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Paper title: Towards a detailed soot model for internal combustion engines

Authors: S. Mosbach[1], M. Celnik[1], M. Kraft[1,*], H. R. Zhang[2], S. Kubo[3], K.-O. Kim[4]

Affiliation: [1]University of Cambridge, United Kingdom
[2]University of Utah, USA
[3]Toyota Central R&D Labs. Inc., Japan
[4]Toyota Motor Corporation, Japan

[*]Corresponding author:
Dr. M. Kraft
Department of Chemical Engineering
University of Cambridge
Pembroke Street
Cambridge CB2 3RA, United Kingdom
Phone: +44(0)1223 762784
Fax: +44(0)1223 334796
E-mail: mk306@cam.ac.uk

Extended summary:

In this work, we integrate previously developed models for engine combustion and soot formation.

The engine code we consider is the Stochastic Reactor Model (SRM), which uses detailed chemistry and takes into account convective heat transfer and turbulent mixing. The main strength of the SRM is its capability of qualitatively predicting emission trends of CO, CO_2, NO_x, and unburnt hydrocarbons at reasonable computational cost of 1-2 hours per engine cycle. This enables convenient multi-cycle, sensitivity, and parameter studies.

As soot model, we use SWEEP, a population balance solver based on a Monte Carlo method. One of the most striking features of SWEEP is its ability to accommodate up to thousands of internal coordinates, or in other words highly detailed particle descriptions covering aggregate structure and chemical composition.

In order to couple the two codes, a detailed chemical kinetic mechanism describing the combustion of Primary Reference Fuels (PRFs, mixtures of n-heptane and iso-octane) is extended to include small Polycyclic Aromatic Hydrocarbons (PAHs) such as pyrene, which function as soot precursor species for particle inception in the soot model. The extended chemical kinetic mechanism contains 208 species, about 50 of which are involved in the soot precursor chemistry, and 1002 reactions. We validate the mechanism against a variety of experimental data sets for fuel-rich laminar flames obtained from literature.

The integrated model provides not only averaged quantities as functions of crank angle like soot mass, volume fraction, aggregate diameter, and the number of primary particles per aggregate for example, but also more detailed information such as aggregate and primary particle size distribution functions, and specifics about aggregate structure including images similar to those produced with Transmission Electron Microscopes (TEMs). In addition to that, the chemical composition of soot aggregates is modelled in quite some detail. Surface chemistry, including growth and

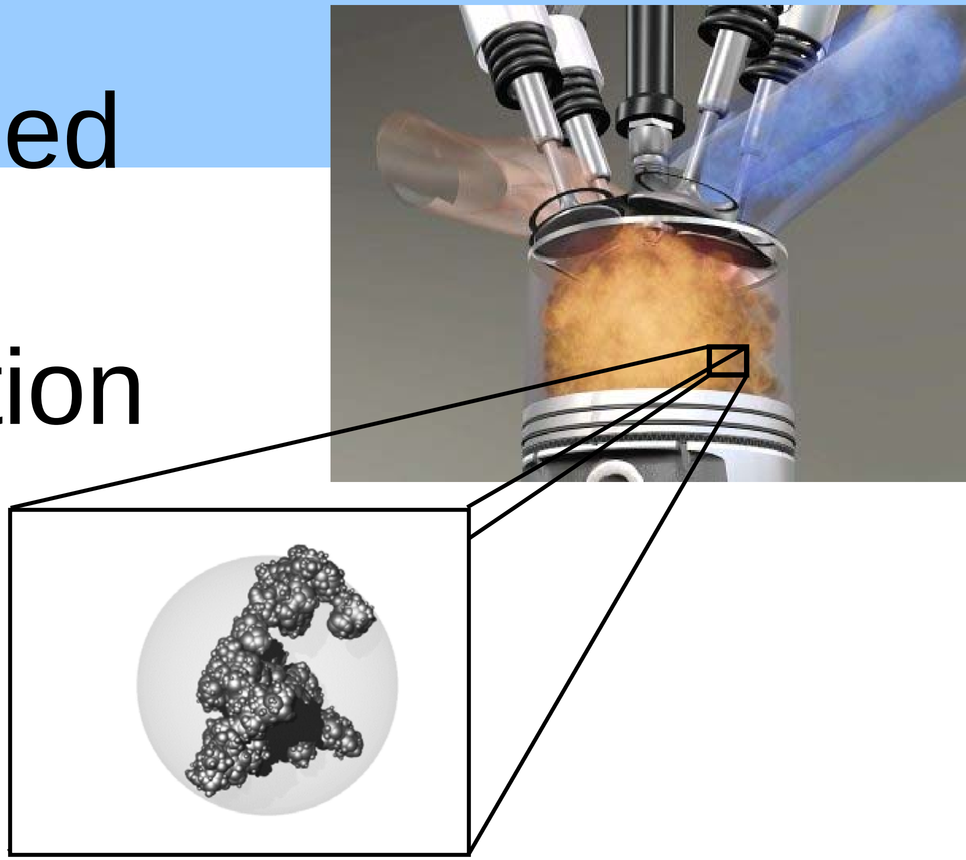
oxidation reactions at functional sites on the surface of particles, i.e. edges of PAHs, are taken into account. Since tracking every reaction of every molecule is computationally prohibitive, a statistical representation of PAHs and their functional sites is employed. This chemical description allows for example to plot distributions of aggregate C/H ratio and PAH ring count versus aggregate collision diameter.

The combined model is applied to simulate an n-heptane fuelled Homogeneous Charge Compression Ignition (HCCI) engine which is operated throttled at an equivalence ratio of 1.93 with an Exhaust Gas Recirculation (EGR) rate of about 20%. In-cylinder pressure and heat release predictions show satisfactory agreement with measurements. Particle-laden gases are extracted from within the cylinder through a snatch sampling valve and are analyzed by means of a Scanning Mobility Particle Sizer (SMPS) and a High-Resolution Transmission Electron Microscope (HR-TEM). We find that our simulated aggregate size distributions as well as their time evolution qualitatively agree with those obtained experimentally. It is also seen both in the experiment and in the simulation that soot emissions in terms of mass stem mostly from recirculated aggregates, whereas in terms of number mostly from newly formed ones. The simulation also shows that the largest aggregates are recirculated in the trapped residual gases for possibly several cycles before being emitted from the engine.

An important open question in soot research is the transition from pure gas-phase chemistry to the particulate phase, i.e. molecules held together in a particle through physical forces. In our model, two possible pathways from the gas-phase to the particulate phase are considered: inception, i.e. dimerization of pyrene molecules, and condensation, i.e. addition of a pyrene molecule taken from the gas-phase to an existing particle. We studied how the ratio between the rates of these two processes affects aggregate morphology. In line with expectation, we find that, if inception dominates, aggregates consist of large numbers of very small primary particles, whereas if condensation dominates, aggregates consist of comparatively small numbers of large primaries. We note that the peak of the aggregate size distribution early in the formation phase, which is found here well below 10 nm, moves towards larger sizes with increasing importance of condensation, while the collision diameter of the largest aggregates remains largely unaffected.

The present study focused on fully premixed engine operation. However, we have also taken first steps to extend this work towards operating modes which utilize direct injection such as partially stratified HCCI as well as conventional Compression Ignition Direct Injection (CIDI) engines. The time evolution of the cylinder charge in the Kamimoto diagram, i.e. in equivalence ratio/temperature phase space, proves particularly useful when analyzing emission formation for stratified operation.

Towards a detailed soot model for internal combustion engines



S. Mosbach, M. S. Celnik, M. Kraft,

H. R. Zhang, S. Kubo, K.-O. Kim

23 June 2008

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GROUP

Markus Kraft
mk306@cam.ac.uk

Engine model: SRM

Stochastic Reactor Model (SRM)

$$\frac{\partial}{\partial t}\mathcal{F}(\psi;t) = \underbrace{-\sum_{j=1}^{S+1}\frac{\partial}{\partial \psi_j}\left[G_j(\psi)\mathcal{F}(\psi;t)\right]}_{\text{chemical reactions, volume change}} + \underbrace{\sum_{j=1}^{S+1}\frac{\partial}{\partial \psi_j}\left[\frac{C_\phi}{2\tau}(\psi_j - \langle\psi_j\rangle)\mathcal{F}(\psi;t)\right]}_{\text{IEM mixing}} -$$

