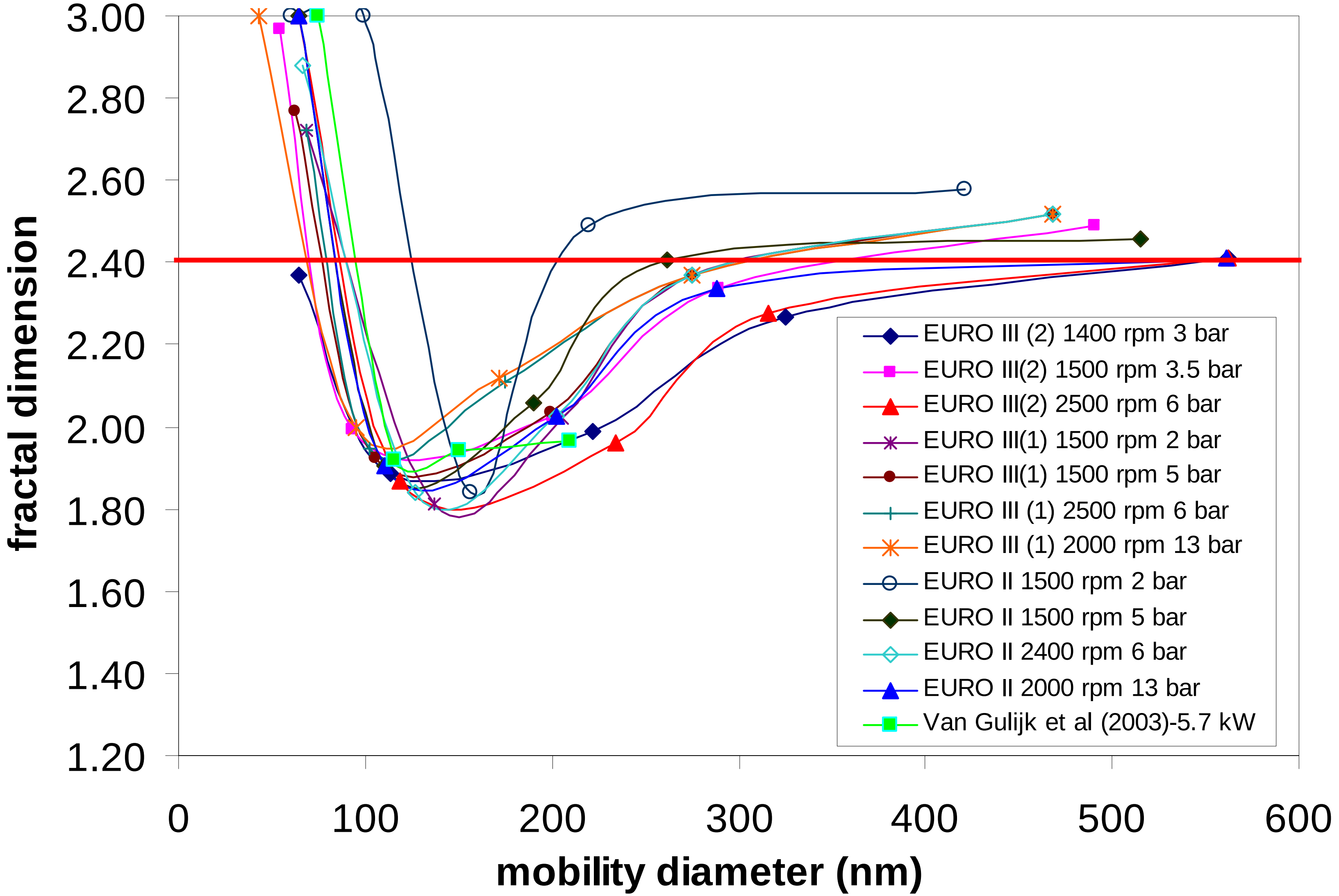
$$\frac{\rho_0}{\rho_1} = \frac{D_{ae}^2 C_c(D_{ae}) f\, h_{KR}^{D_f}}{D_{me}^{D_f - 1} d_0^{3-D_f} C_c(D_{me})}$$

$$\frac{\rho_{\text{eff}}}{\rho_0} = \frac{1}{f\, h_{KR}^{D_f}} \left[\frac{D_{me}}{d_0}\right]^{D_f - 3}$$

