15th ETH-Conference on Combustion Generated Nanoparticles
June 26-29 2011

The microstructure of soot aggregates produced by the Combustion Aerosol Standard (CAST) generator

Athanasios G. Konstandopoulos[1,2], Emmanouil Daskalos[1], Nickolas Vlachos[1],

[1]Aerosol and Particle Technology Laboratory (APTL), Chemical Process Engineering Research Institute, Center for Research and Technology-Hellas (CPERI/CERTH), 6th km Charilaou-Thermi Rd. 57001, Thessaloniki, Greece
[2]Department of Chemical Engineering, Aristotle University, P.O. Box 1517, Thessaloniki 540 06, Greece

Introduction

In the present work we characterize the microstructure of soot nanoparticles produced by a well-known soot generator, the Combustion Aerosol Standard (CAST), which produces soot aggregates with selected size distributions in a repeatable manner, in a range of mobility diameters from 30 to 200 nm. To our knowledge these are the first systematic characterization results for the structure of CAST aggregates and they should be useful in future research where the effect of soot aggregate structure on the phenomena under investigation needs to be studied.

Nanoparticle Characterization

In order to achieve the unbiased sampling of the original aggregate structure, a thermophoretic sampler was designed and evaluated. The evaluation was based on digital image processing and analysis of electron microscopy images of the CAST aggregates. We reconstructed the size distribution of the particles through projected area aggregate analysis and it was found to be in good agreement with the results obtained with a Scanning Mobility Particle Sizer (SMPS), validating thus the thermophoretic sampler. Subsequently for each of the pre-set soot size distributions (with nominal mean diameter D) of the CAST we evaluated the actual mobility diameter D_{me}, the size of primary soot particles, the average number of primary particles per aggregate, <N>, the fractal dimension, D_f, the fractal prefactor, k_g, the filling factor, f, and the overlap coefficient, C_{ov} . The fractal dimension and fractal prefactor of the CAST soot aggregates are given in Table 1.

Table 1. Morphological properties of CAST soot aggregates

Nominal, D (nm)	D_{me} (nm)	D_f	k_g
190	*210*	2.25	3.95
143	*156*	2.27	3.69
128	*138*	2.14	4.28
106	*121*	2.13	6.69
91	*104*	2.03	5.25
60	*78*	2.15	4.97

The D_f results were compared with those obtained with an independent method based on combined inertial (ELPI) and mobility (SMPS) size measurements [1] and are shown to be in good agreement (Figure 1), while the k_g values are in agreement with those known for soot aggregates in the literature. To facilitate use of these results in computational studies involving the transport and deposition of soot aggregates a computational engine has been developed ("Digital CAST", d-CAST), allowing the generation of soot aggregates with the same microstructural properties as those found in this work (Figure 2).

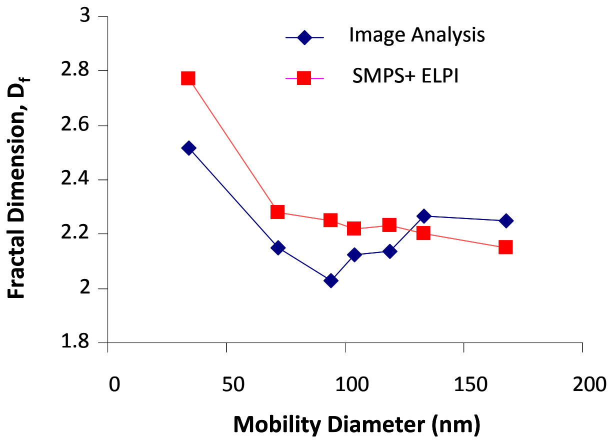


Figure 1. Fractal Dimension against Mobility Diameter obtained with two independent methods

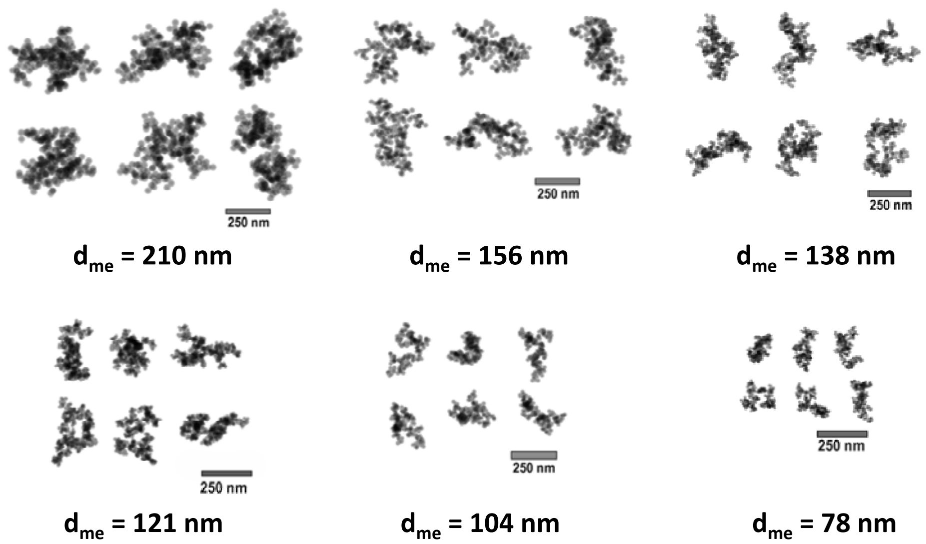


Figure 2. Digital-CAST (d-CAST) generated aggregates with same properties as actual CAST aggregates.

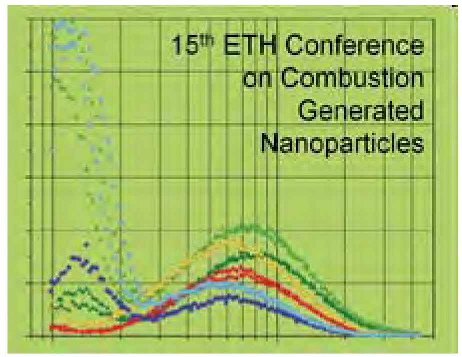
Conclusions

Soot aggregates produced at different operating points of the CAST burner have been characterized in terms of their structure and a database of D_f, k_g, C_{ov}, N_A, d_0 has been established. The morphological results obtained by image analysis were found to be in good agreement with an independent method based on parallel SMPS-ELPI measurements and a previously developed theory that allows the calculation of D_f from such measurements. The d-CAST software provided faithful aggregate particle morphologies that were used for ab initio simulations in a variety of applications, including filtration and deposition in realistic geometries.

References

1. J. Ristimäki, A. Virtanen, M. Marjamäki, A. Rostedt and J. Keskinen, On-line measurement of size distribution and effective density of submicron aerosol particles, Journal of Aerosol Science 33 (2002), pp. 1541–1557.

The microstructure of soot aggregates produced by the Combustion Aerosol Standard (CAST) generator



Athanasios G. Konstandopoulos[1,2],
Emmanouil Daskalos[1] and Nickolas Vlachos[1]

[1]Aerosol & Particle Technology Laboratory, CPERI/CERTH
[2]Department of Chemical Engineering, Aristotle University of Thessaloniki