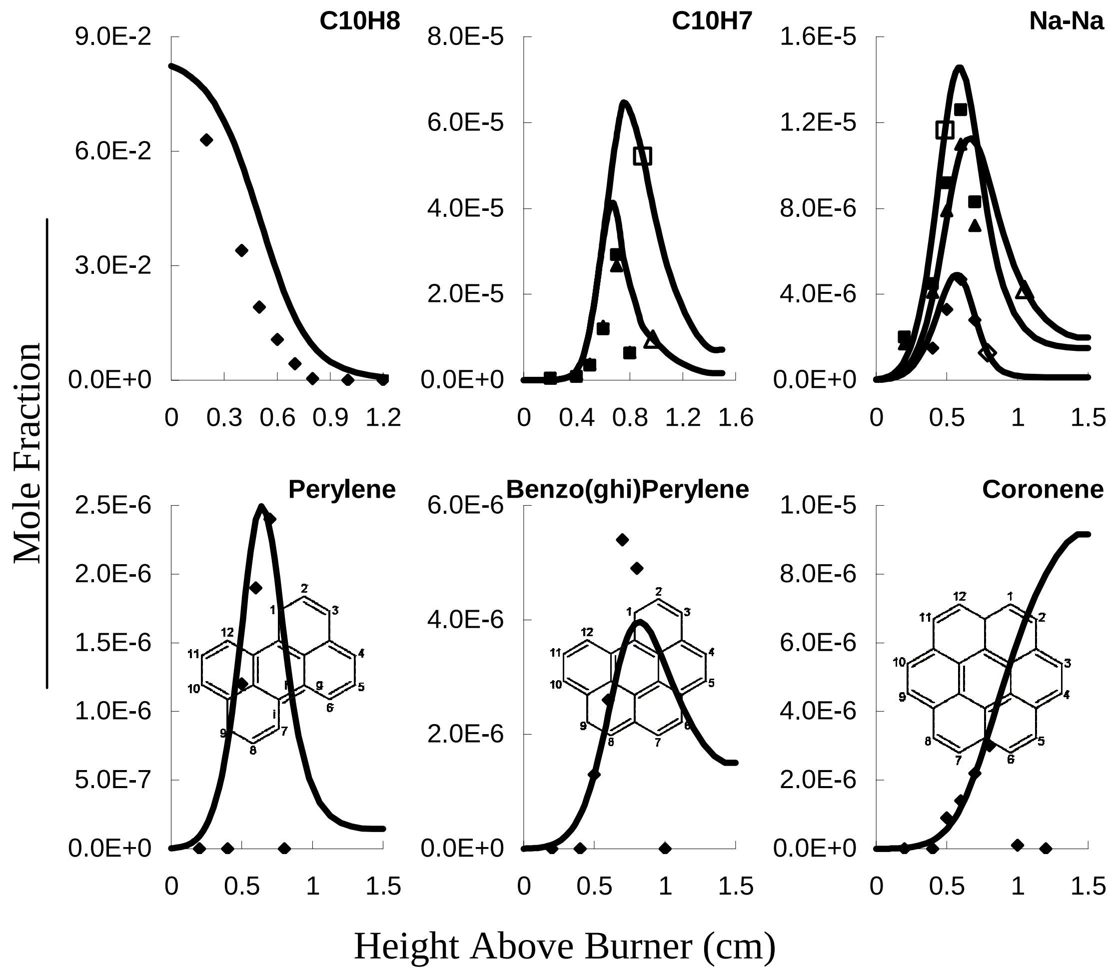
$$\underbrace{-\frac{\dot{V}}{V}\mathcal{F}(\psi;t)}_{\text{piston movement}} - \underbrace{\frac{1}{h}\left[U(\psi_{S+1}+h)\mathcal{F}(\psi_1,\ldots,\psi_S,\psi_{S+1}+h;t) - U(\psi_{S+1})\mathcal{F}(\psi;t)\right]}_{\text{heat transfer}}$$

- Detailed chemical kinetics → Chemical mechanism: PRF + small aromatics (extended by H. R. Zhang) 208 species, 1002 reactions
- Turbulent mixing
- Convective heat transfer
- Computationally cheap (1-2 CPU-hrs/cycle)

Markus Kraft
mk306@cam.ac.uk

PAHs in gas-phase chemistry

- Hongzhi R. Zhang
- Before: PRF+NOx, 157 species
- After: PRF+NOx+ variety of PAHs and highly unsaturated HCs, 208 species
- Validation against fuel-rich flame experiments



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GROUP

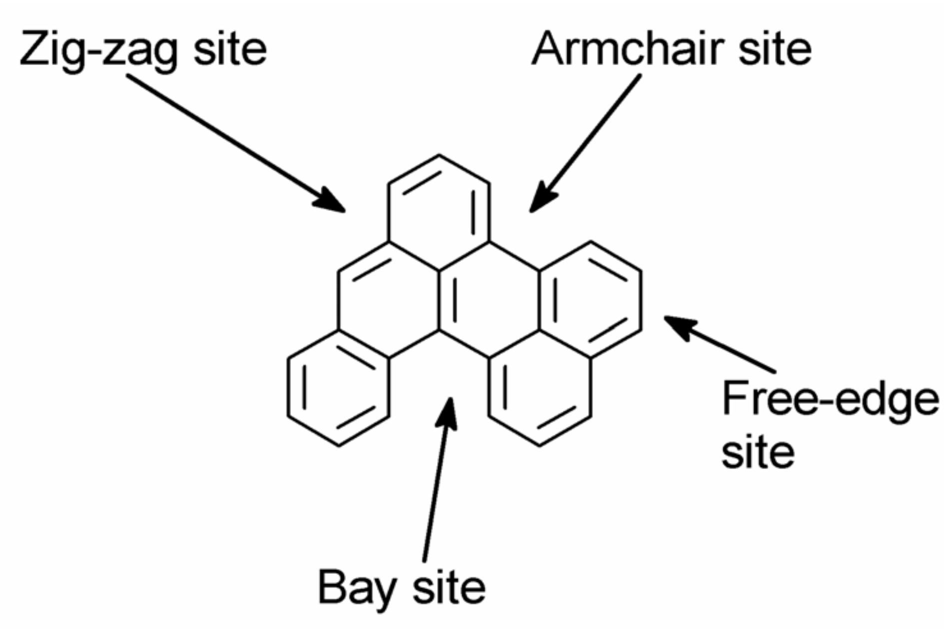
Markus Kraft
mk306@cam.ac.uk

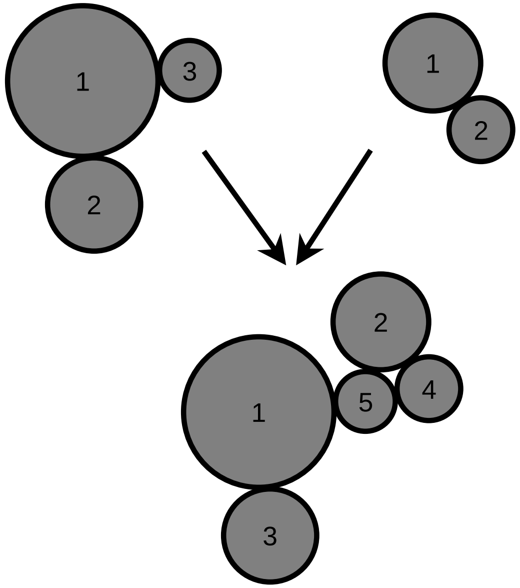
Soot model: site-counting

Describe soot particles by $9+N$ dimensional state space (ARS-SC-PP model):

$$E = \left(C, H, S_a, N_{ed}, N_{zz}, N_{ac}, N_{bay}, N_{R5}, N_{PAH}, PP_{(1\text{-}N)}\right)$$

PP = primary particle list





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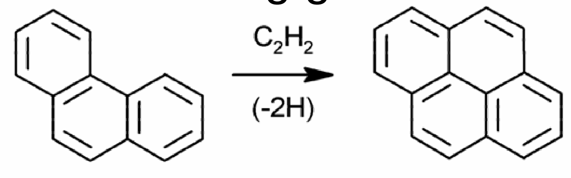
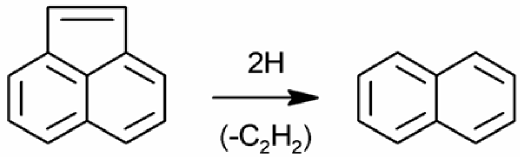
Markus Kraft
mk306@cam.ac.uk

PAH reaction steps

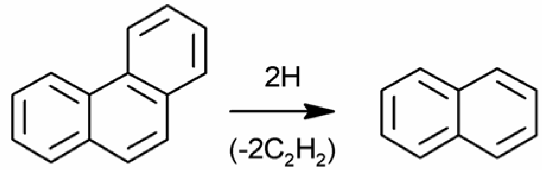
Armchair ring growth



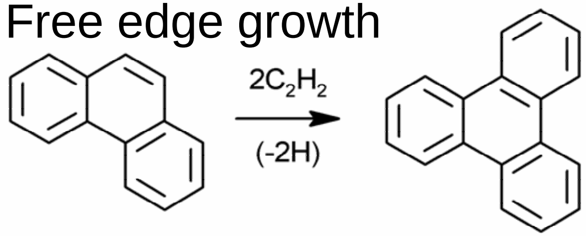
5-member ring desorption



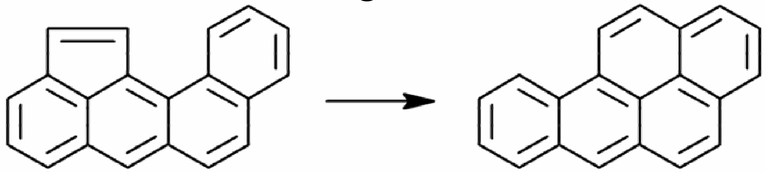
6-member ring desorption



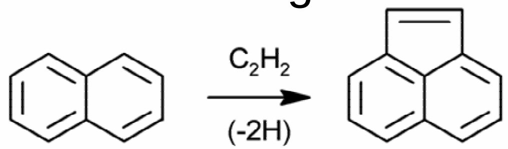
Free edge growth



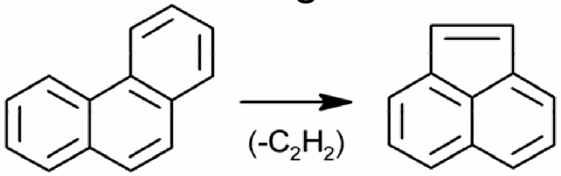
5-member ring conversion at AC



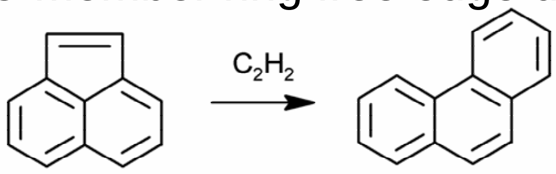
5-member ring addition



6- to 5-member ring conversion

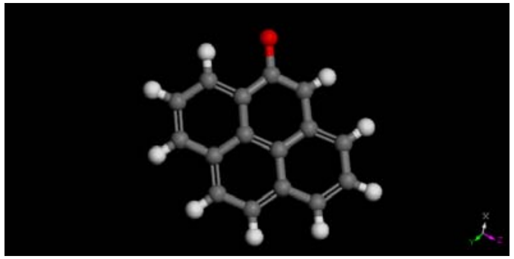


5-member ring free edge desorption



Frenklach, Schuetz, Ping. *Proc. Combust. Inst.* 30, 2005

Oxidation steps:
rates from
quantum chemistry



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Markus Kraft
mk306@cam.ac.uk

Soot in engines!