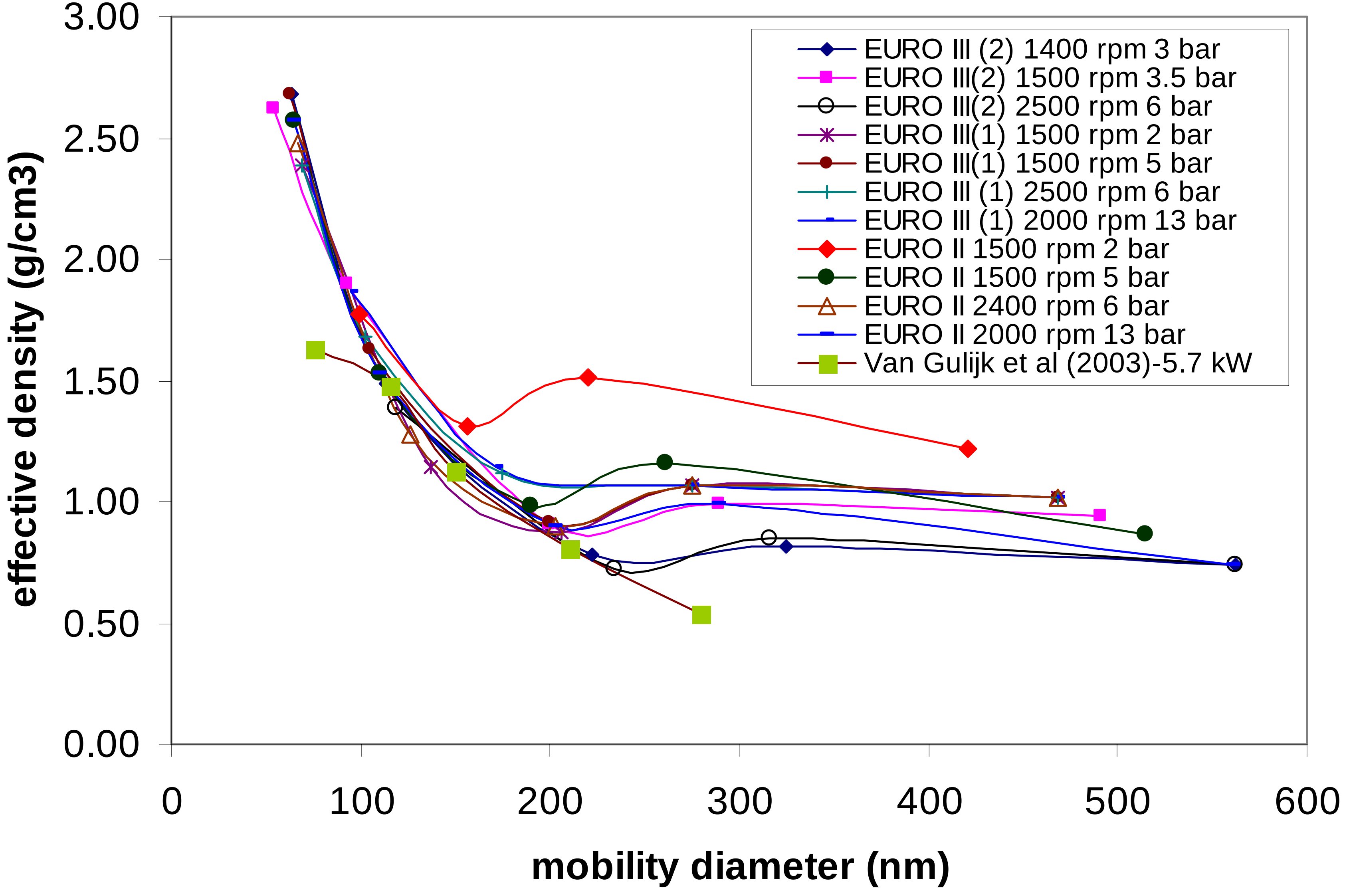
$$D_f$$

DIESEL SOOT AGGREGATE FRACTAL DIMENSION

3 different diesel engines & 1 gen set

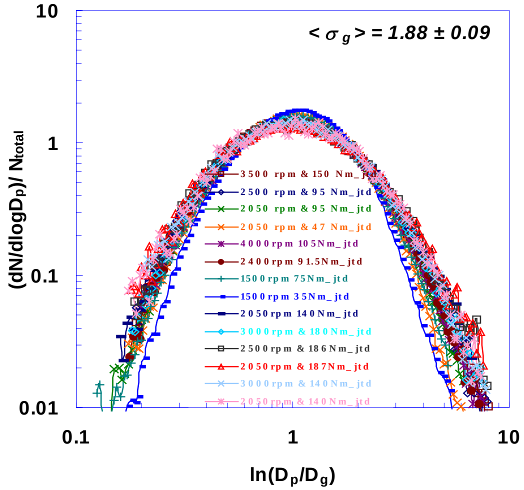


DIESEL SOOT AGGREGATE EFFECTIVE DENSITY

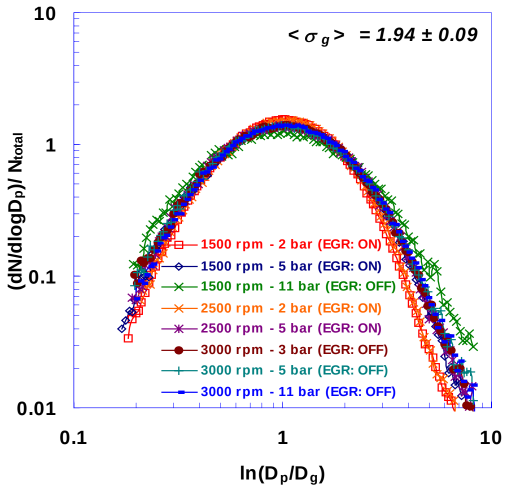


Diesel aggregate size distribution:
5 Engines (1996-2003) with engine displacement 1.9-2.4 litres