Zurich, June 26th – 29th, 2011

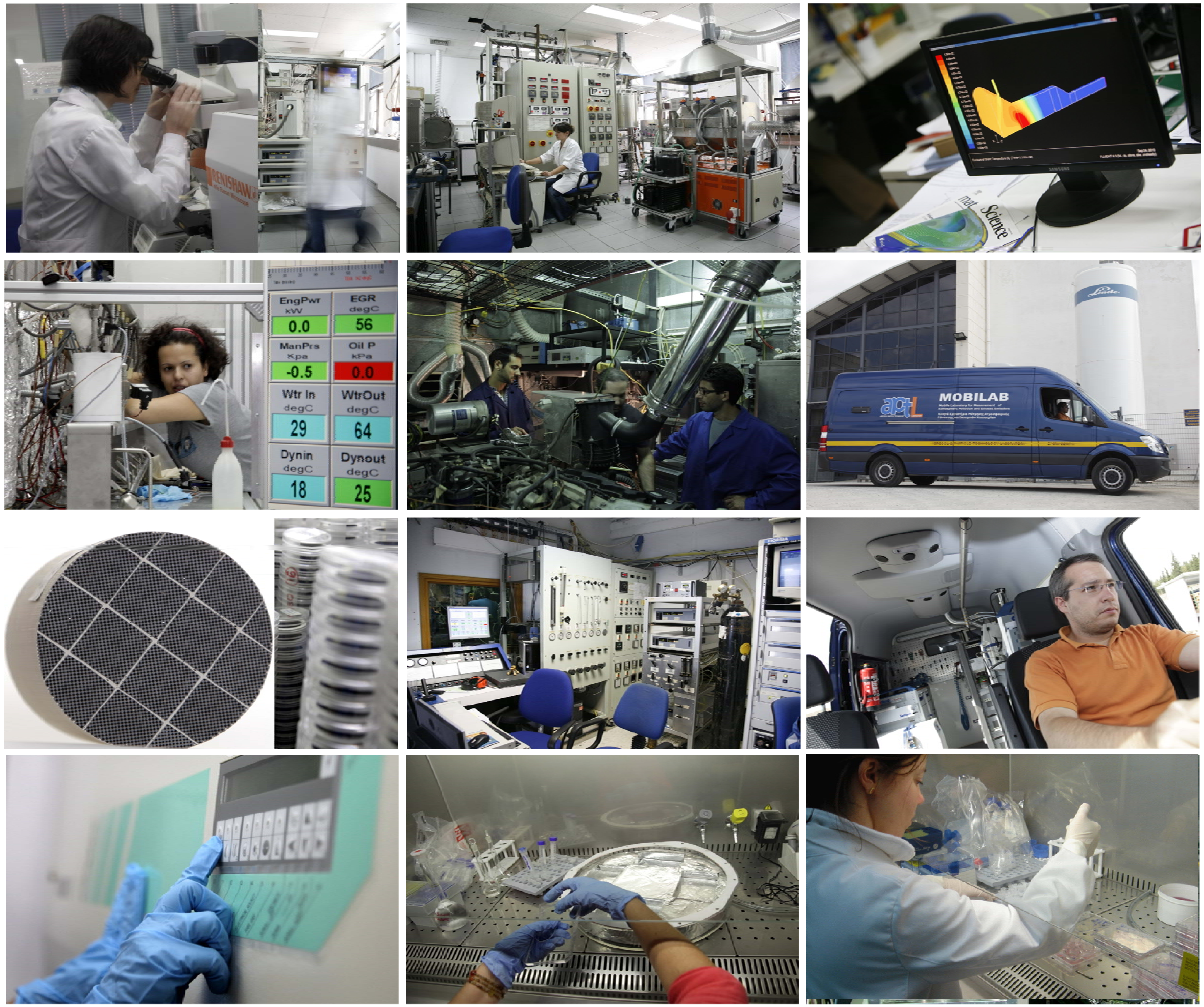
A Day in the Life of the Aerosol & Particle Technology Laboratory

15 YEARS

aptL

AEROSOL & PARTICLE TECHNOLOGY LABORATORY

1996-2011



Motivation

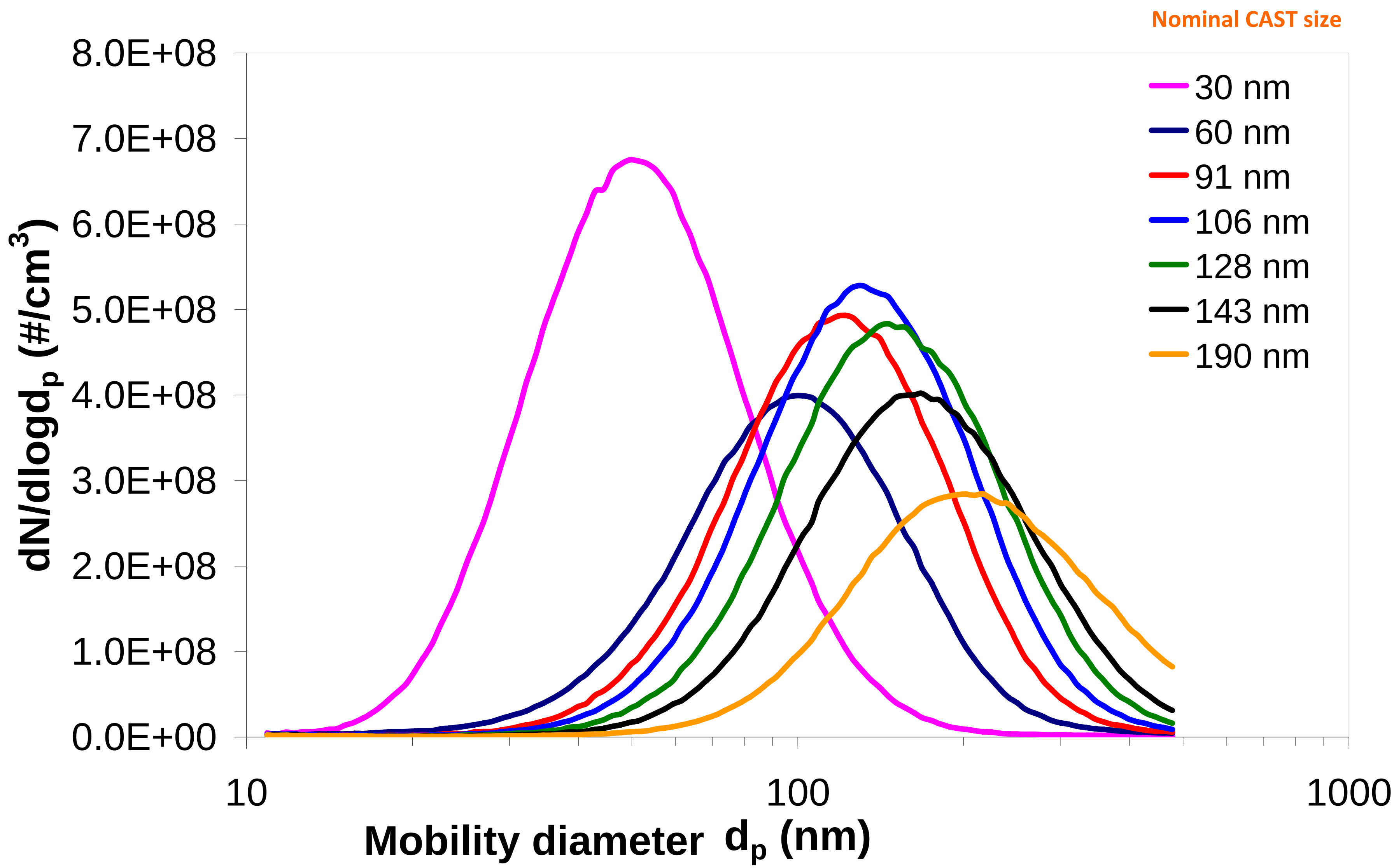
Structure of combustion generated aggregates is important for a number of applications

- 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

CAST: Combustion Aerosol Standard

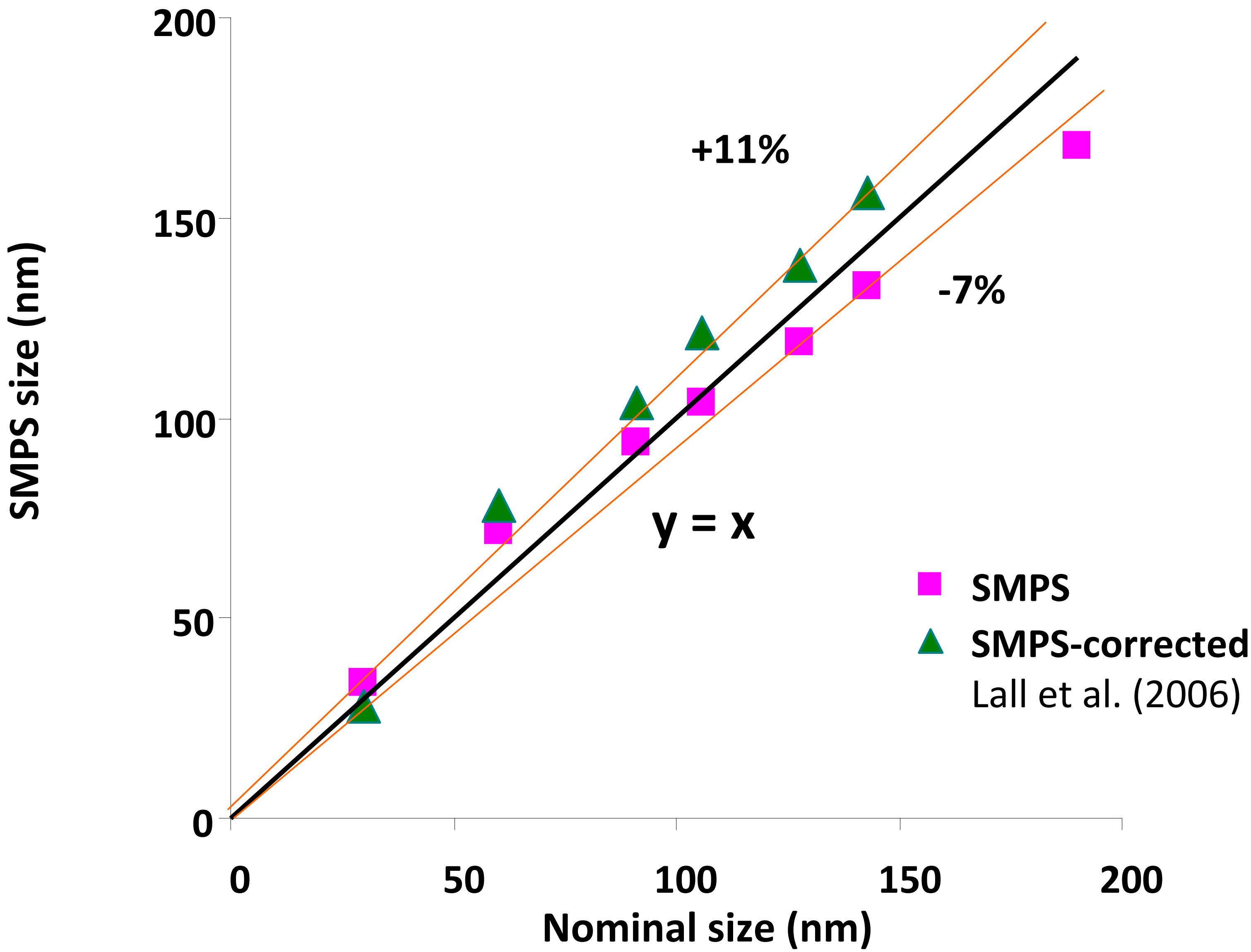
Provides Reference Soot Size Distributions but not enough quantitative data on their morphology exist