- HCCI, n-heptane
- Compression ratio 12
- Equivalence ratio 1.93
- Throttled, 20% EGR

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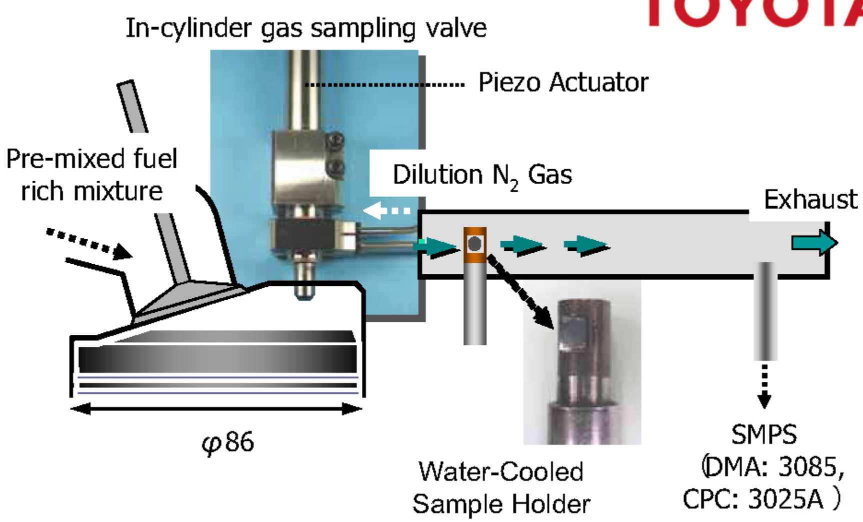

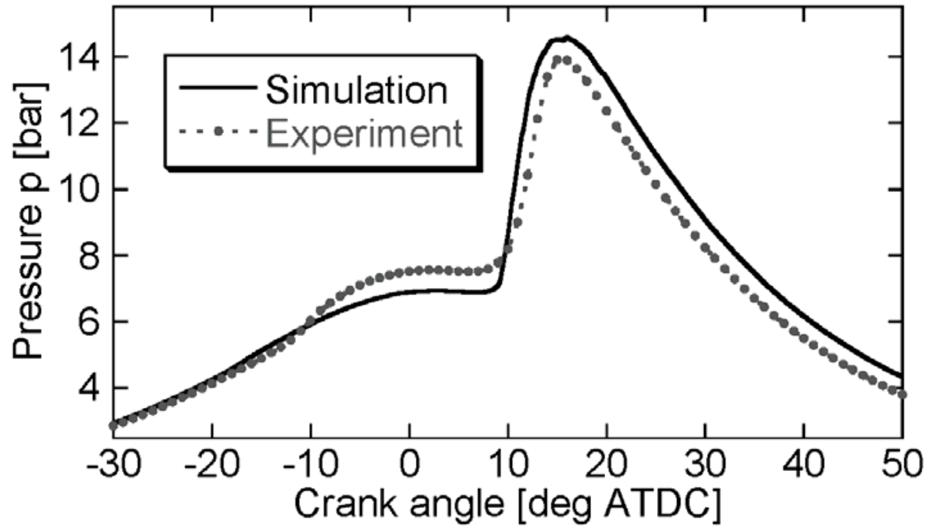

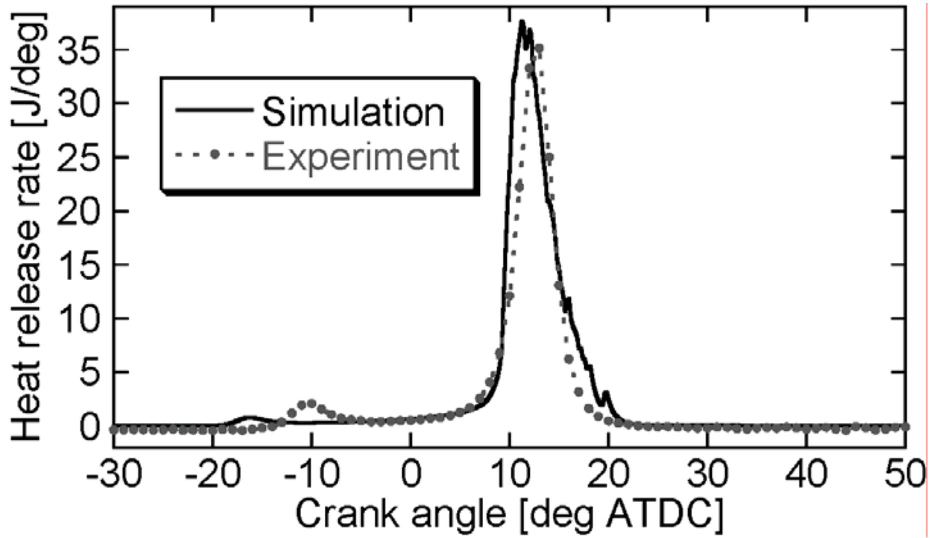

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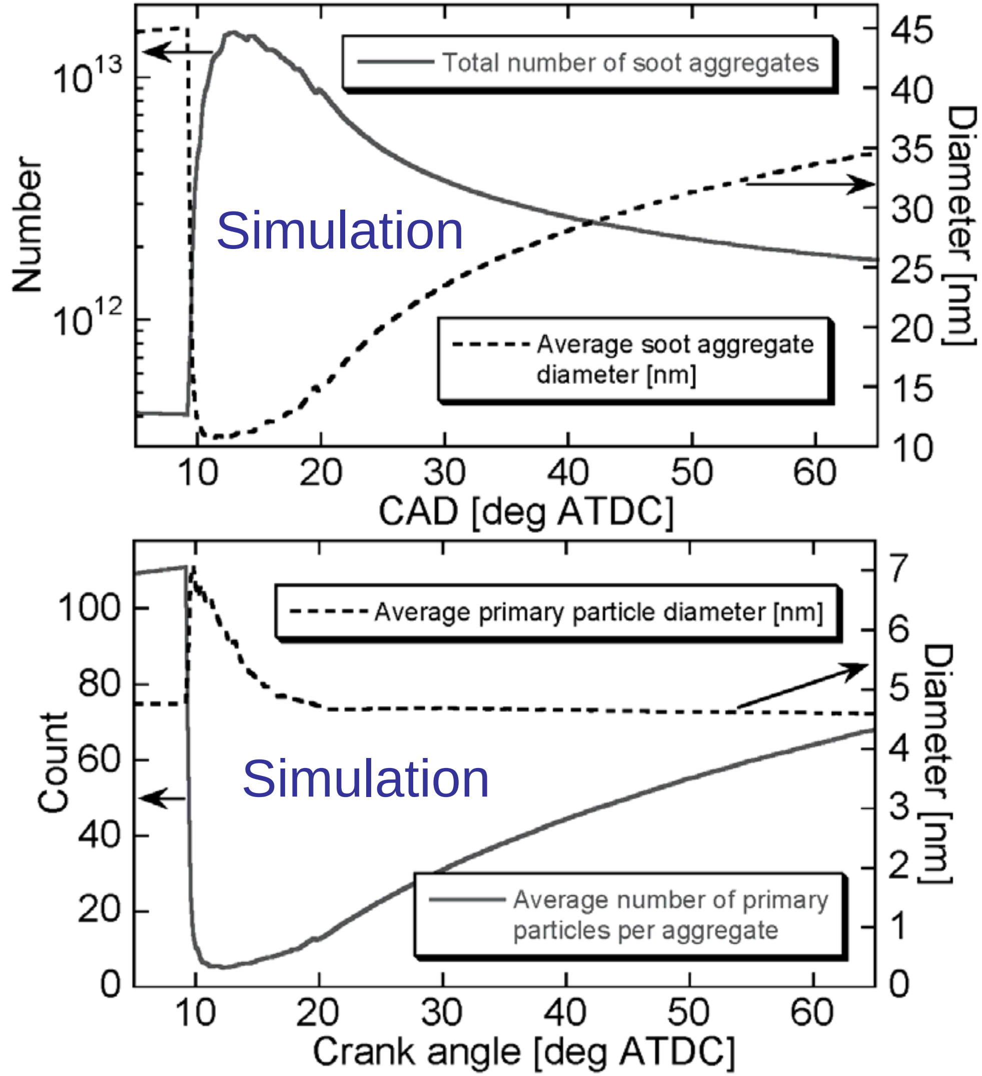
Markus Kraft
mk306@cam.ac.uk

Averaged soot quantities



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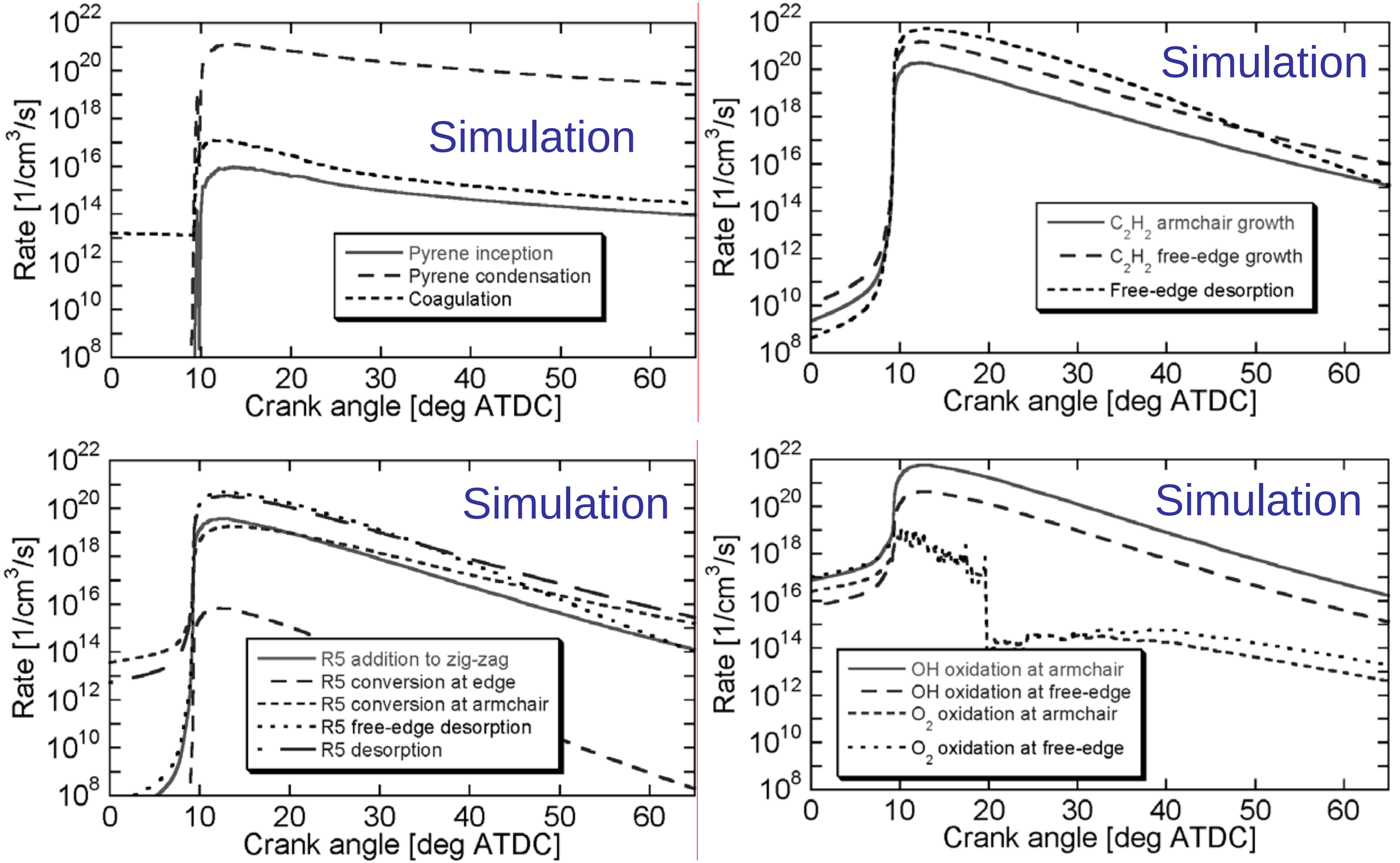
Markus Kraft
mk306@cam.ac.uk