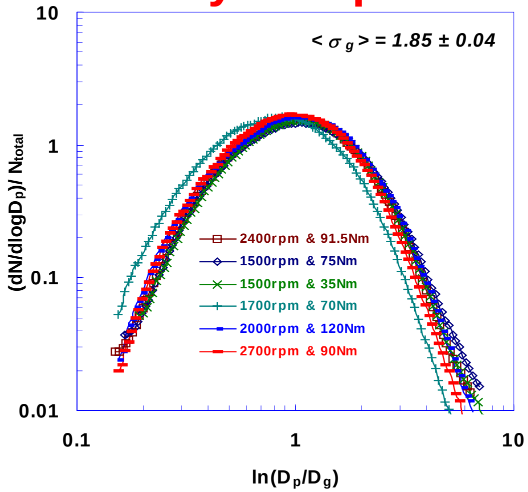
Common Rail – 1



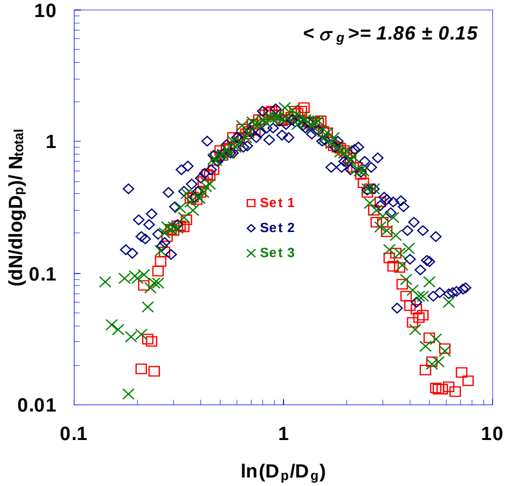
Common Rail-2



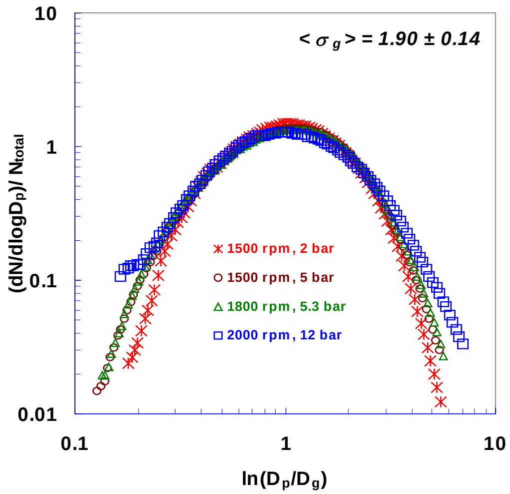
Rotary Pump



Common Rail-3



Pump Unit Injector

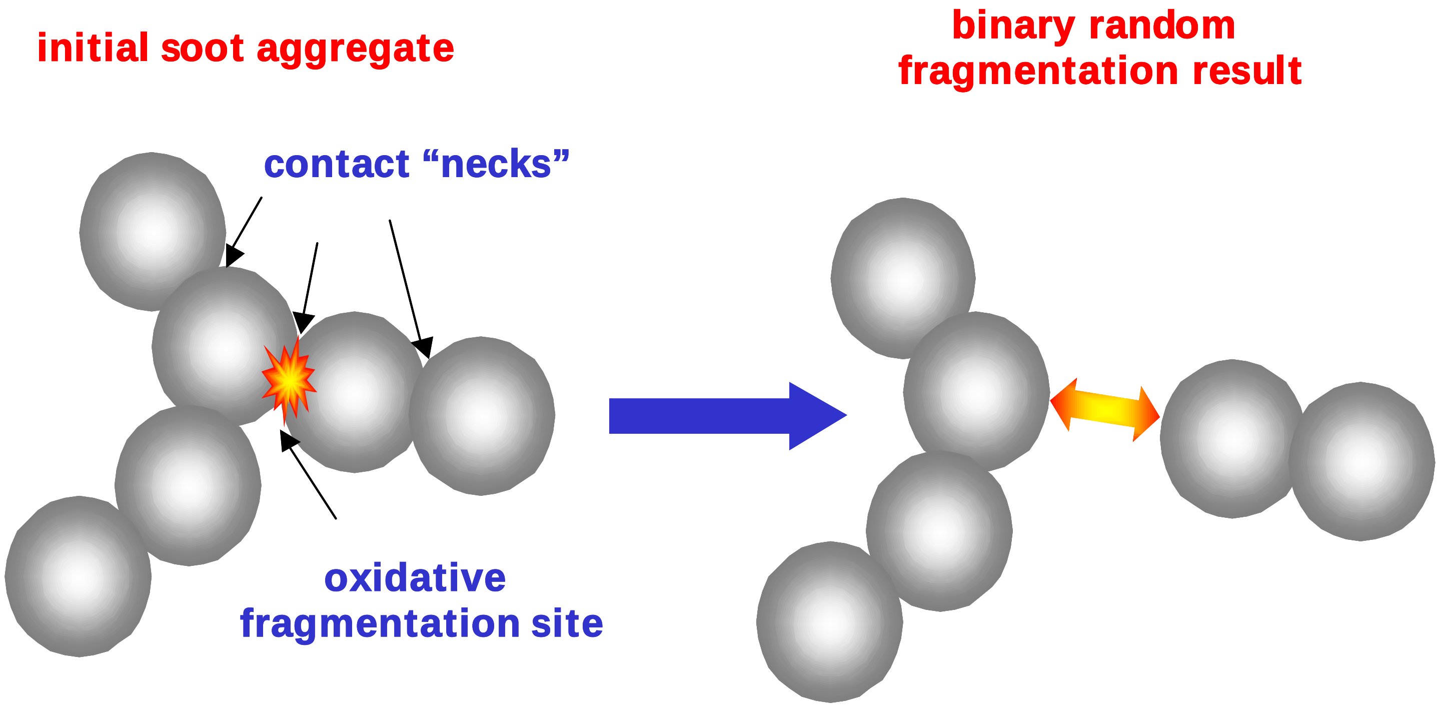


Universal Lognormal Shape:
σ_g=1.89 +/- 0.08

cf. Harris & Maricq (2002) 1.7-1.8

Kostoglou & Konstandopoulos (2003)

Steady state shape is determined from the ratio of oxidative fragmentation to coagulation rate



Continuous, binary random fragmentation process with size dependent rate:

$$S_i = Ai^b = Ai^{1/D_f}$$

In the large aggregate limit it can be shown that $\ln \sigma_g = \ln(6)/2(1+b)$

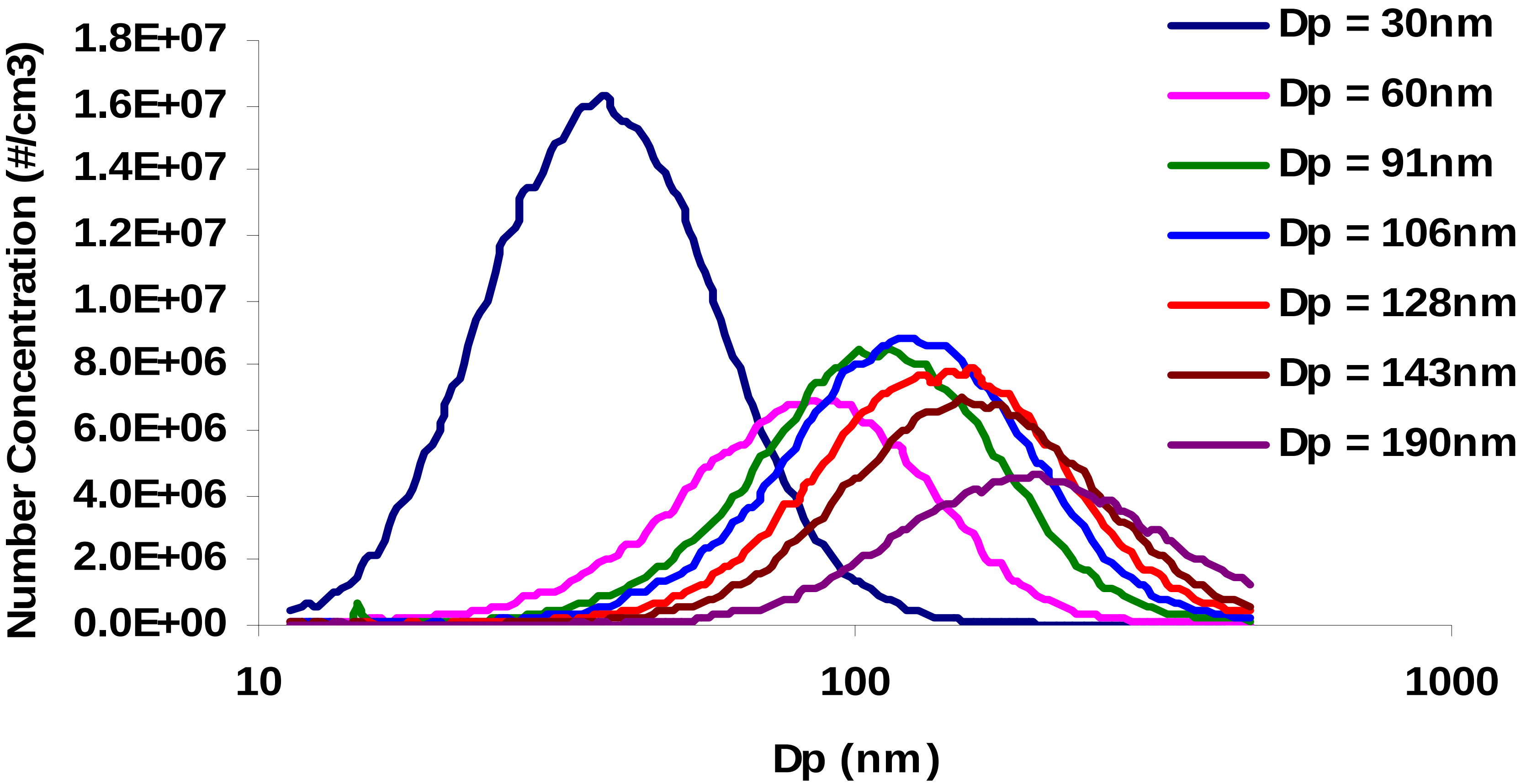
For σ_g=1.89 , b = 0.42 and D_f = 2.38

Kostoglou & Konstandopoulos (2003)