CAST Soot size distributions



A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

Nominal vs. Measured (SMPS) Size

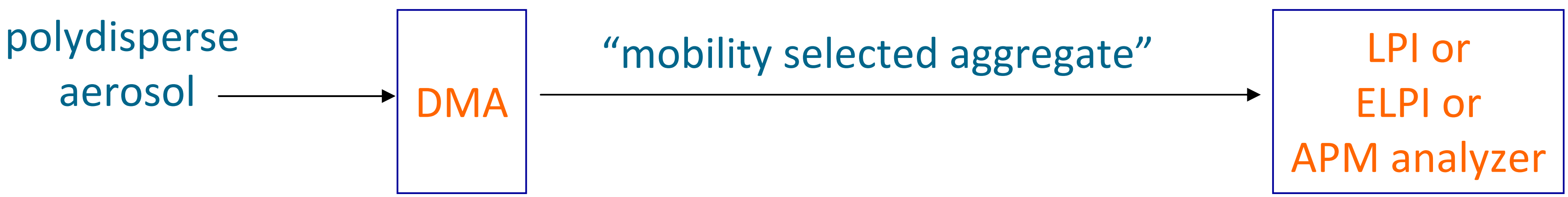


A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

Methods to Measure Aggregate Morphology

- 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

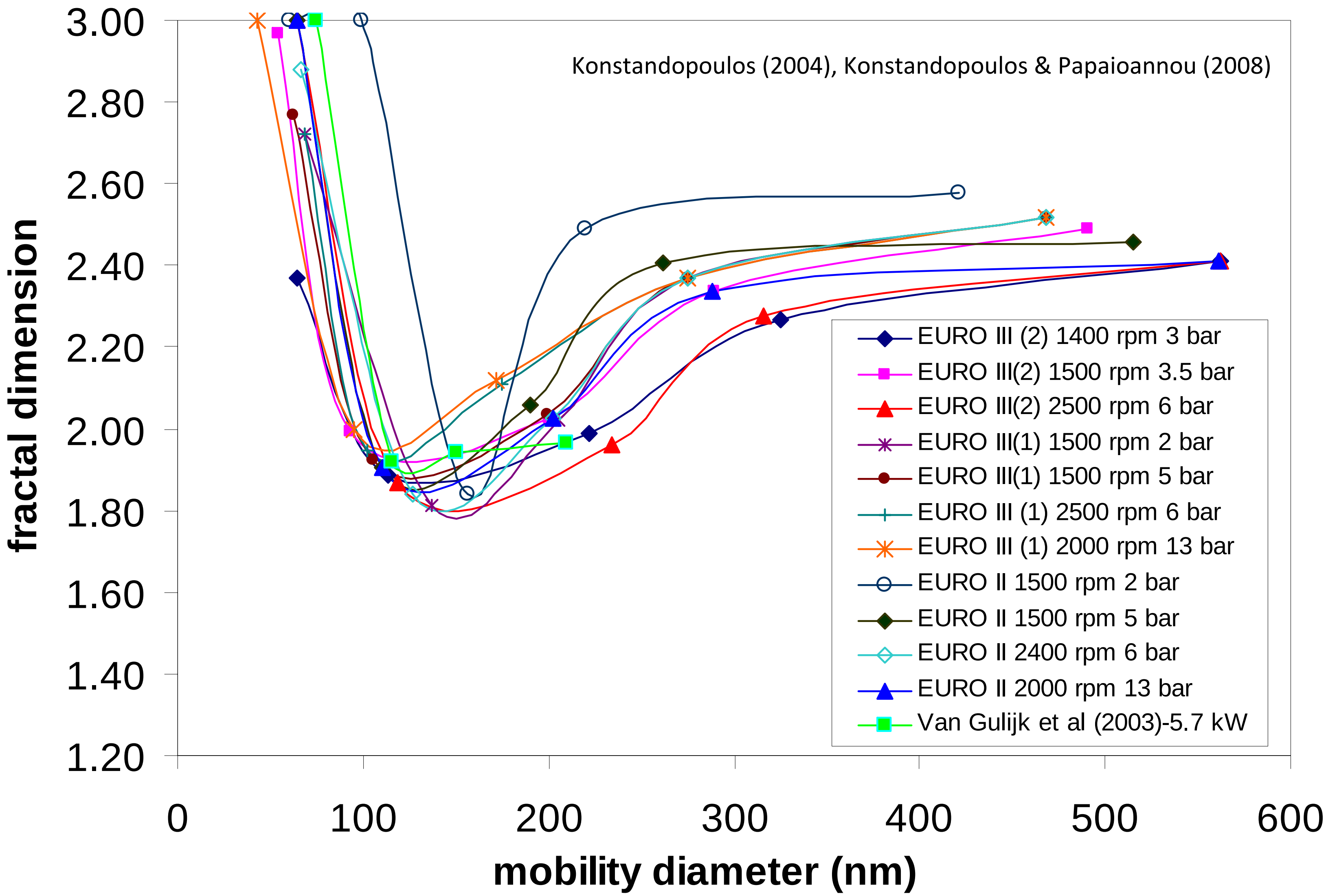
  Ristimaaki et al (2002), Present work

- Optical methods and microscopy

  Sorensen and co-workers (1995-2004), di Stasio et al. (1999-2004), Brazil et al. (1999), Wentzel et al. (2003), Present work