Rates of soot processes



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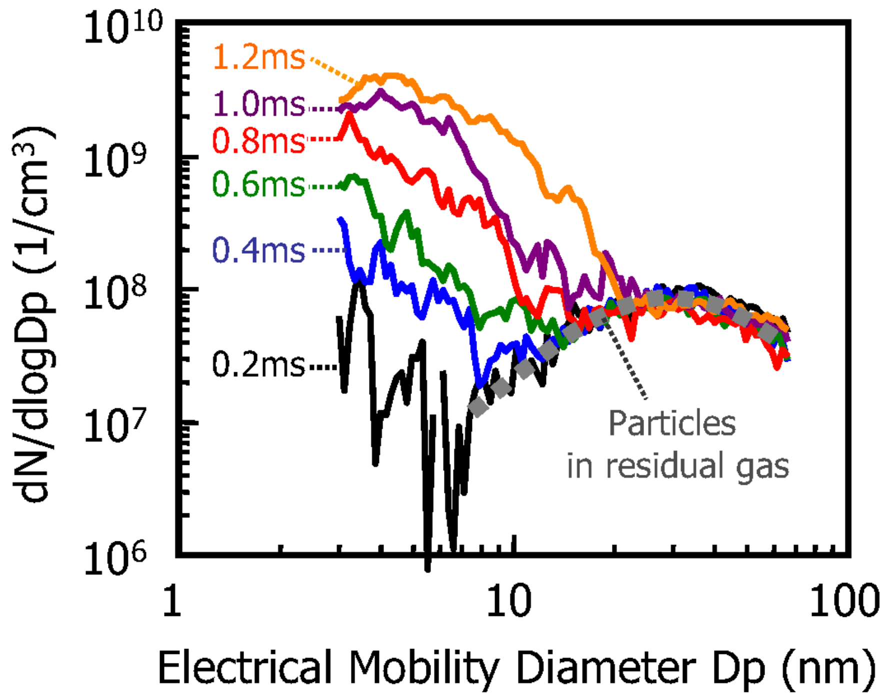
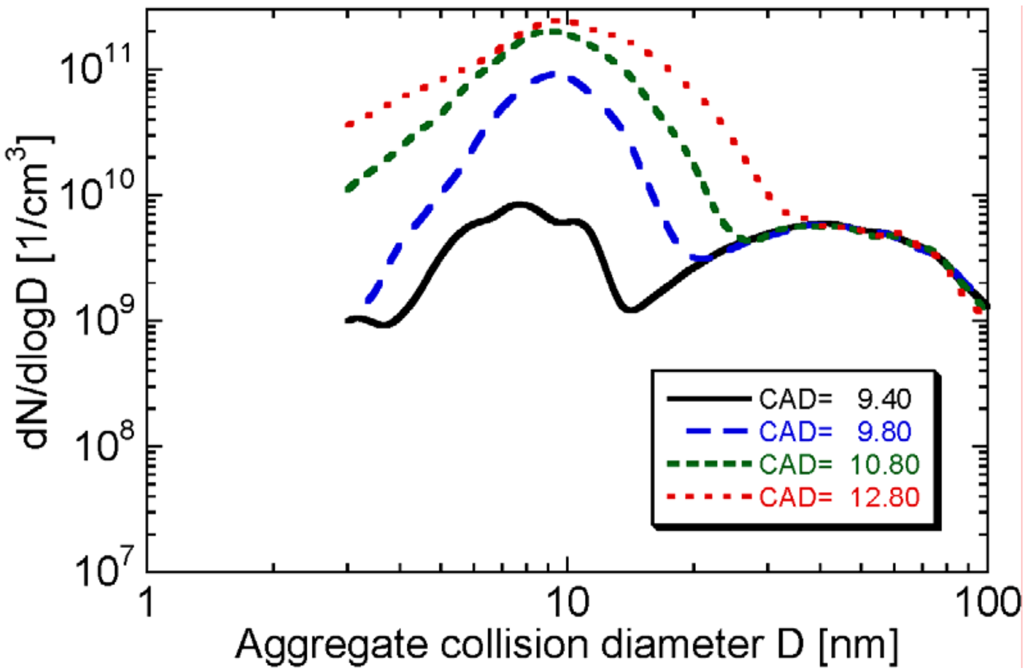
mk306@cam.ac.uk

Aggregate size distributions (I)

Experiment

Simulation





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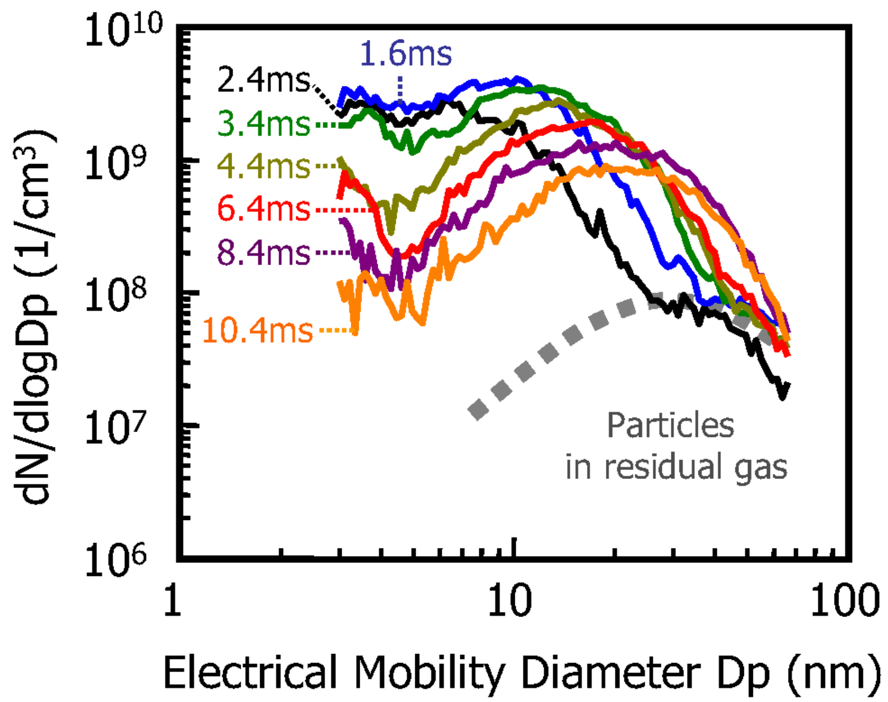
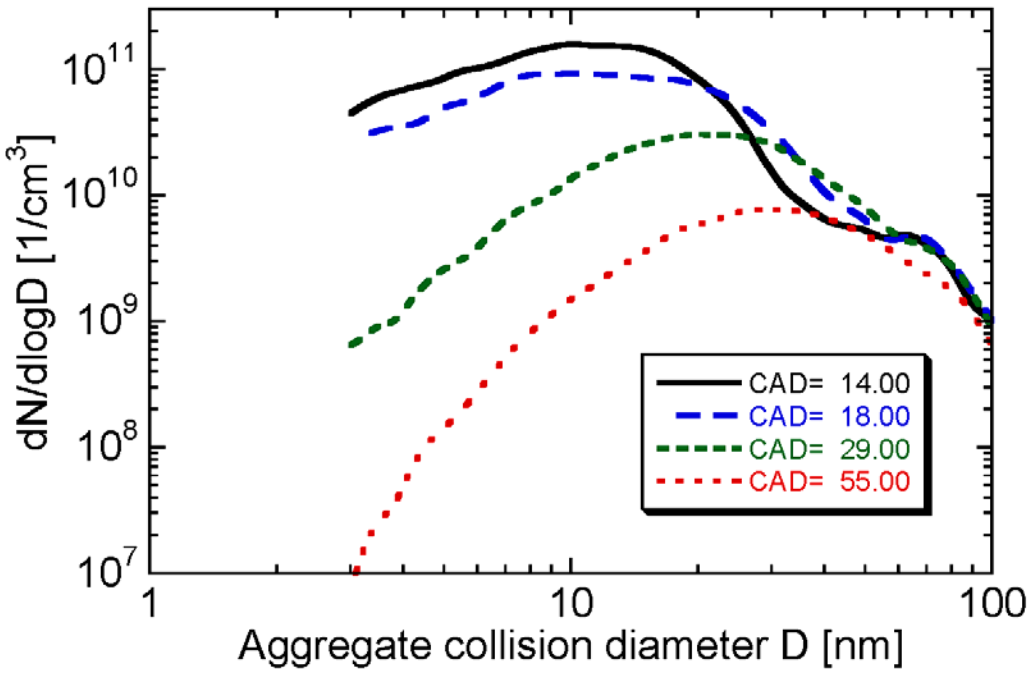
Markus Kraft
mk306@cam.ac.uk