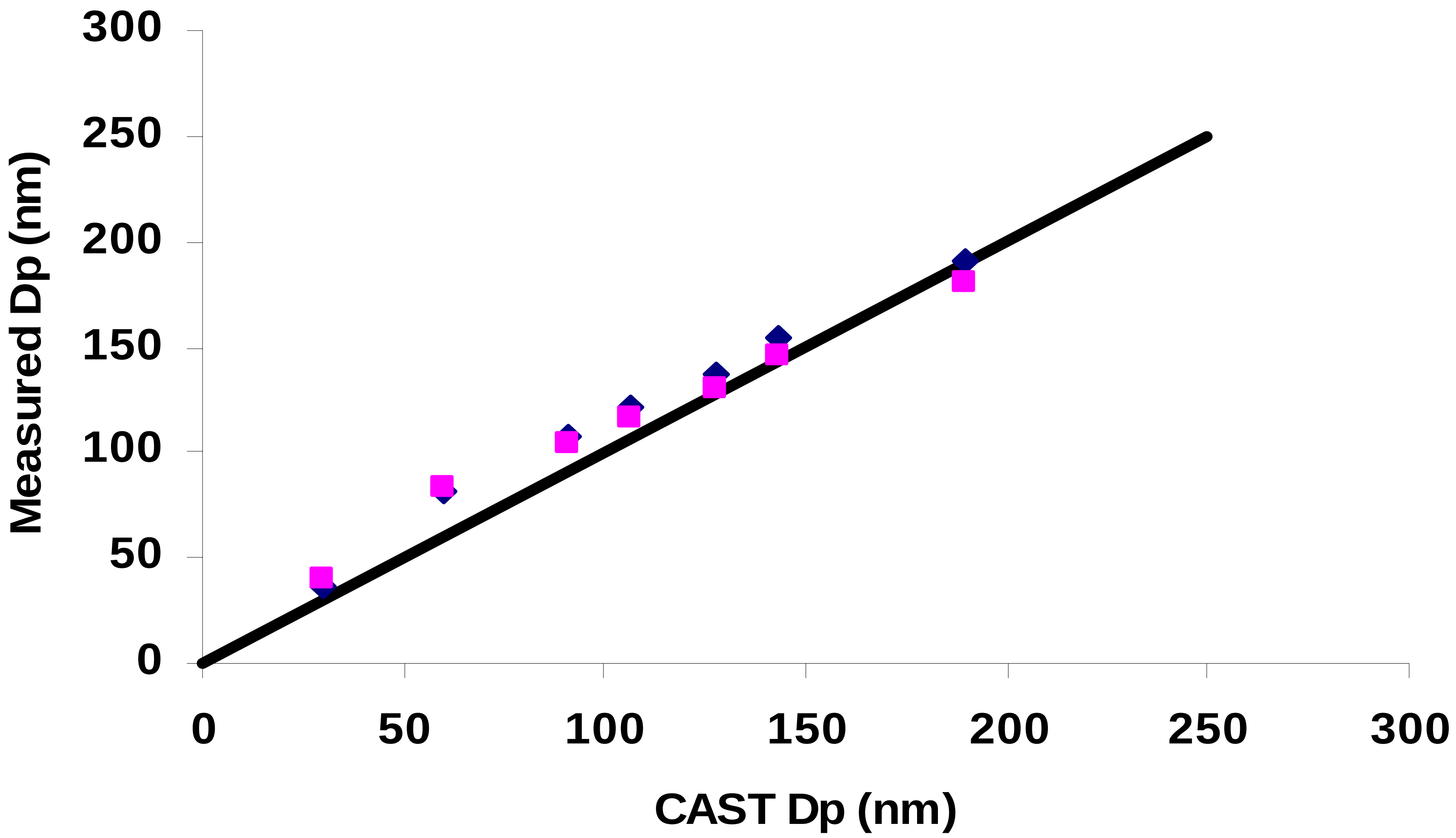
CAST SOOT GENERATOR

Provides Reference Soot Size Distributions

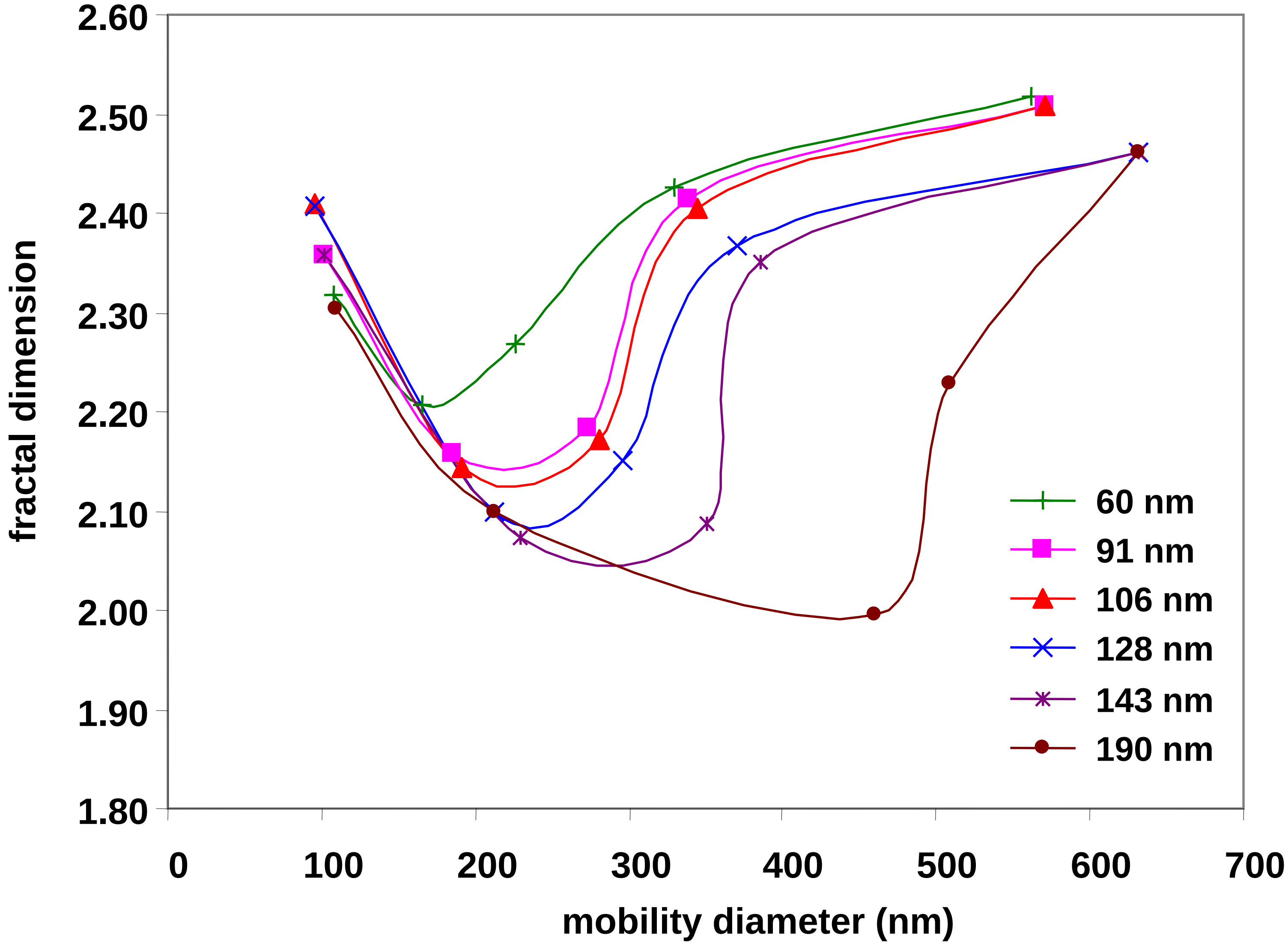
SIZE DISTRIBUTIONS FROM CAST