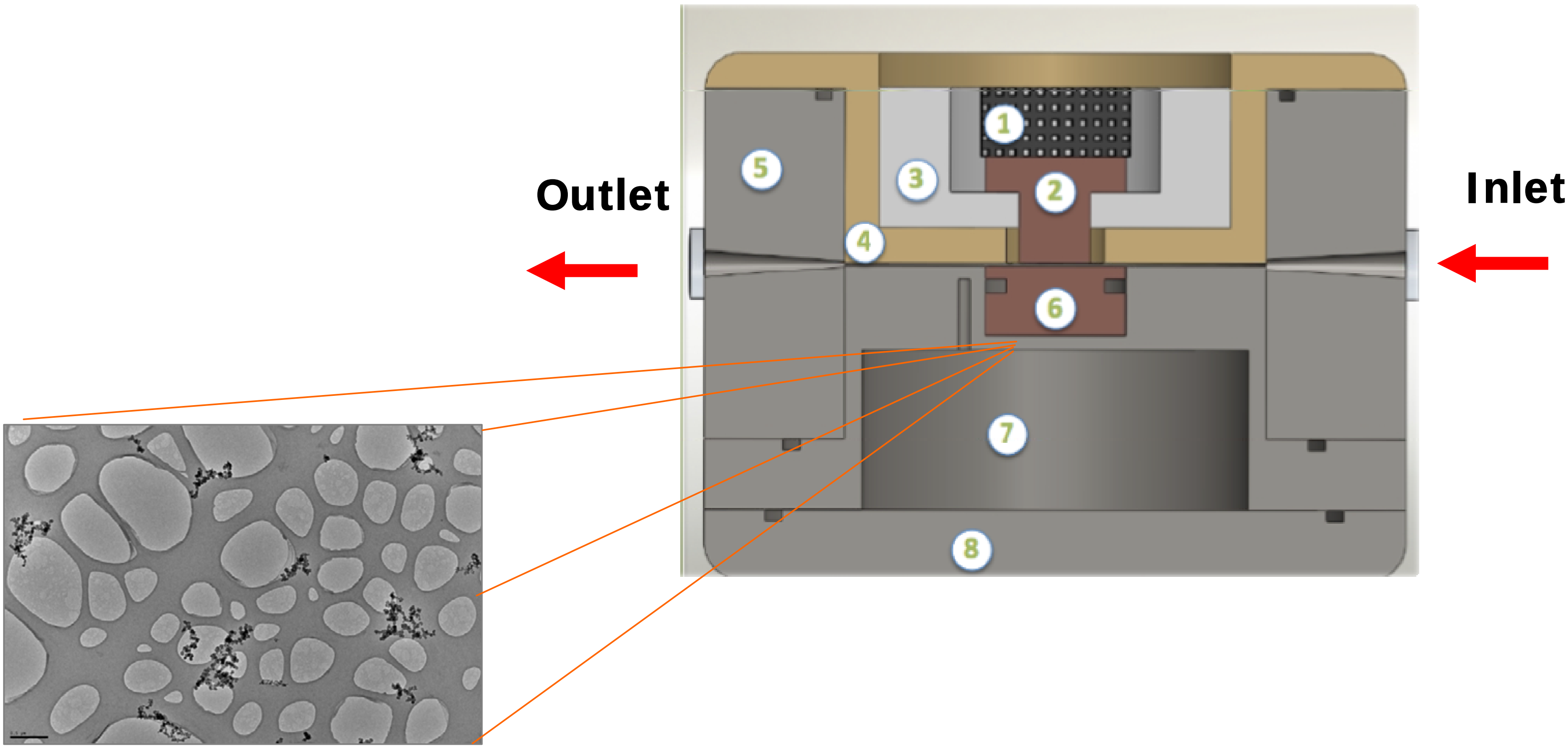
Example: Diesel Soot Characterization

Combined SMPS-ELPI Measurements



A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

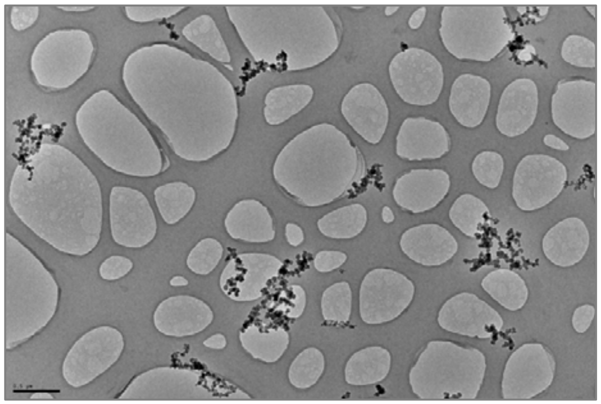
Thermophoretic Sampler

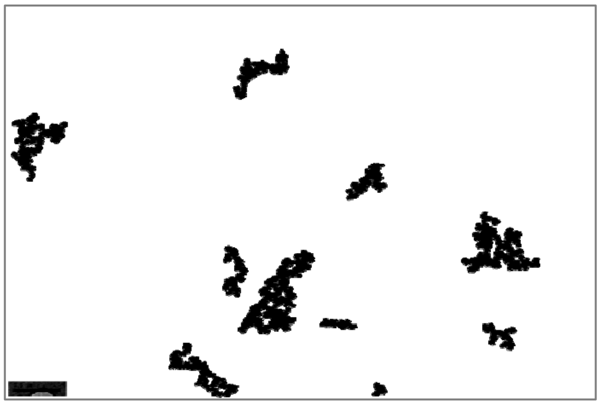


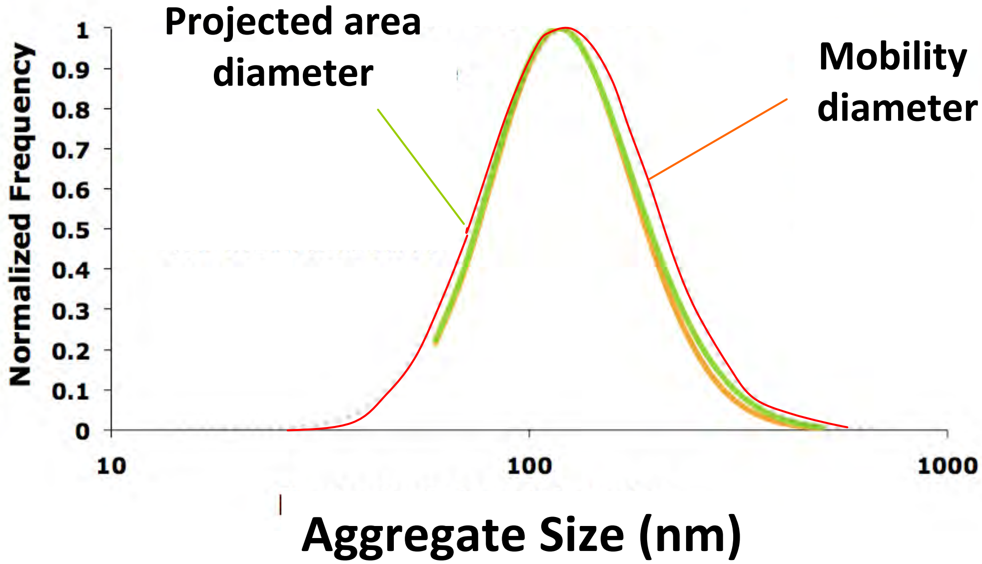
A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

Image Analysis

Aggregate Projected Area, Ap, d_{pa}



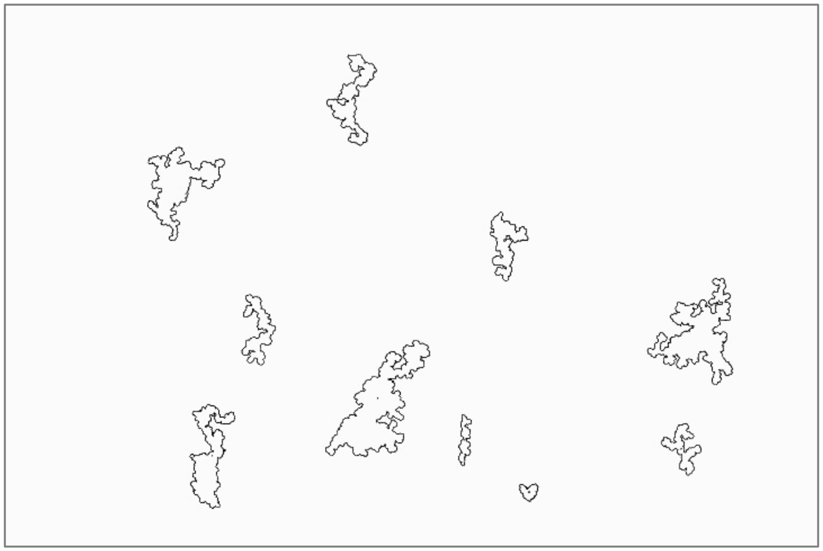




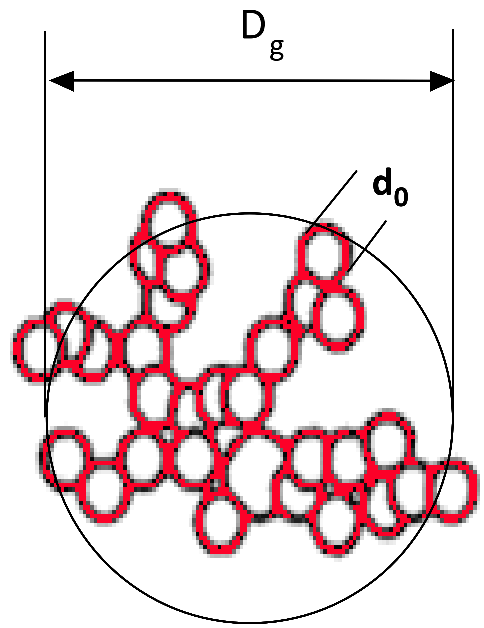
A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