Aggregate size distributions (II)

Experiment

Simulation





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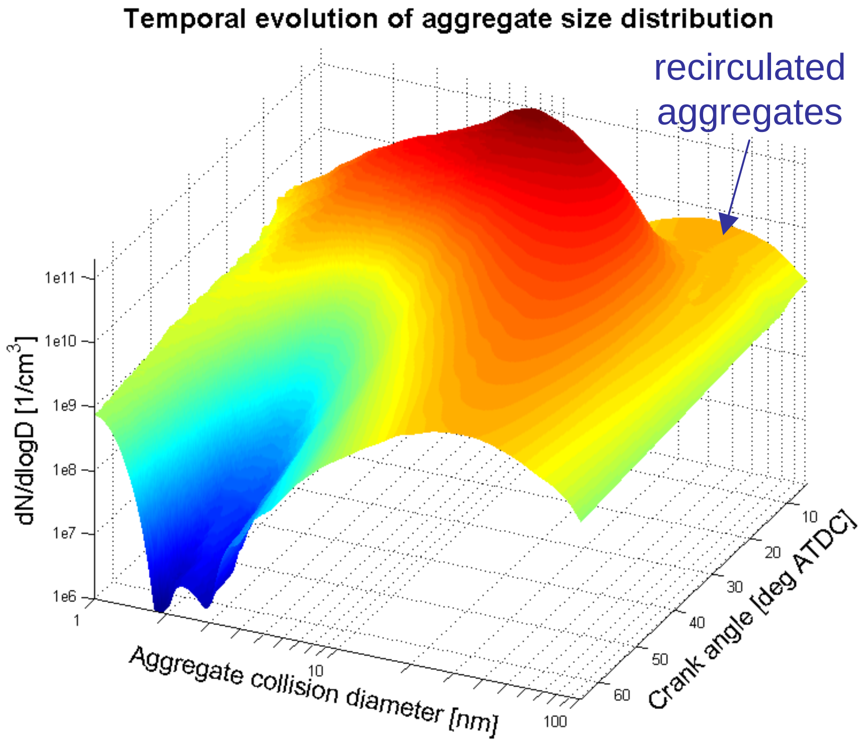
Markus Kraft
mk306@cam.ac.uk

Aggregate size distributions (III)

Simulation



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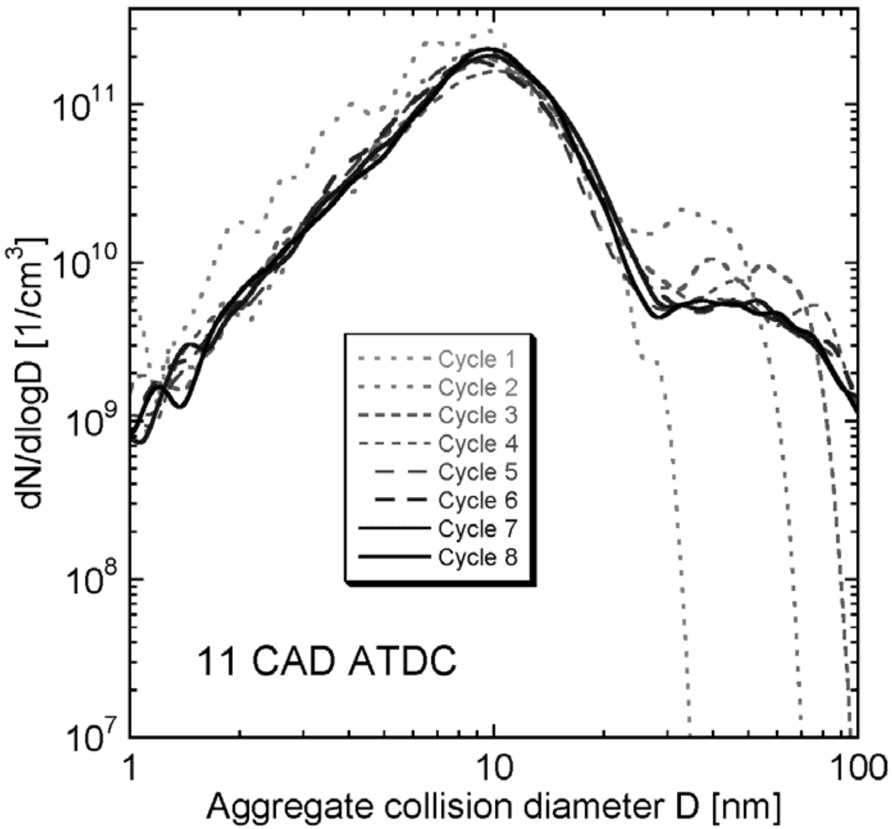
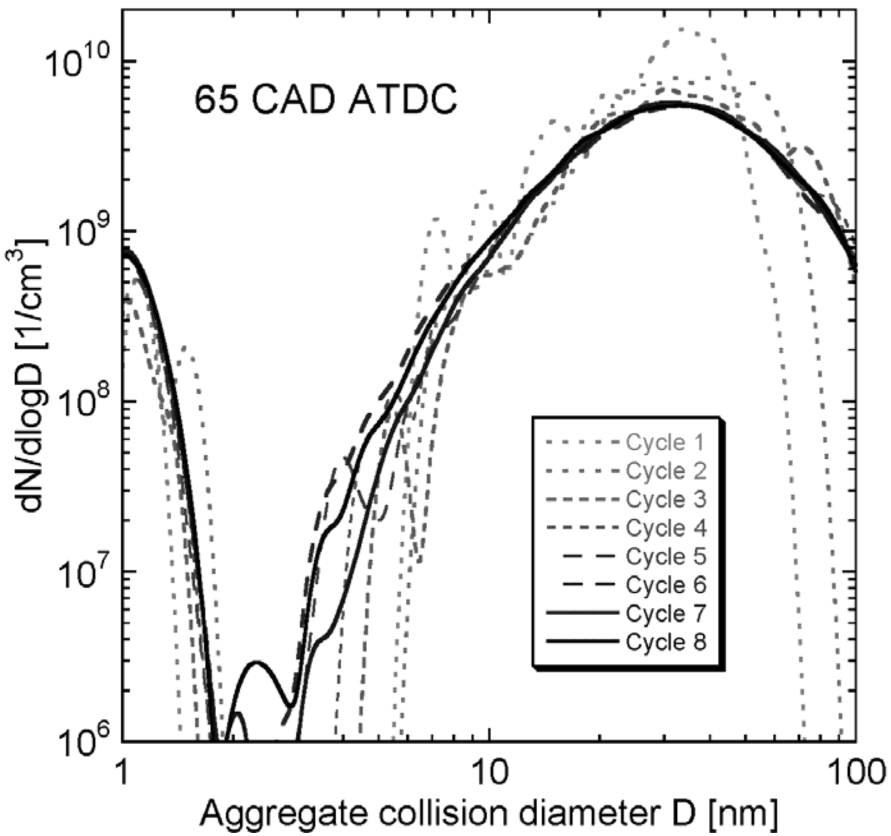
Markus Kraft
mk306@cam.ac.uk

Role of EGR

Simulation



Simulation



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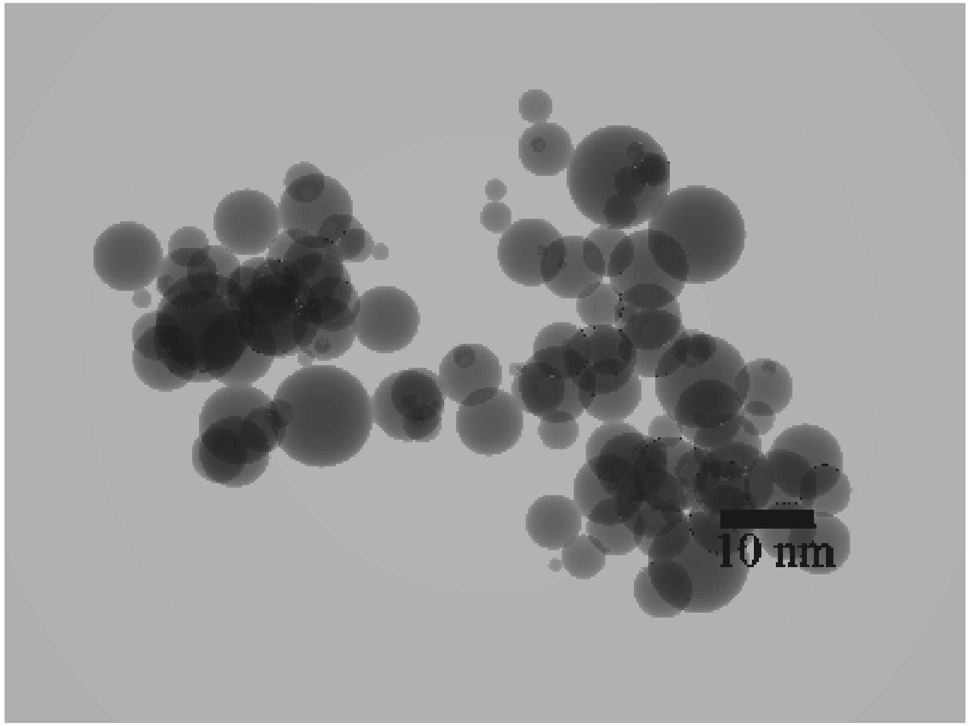
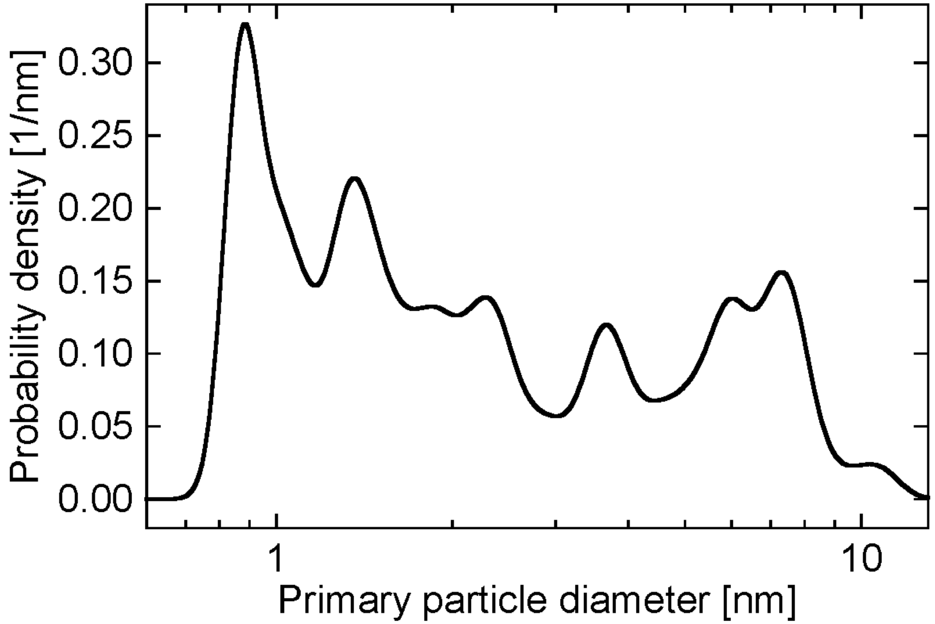
Markus Kraft
mk306@cam.ac.uk