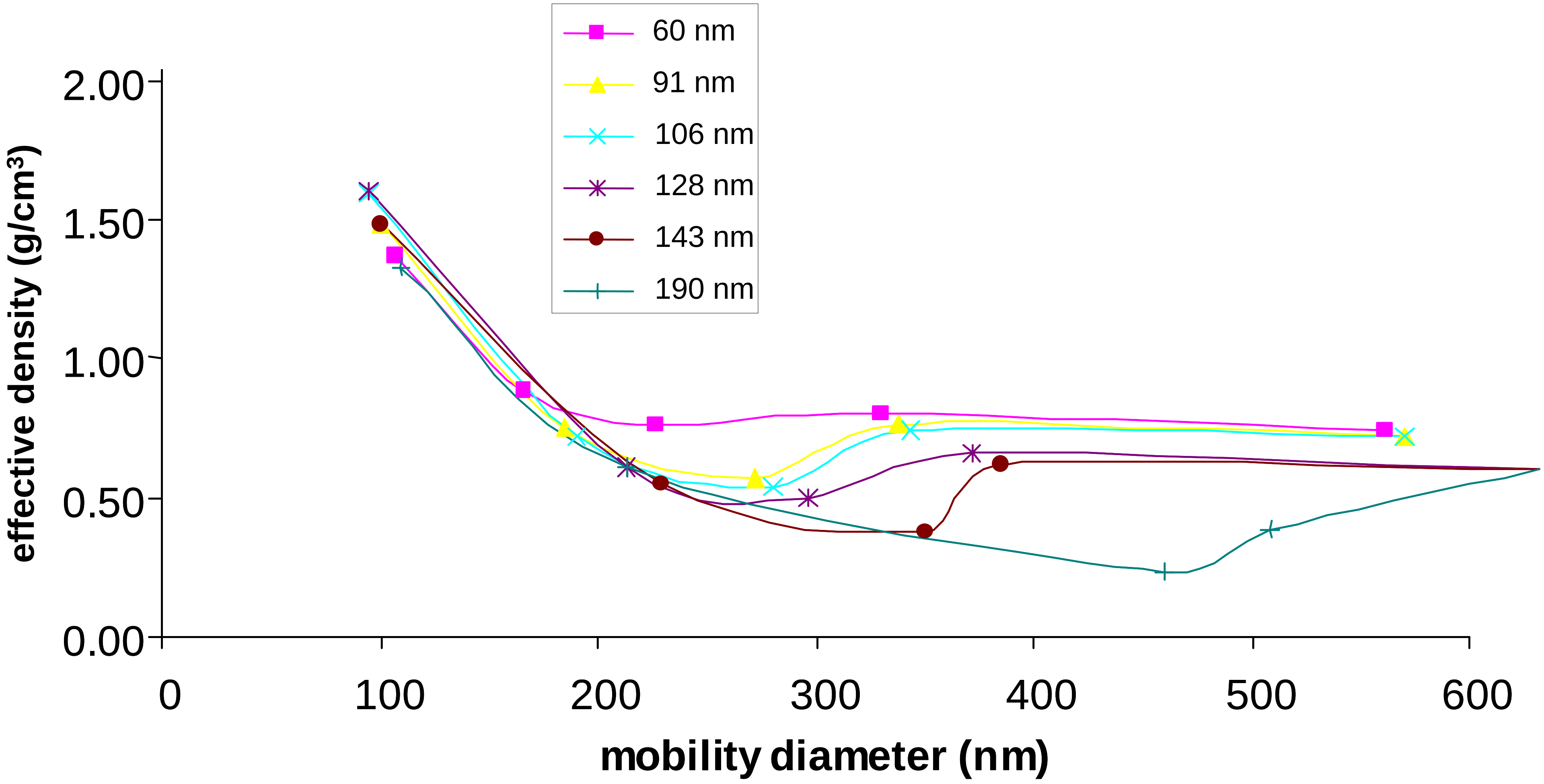
CAST CALIBRATION WITH SMPS



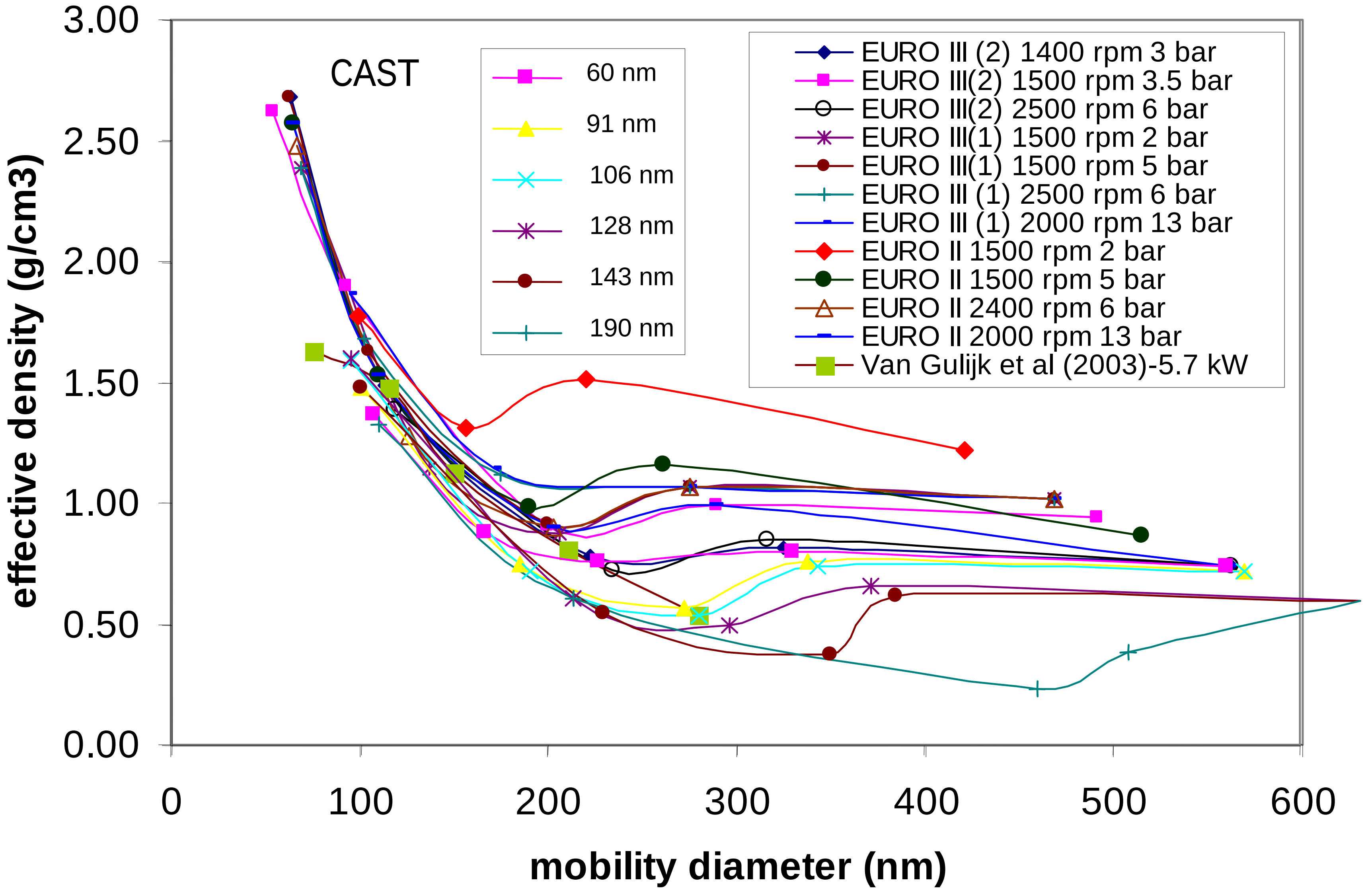
CAST SOOT AGGREGATE FRACTAL DIMENSION