From Projections to 3-D Structures

Aggregate Projected Area, A_p, d_{pa}



From 2-D to 3-D



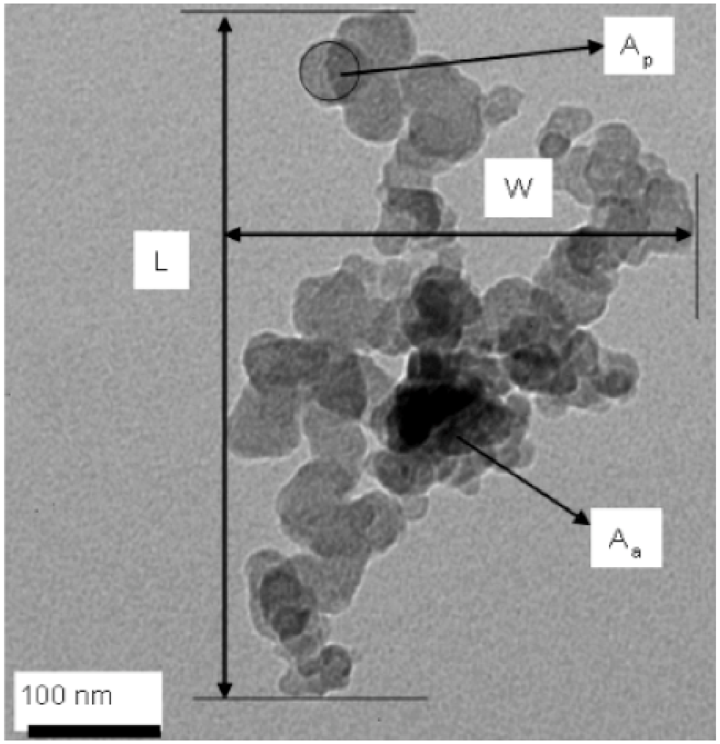
$$N_A = k_g \left[\frac{D_g}{d_0} \right]^{D_f}$$

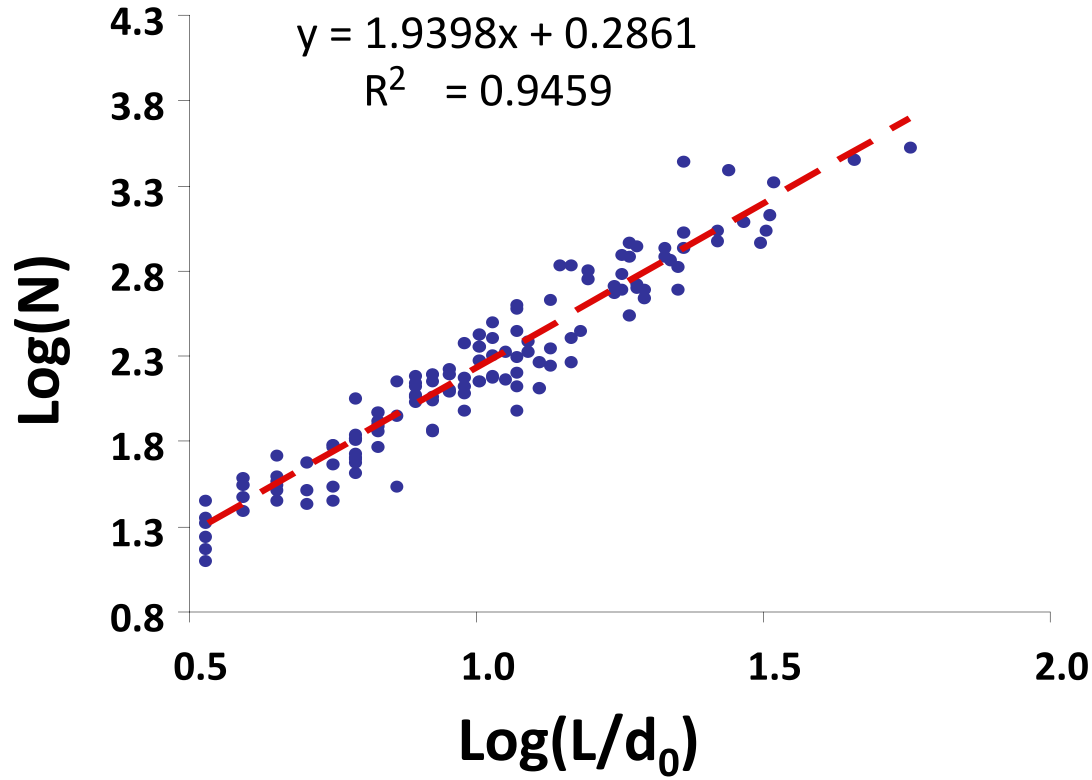
$$N_A = k_a \left(\frac{A_a}{< A_p >} \right)^a$$

Method as in:

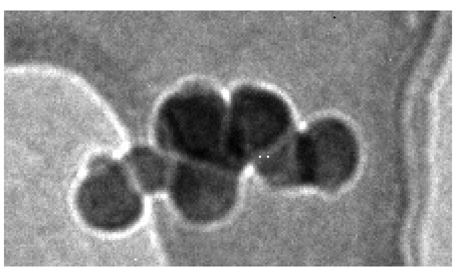
Oh and Sorensen (1997)
Brazil et al. (1999),
Wentzel et al. (2003)
Park et al. (2003)
and others