Sampled aggregates (I)

Simulation



Simulation

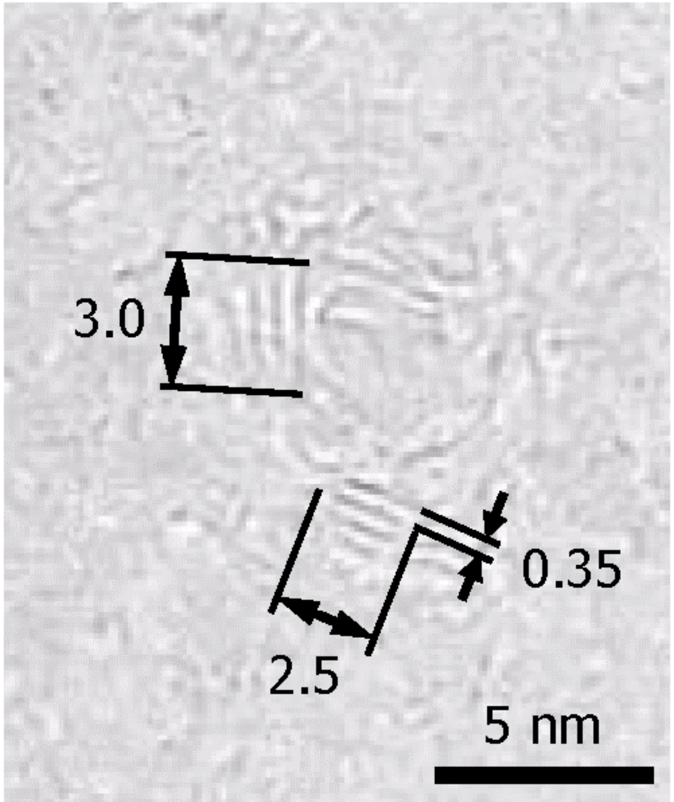


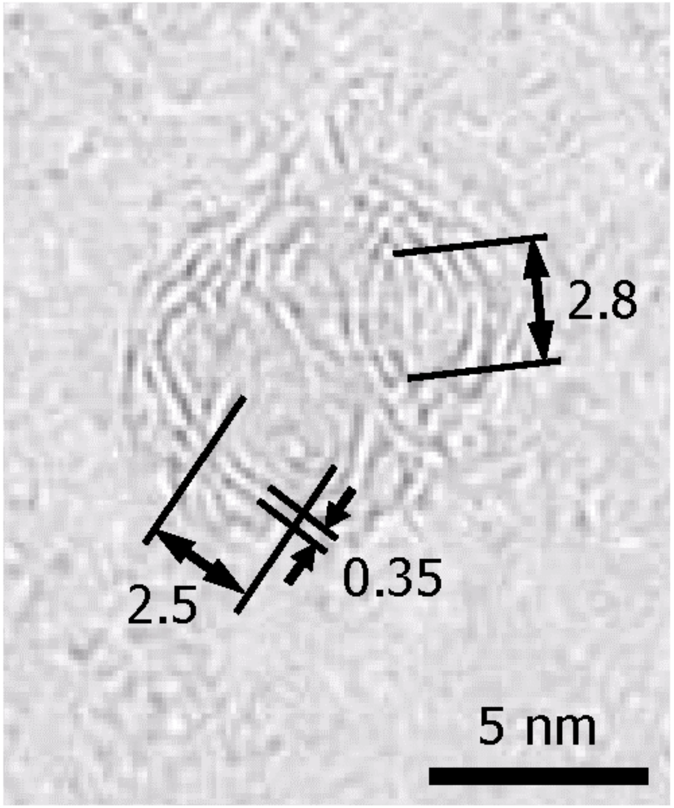
49.4 CAD ATDC, 129 primaries, coll. diam. 64 nm

Markus Kraft
mk306@cam.ac.uk

Sampled aggregates (II)

Experiment, sampled at ~16 CAD ATDC





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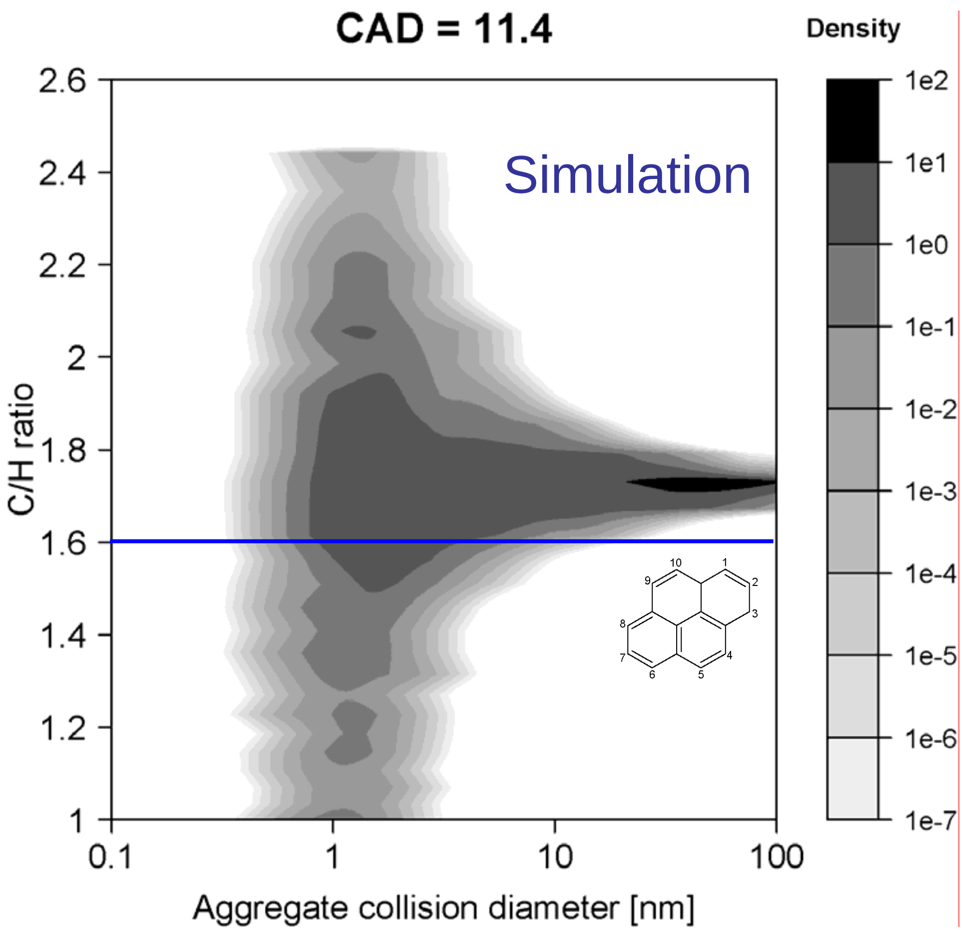
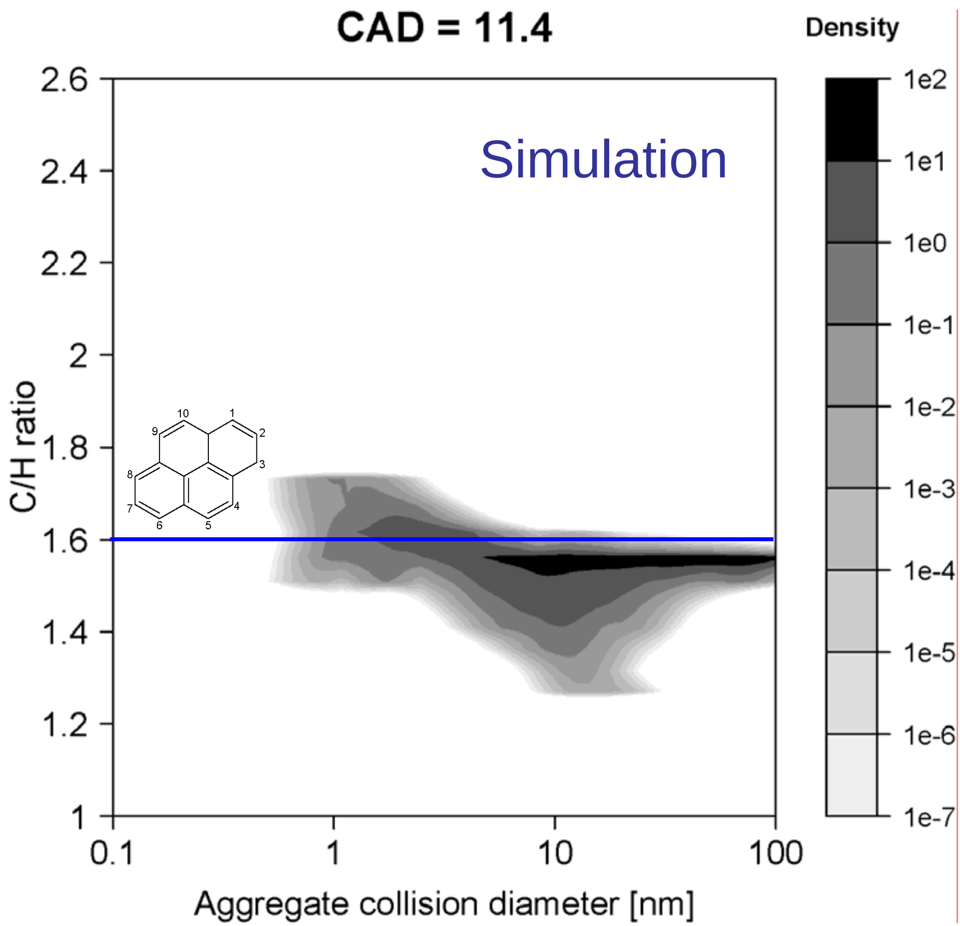
Markus Kraft
mk306@cam.ac.uk