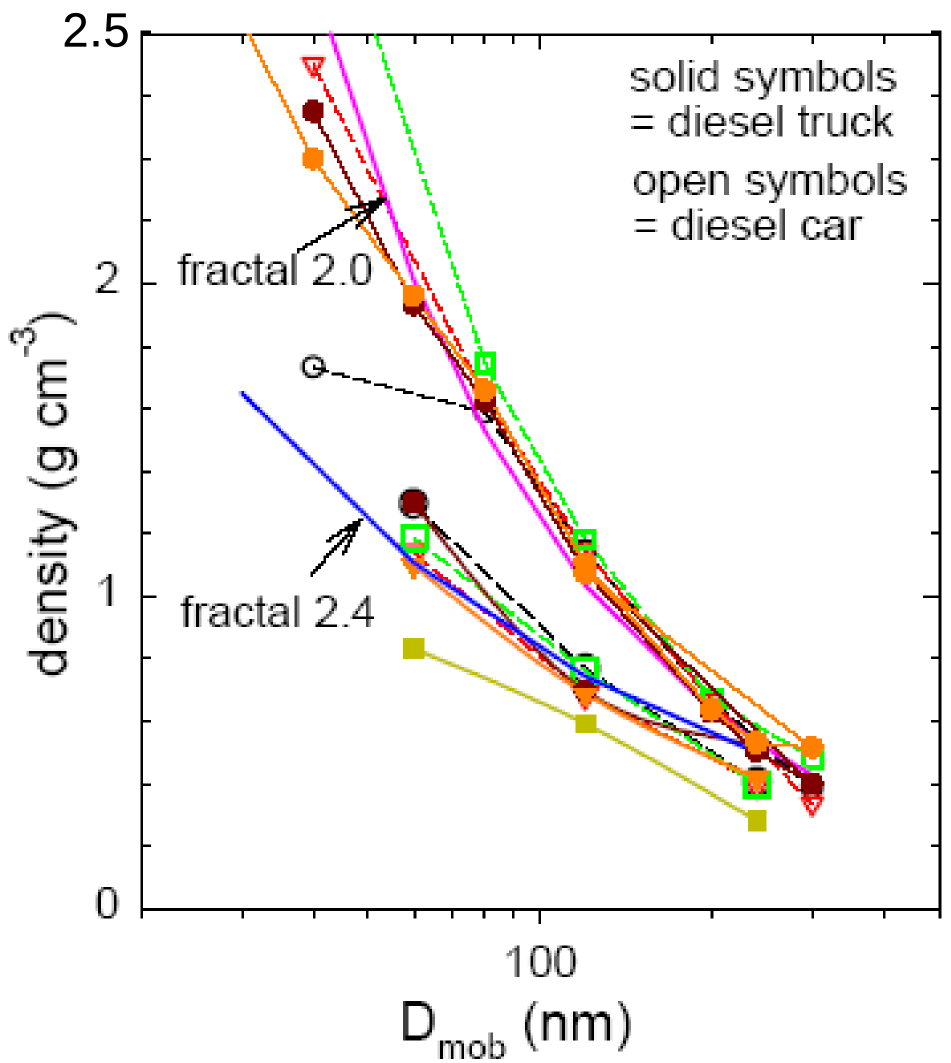
CAST SOOT AGGREGATE EFFECTIVE DENSITY



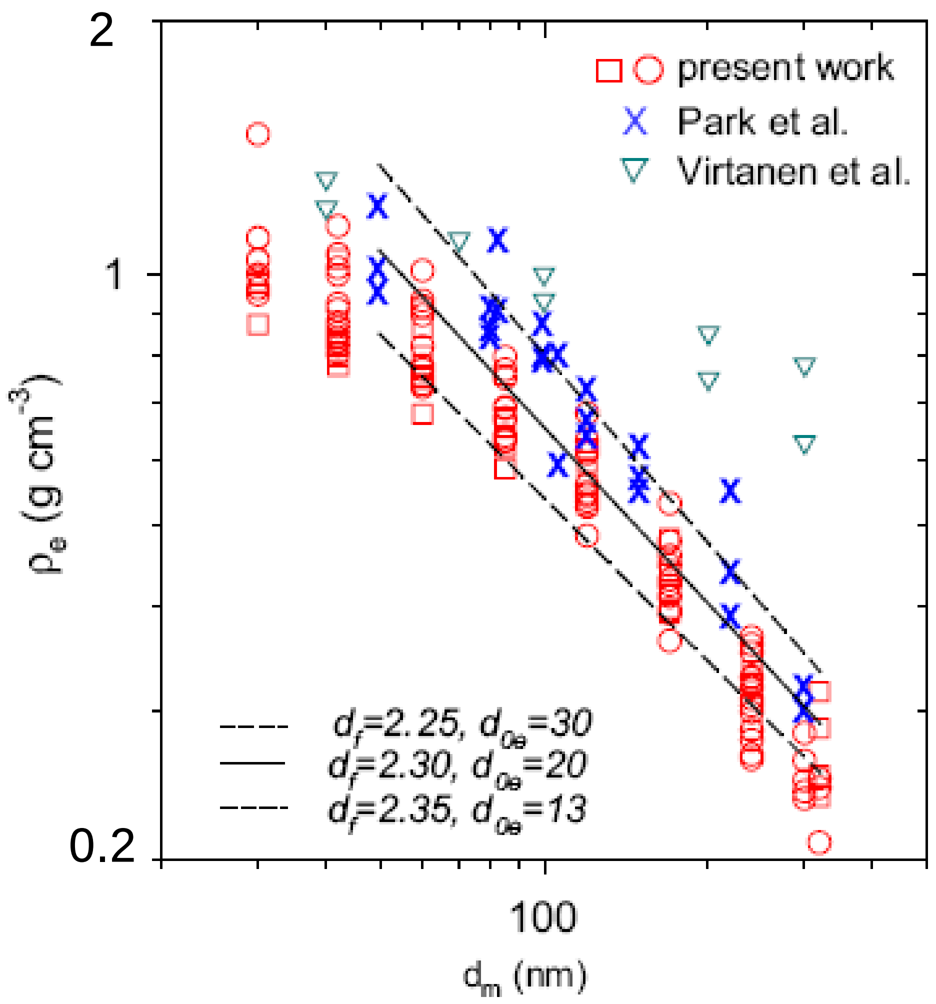
SOOT AGGREGATE EFFECTIVE DENSITY: CAST & DIESEL