Scaling Analysis

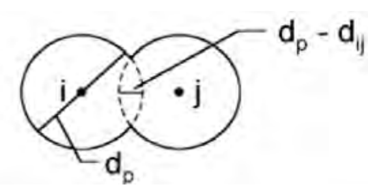




Computing overlap

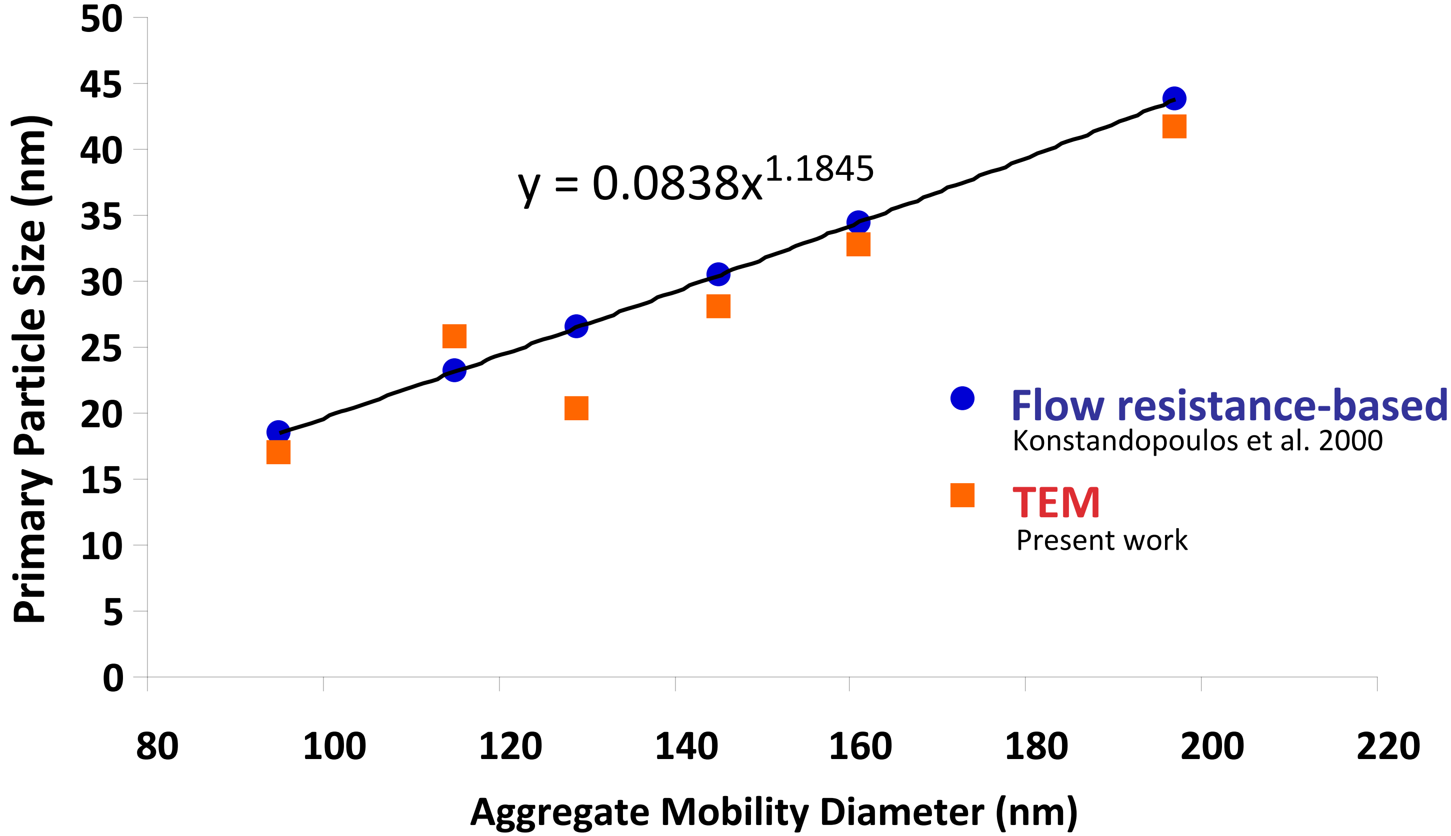


$$C_{ov} = \frac{d_p - d_{ij}}{d_p}$$



A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

CAST Primary vs. Aggregate particle size



A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

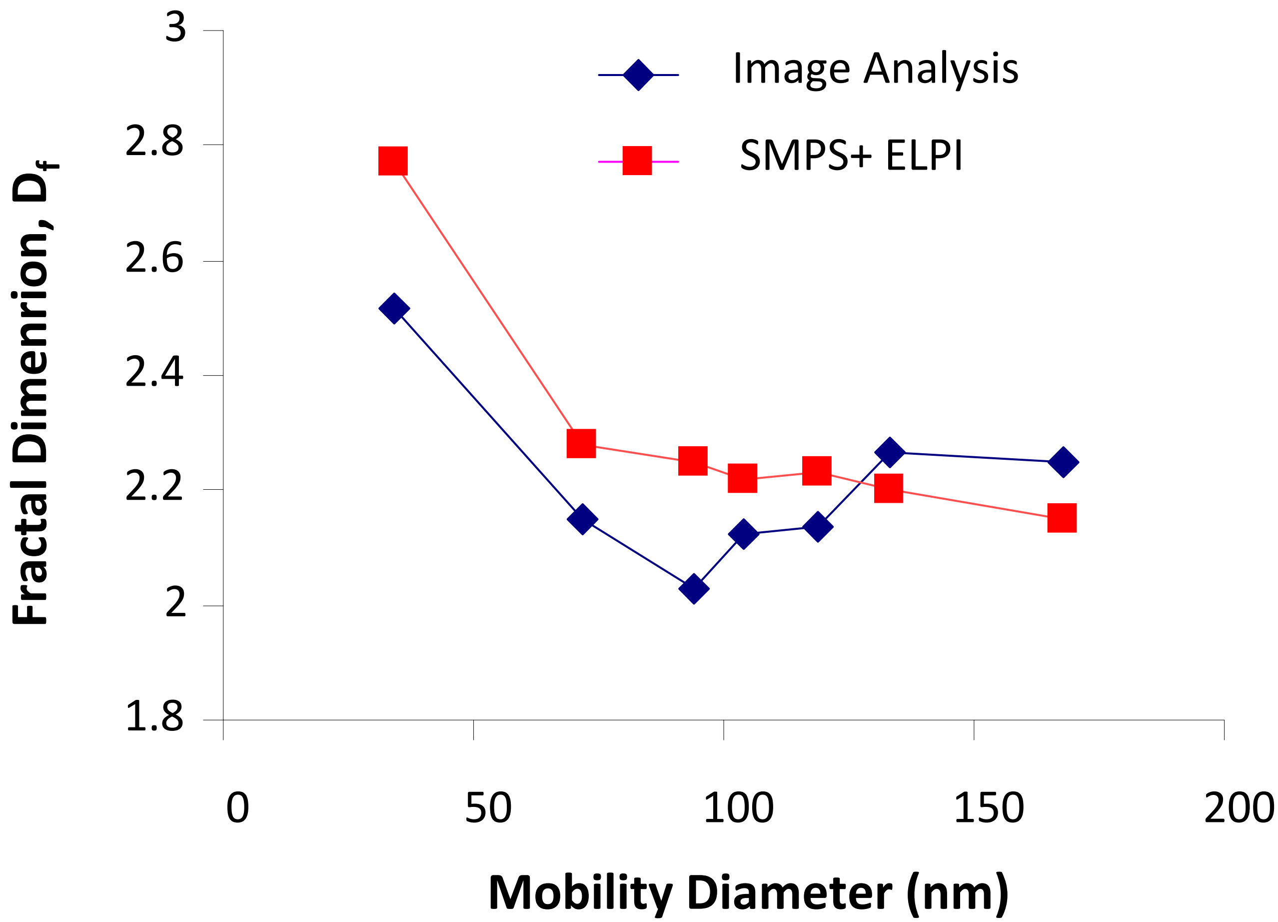
CAST Nanoparticles Characterization

Nominal, D (nm)	D_{me} (nm)	D_f	k_g	C_{ov}
190	*210*	2.25	3.95	0.36
143	*156*	2.27	3.69	0.34
128	*138*	2.14	4.28	0.40
106	*121*	2.13	6.69	0.52
91	*104*	2.03	5.25	0.48
60	*78*	2.15	4.97	0.44

$$D_g = d_0 \left[\frac{N_A}{k_g} \right]^{\frac{1}{D_f}}$$

A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

Comparison with Other Method

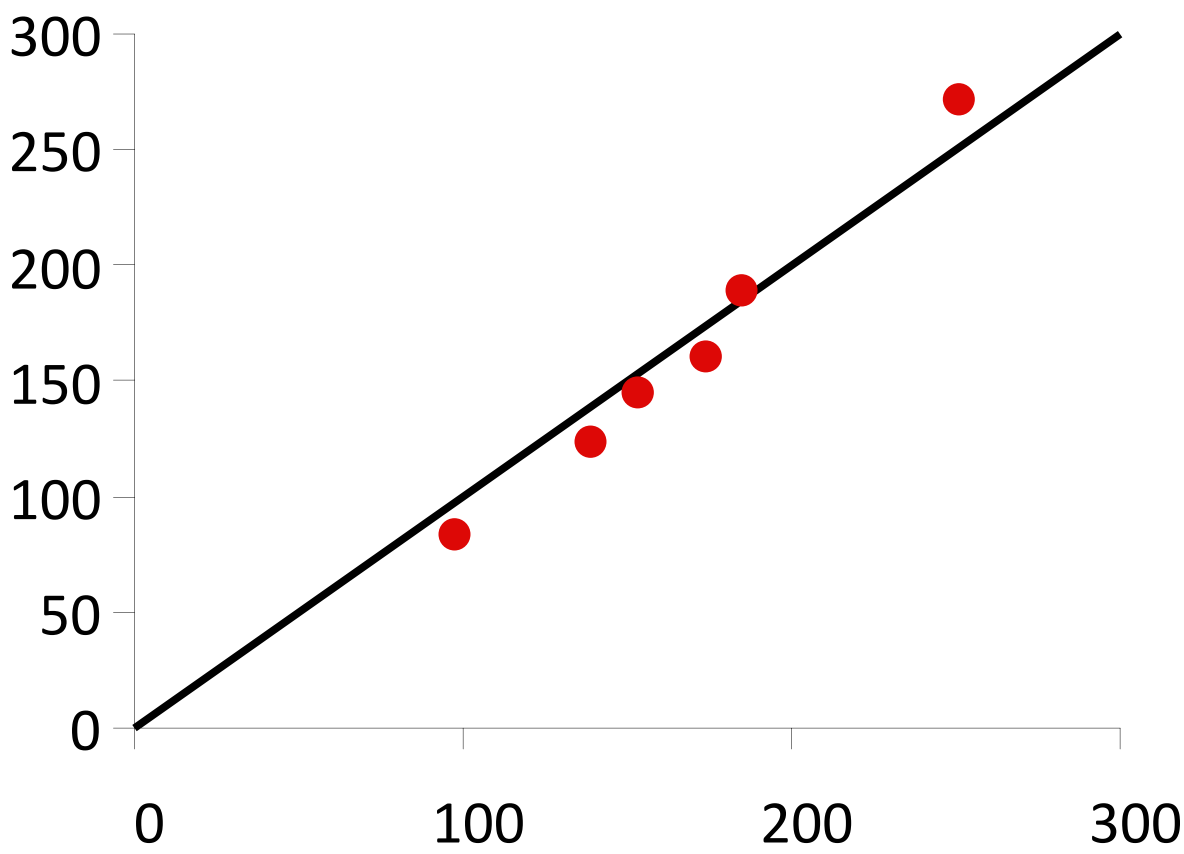


A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

Comparison with Theory

Gyroscopic Diameter from Image Analysis vs Computation from Mobility Diameter

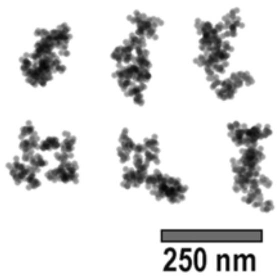
$$D_g = d_0\left[\frac{N_A}{k_g}\right]^{\frac{1}{D_f}}$$