Aggregate composition pdfs (I)

large inception rate



large condensation rate



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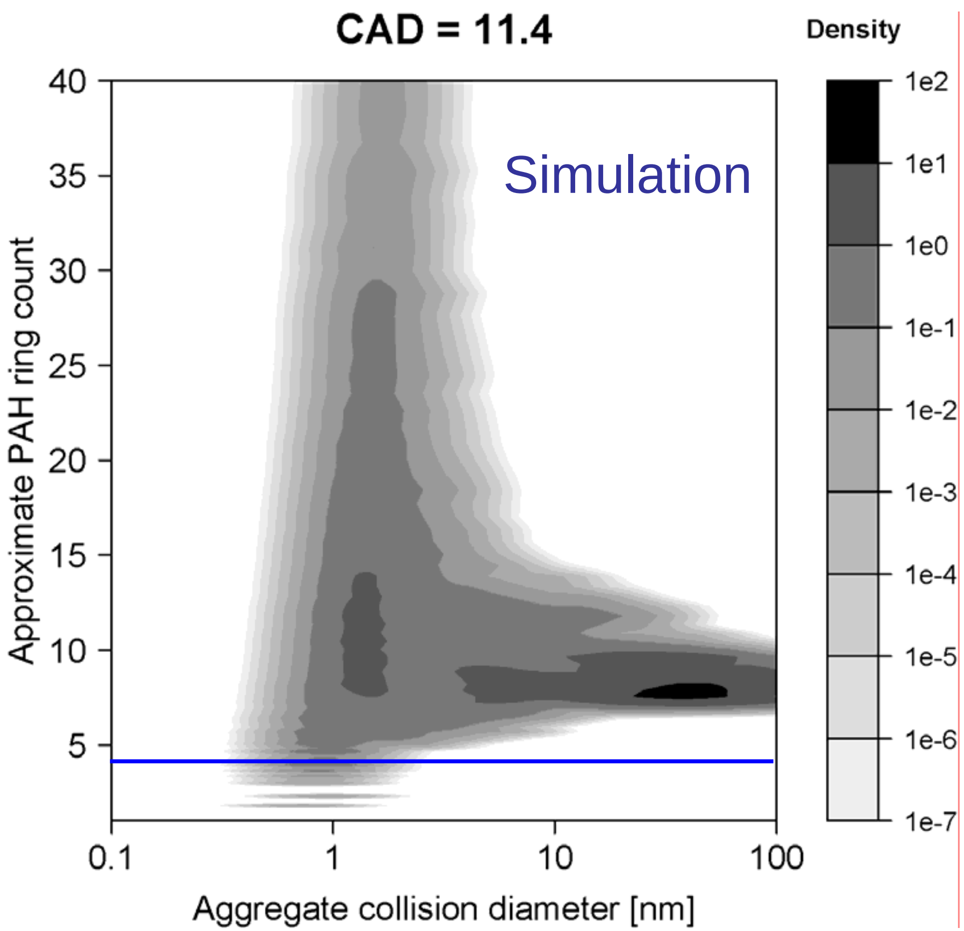
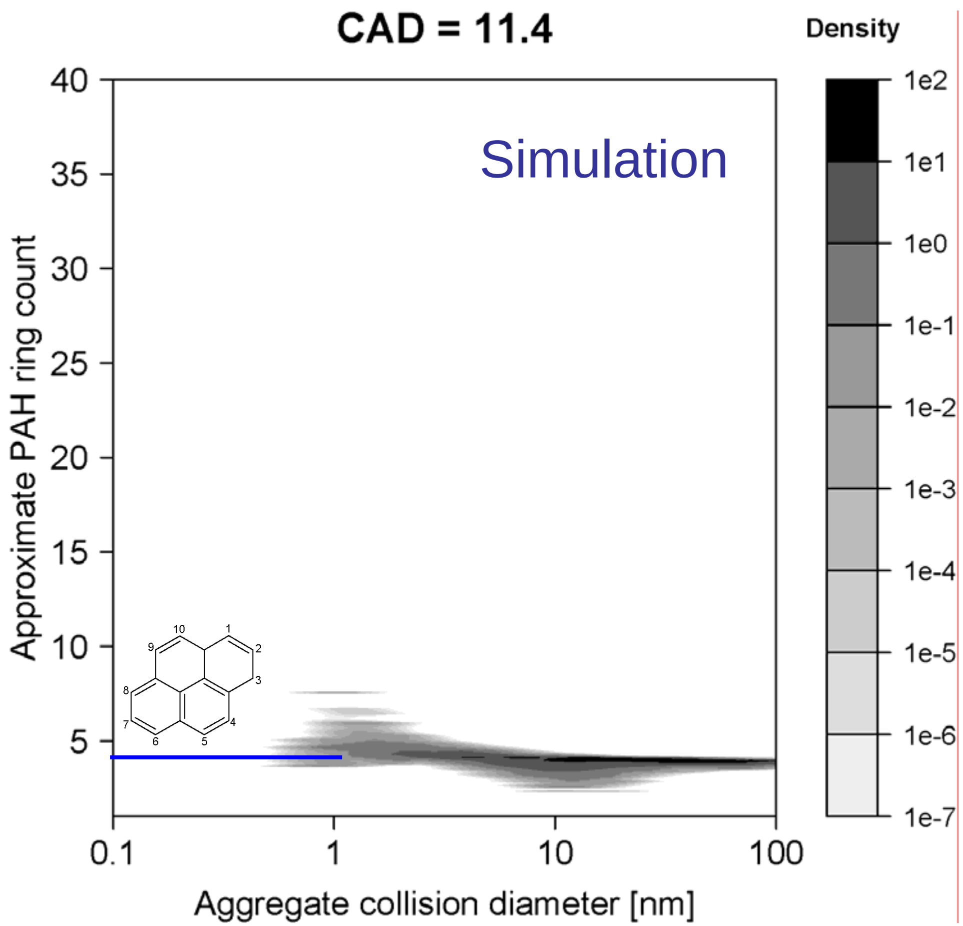
Markus Kraft
mk306@cam.ac.uk

Aggregate composition pdfs (II)

large inception rate



large condensation rate



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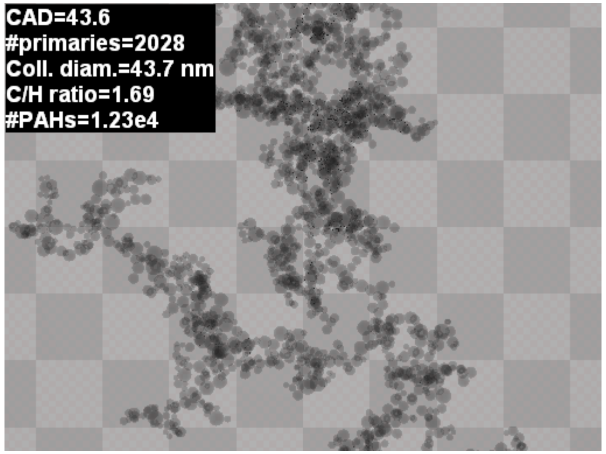
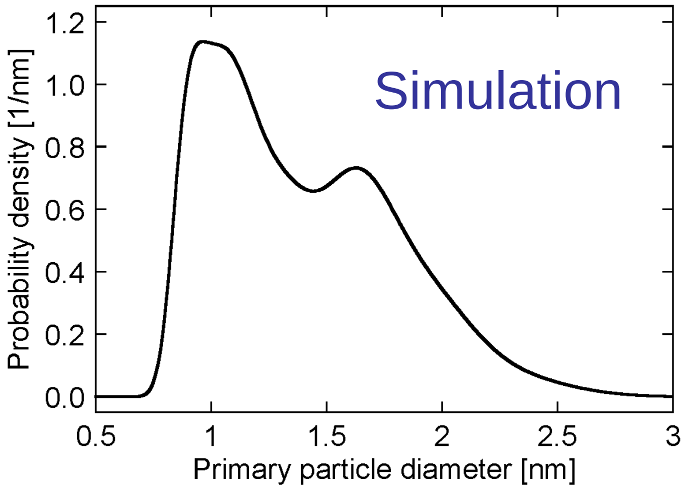
Markus Kraft
mk306@cam.ac.uk