OTHER STUDIES

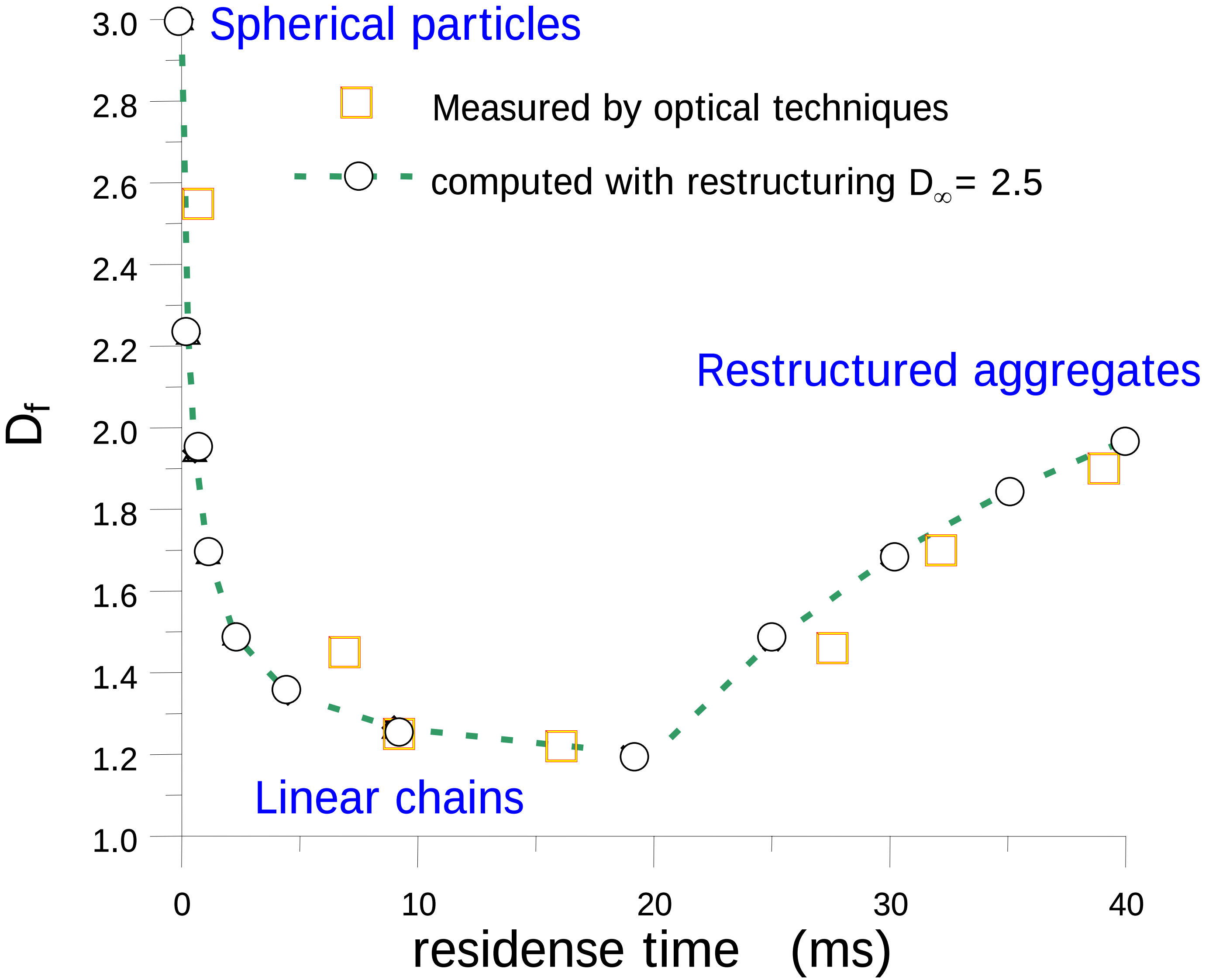


Maricq 2003 ELPI Workshop



Maricq & Xu (in press)

SOOT FRACTAL DIMENSION EVOLUTION IN DIFFUSION FLAME



Di Stasio, Konstandopoulos & Kostoglou (2002)

CONCLUSIONS

- Fractal dimension of soot from 3 diesel engines and a CAST burner changes non-monotonically with mobility diameter
- For diesel soot aggregates D_f decreases sharply from 3 down to ~1.8 – 1.9 with aggregate size up to about 100 nm
- For larger than 100 nm aggregate sizes D_f increases up to ~ 2.4 – 2.5
- An average D_f = 2.4 for the entire aggregate population is consistent with the universal lognormal σ_g of 1.89 +/- 0.08 of many diesel size distributions based on population dynamics modelling of random oxidative fragmentation and coagulation
- Effective density exhibits a sharp decrease up to aggregate sizes about 200 nm and then a more gradual variation in agreement with the compaction shown by the increase of the D_f up to ~ 2.4 – 2.5.

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- Colleagues at the APT Lab: I. Papageorgiou, S. Skopa