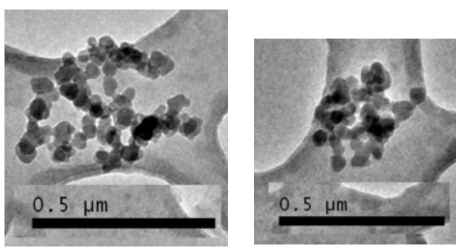
$$D_g = \frac{d_{me}}{(-0.06483D_f^2 + 0.6353D_f - 0.4898)\left(\frac{D_f}{D_f + 2}\right)^{1/2}}$$

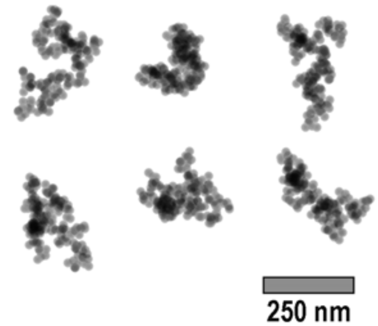
Wentzel et al. (2003)
Konstandopoulos (2003)

Digital Combustion Aerosol Standard: d-CAST

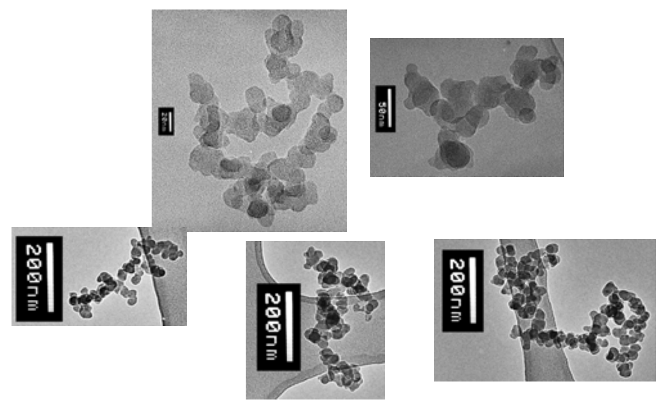


d_{me} = 78 nm

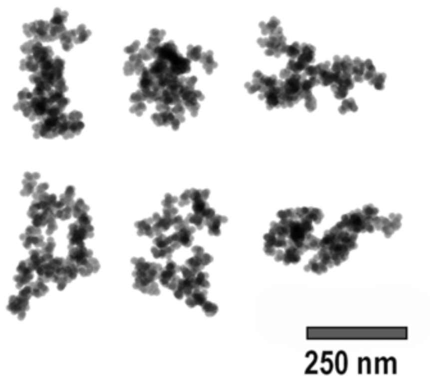




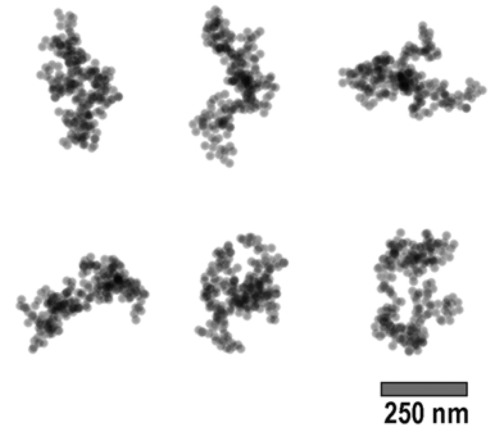
d_{me} = 104 nm



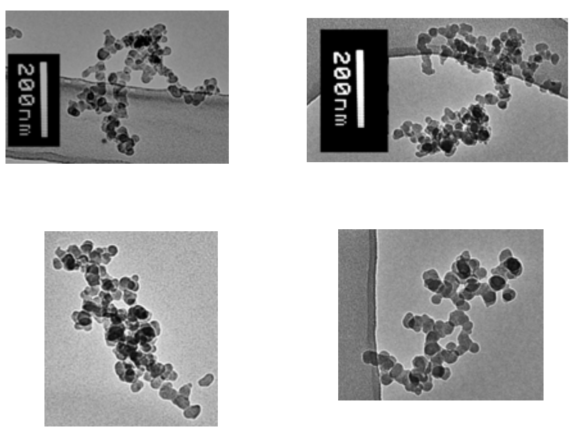
Digital Combustion Aerosol Standard: d-CAST

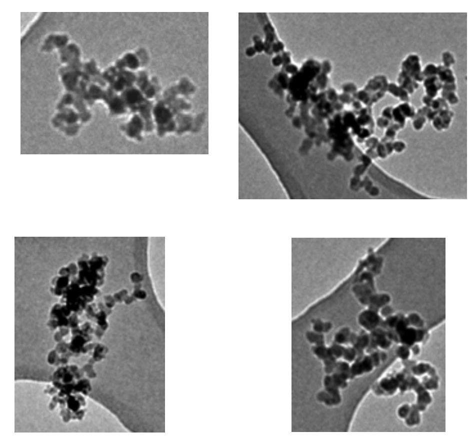


d_{me} = 121 nm



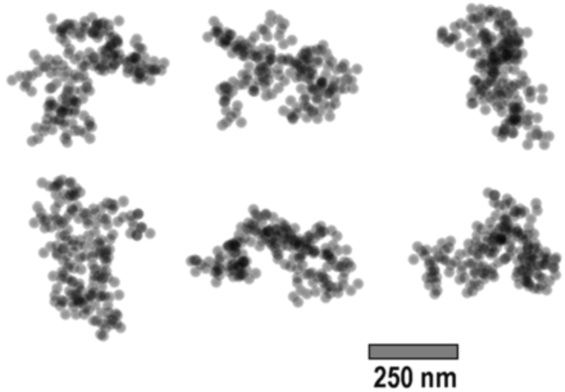
d_{me} = 138 nm



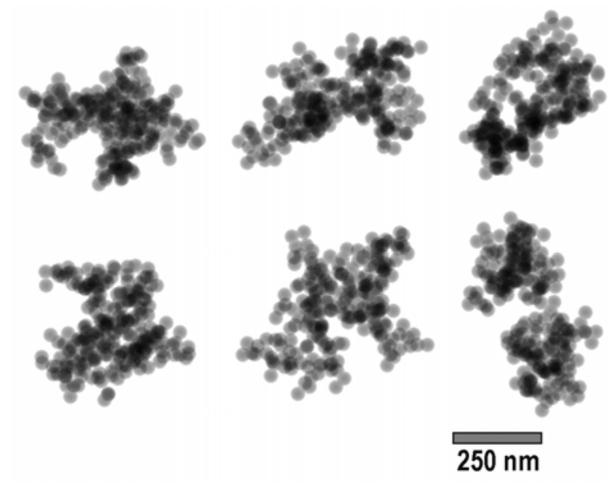


A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

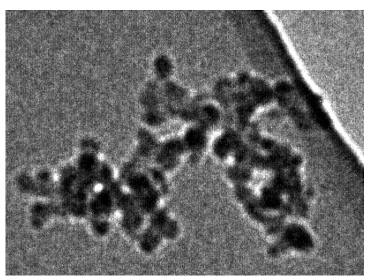
Digital Combustion Aerosol Standard: d-CAST