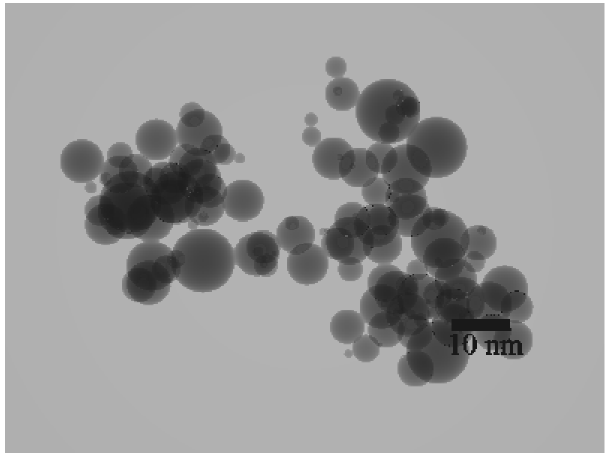
Inception vs. condensation

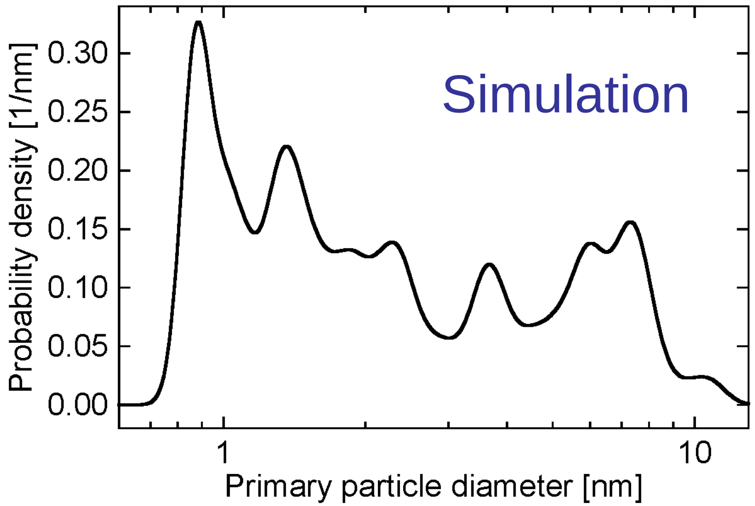
large inception rate





large condensation rate





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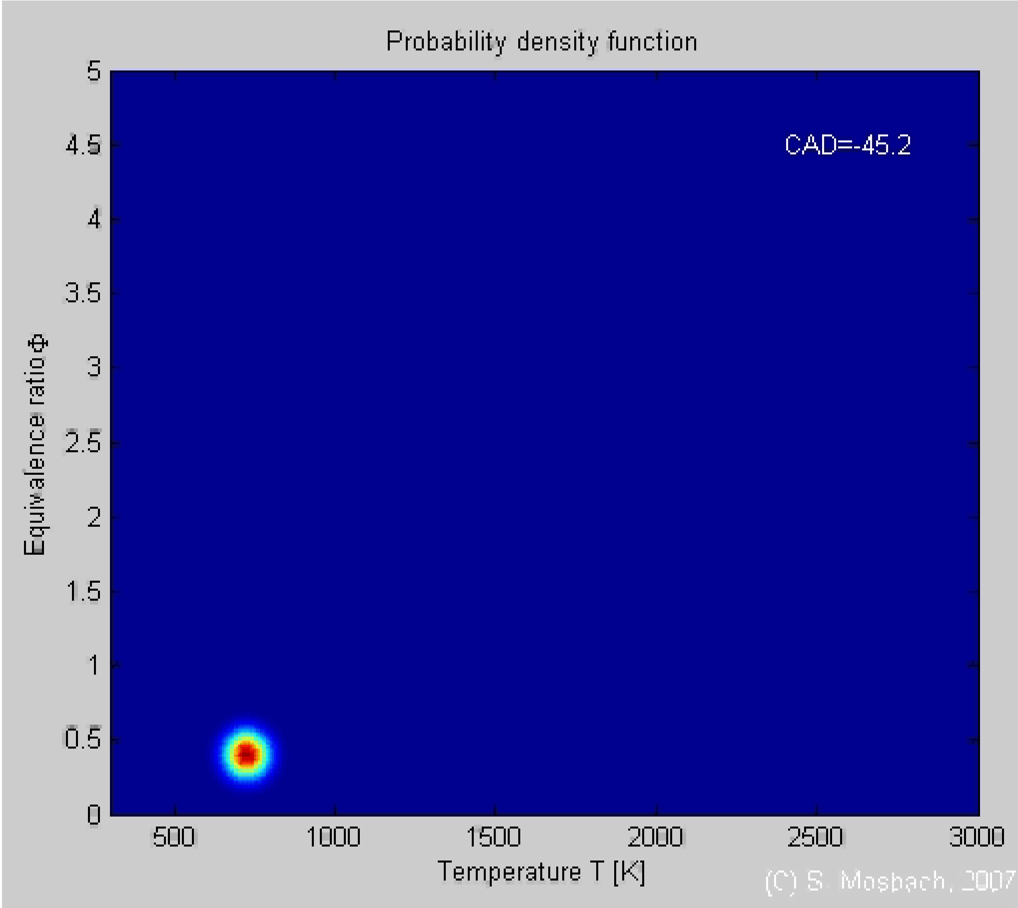
Markus Kraft
mk306@cam.ac.uk

Future engine soot models (I)

- Partially stratified HCCI
- Partially premixed CIDI
- Conventional CIDI
- (Partially stratified) DISI



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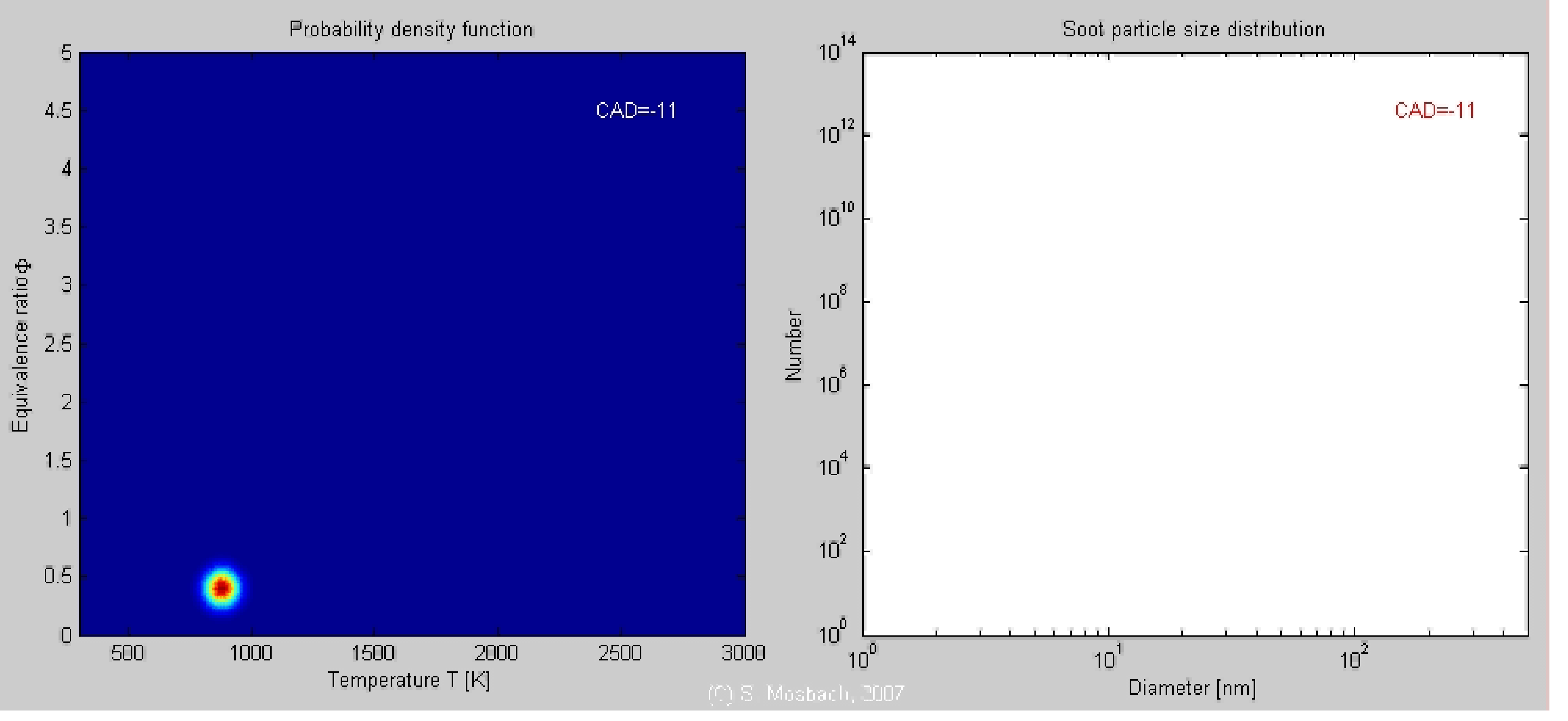
mk306@cam.ac.uk

Future engine soot models (II)

Soot formation in a partially stratified HCCI engine:



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Markus Kraft
mk306@cam.ac.uk

Thank you!

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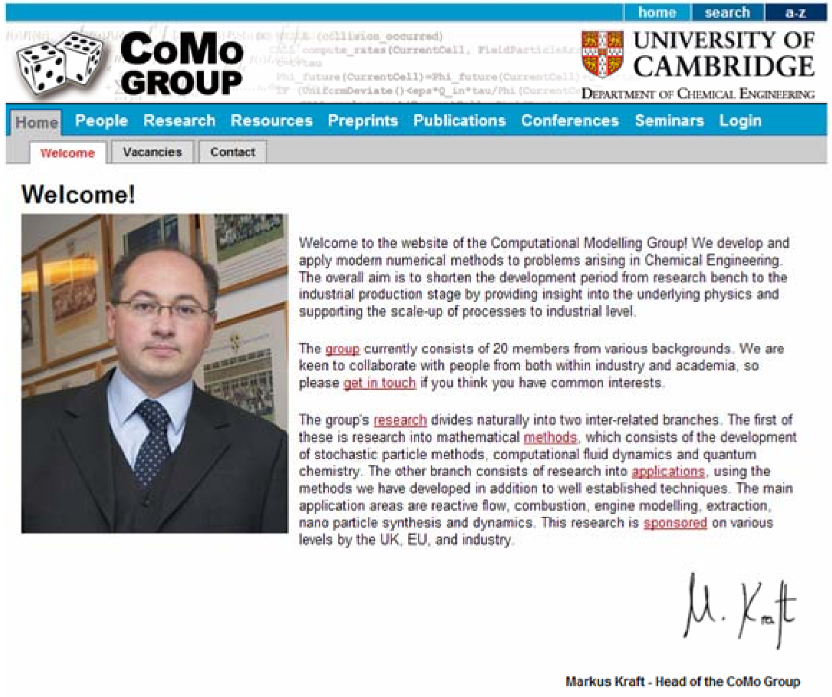

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Markus Kraft
mk306@cam.ac.uk