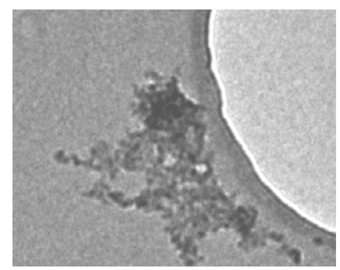


d_{me} = 156 nm

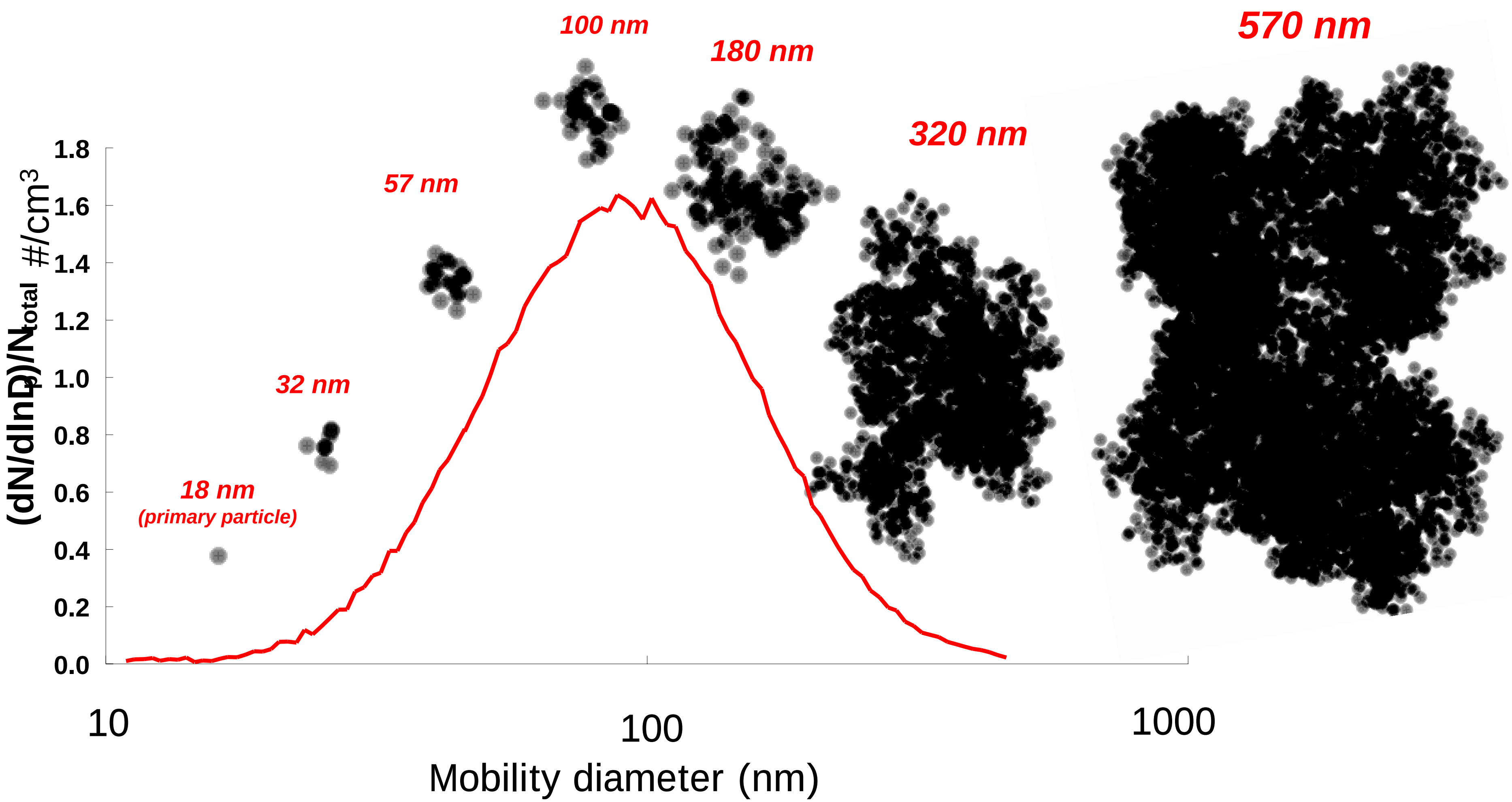


d_{me} = 210 nm



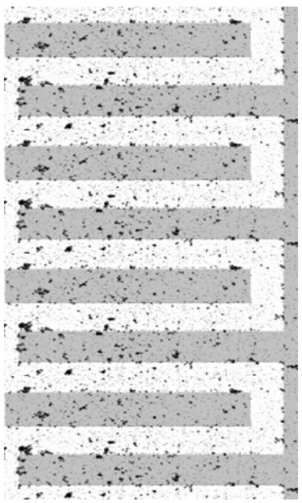


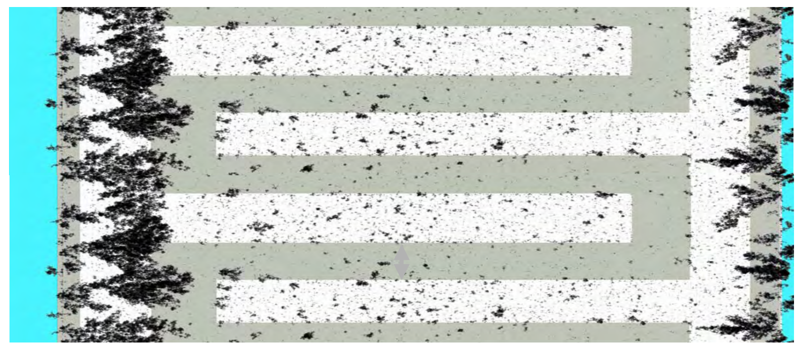
Employ Actual Soot Aggregates in Simulations



A. G. Konstandopoulos, 15 ETH Conference on Combustion Generated Nanoparticles

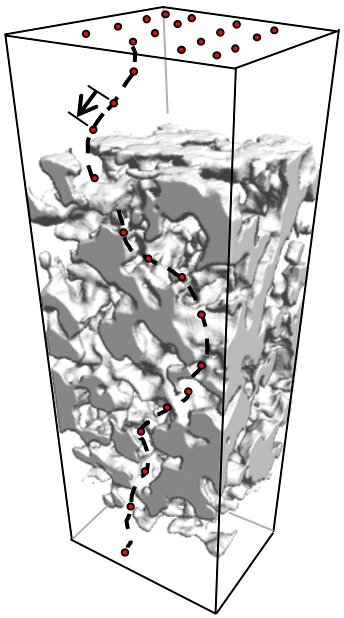
Example: Soot Accumulation on Interdigitated Sensor

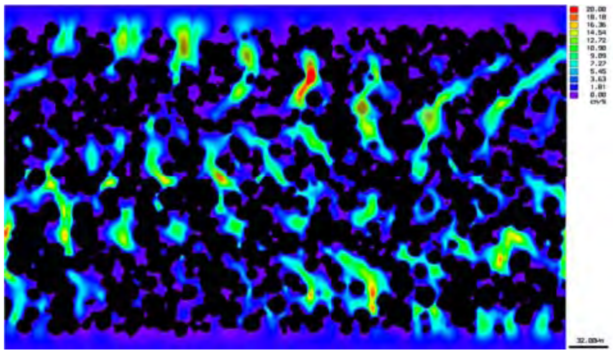


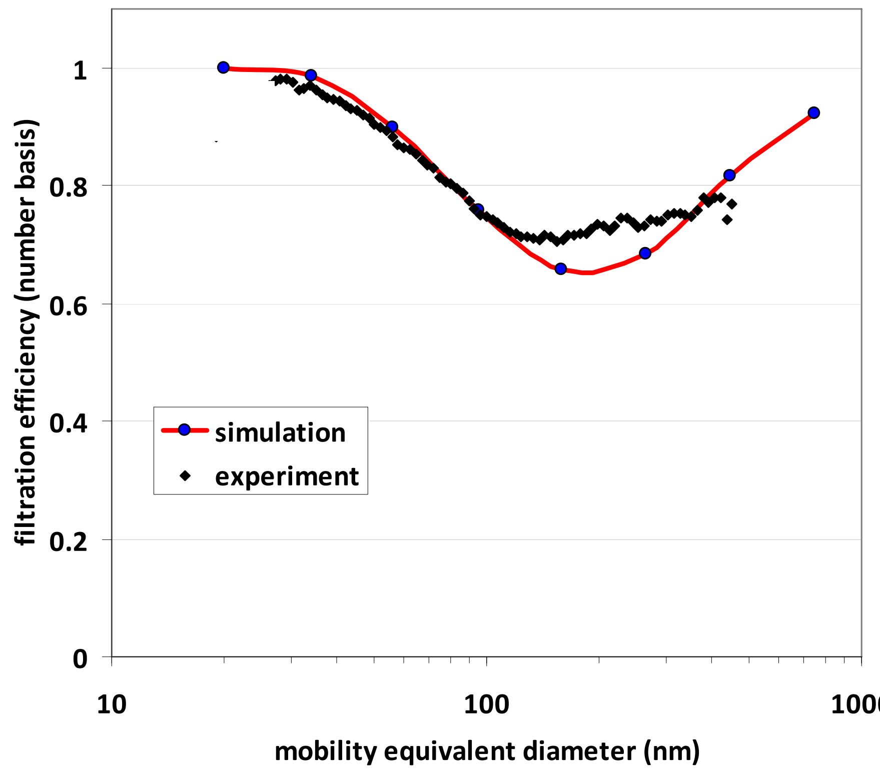


Example: Ab Initio Computation of Filtration Efficiency

See Poster by N. Vlachos, L. Chasapidis and A. G. Konstandopoulos







Conclusions

- Soot aggregates produced at different operating points of the CAST burner have been characterized in terms of their structure and a database of D_f, k_g, C_{ov}, N_A, d_0 has been established

- The morphological results obtained by image analysis are shown to be in good agreeement with an independent method based on parallel SMPS-ELPI measurements and a previously developed theory

- A Digital CAST (d-CAST) has been programmed to provide faithful aggregate particle morphologies for ab initio simulations in a variety of applications

- Current work focuses on extending the size specific CAST characterization in terms of chemical reactivity

Acknowledgements

- European Commission for partially supporting this work through the ATLANTIS project.
- Our colleagues at the APT Laboratory