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# Fuel-Neutral Studies of Particulate Matter Transport Emissions

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# Overview



## Timeline

- Start - June 2008

## Budget

- Total project funding through FY12
  - DOE - $800K
- Funding received in FY11 - $200K
- Funding for FY12 - $200K

## Barriers

- Barriers addressed for enabling of high-efficiency engine technology:
  - B. Lack of cost-effective emission control
  - C. Lack of modeling capability for combustion and emission control
  - F. Lack of actual emissions data on pre-commercial and future combustion engines

## Partners

- General Motors Company - provide project guidance, provide hardware to ERC
- Engine Research Center at University of Wisconsin, Madison - host and operate test engine
- Pennsylvania State University – perform advanced soot analysis

# Relevance and objectives



## Overall objective: Enable application of future high-efficiency engine technology

Barrier: Lack of actual emissions data on pre-commercial and future combustion engines
Objective: Systematic particulate characterization with single-cylinder test engine, guided by industry

Barrier: Lack of cost-effective emission control
Objective: Seek to shorten development time of filtration technologies for future engine technologies

Barrier: Lack of modeling capability for combustion and emission control
Objective: Develop modeling approaches relevant to the likely key challenge for SIDI filtration – high number efficiency at high exhaust temperatures

# Approach



## Advanced Particulate Characterization

### Aerosol methods

- DMA (select by particle mobility)
- APM (select by particle mass - *new in second round tests*)
- SPLAT-II (measures aerodynamic size, mass spectrum for each particle)

- Huge number of particles (O~$10^6$)
- Real time
- Used in conjunction, many properties of particulate populations can be measured or *inferred*
  - Particle size distribution
  - Aggregate shape
  - Avg primary particle size
  - Particle composition (as a function of other parameters listed)

### Micro/nano-scale analysis

- TEM
- HRTEM
- FTIR-ATR
- XPS
- TGA →HRTEM

- Smaller numbers of particles (O~$10^2$)
- Not real time
- *Direct observation* of parameters
  - Particle size distribution
  - Aggregate shape
  - Individual primary particle size

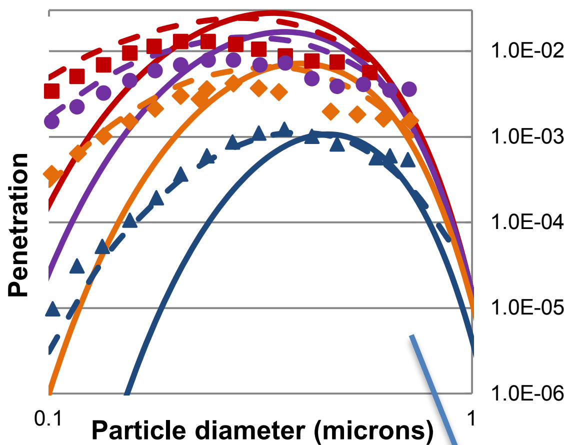
  *Also:*
  - Particle composition
  - Particle nano-structure



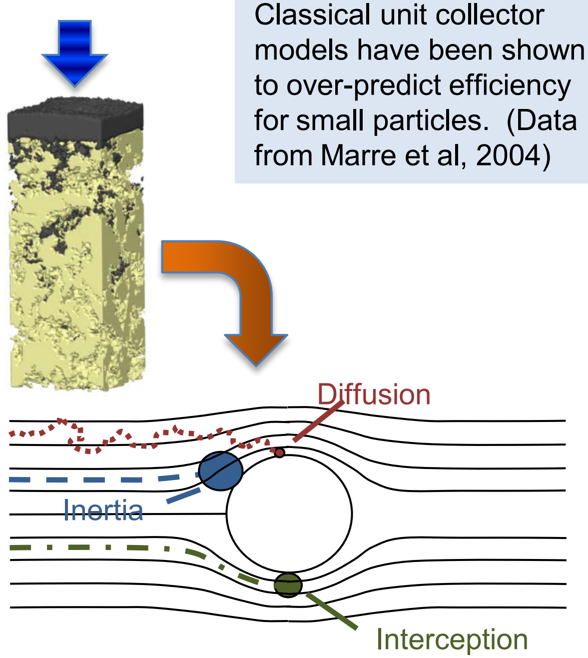
Complimentary

# Approach






Classical unit collector models have been shown to over-predict efficiency for small particles. (Data from Marre et al, 2004)




## Modeling and technology development

- Due to high exhaust temperatures and relatively low soot loadings, SIDI filter systems may not have a significant accumulation of soot
- Current DPF systems rely upon soot cake filtration to achieve very high particulate removal efficiencies
- While many SIDI systems meet current particulate emissions limits based on mass, number-based limits are more challenging
- Unit collector models have been extensively used for design of DPF systems, with a focus on mass-based limits
- Model improvements are needed for prediction of number efficiency across the entire particle size spectrum
- 3-D micro-scale simulations are being explored as a means to enhance lower order device-scale models
- Improved models are being used to help screen candidate filter technologies for advanced internal combustion engines

Marre, S., J. Palmeri, A. Larbot, and M. Bertrand, "Modeling of submicrometer aerosol penetration through sintered granular membrane filters". *JOURNAL OF COLLOID AND INTERFACE SCIENCE, 2004. 274(1): p. 167-182.*

# Technical Accomplishments



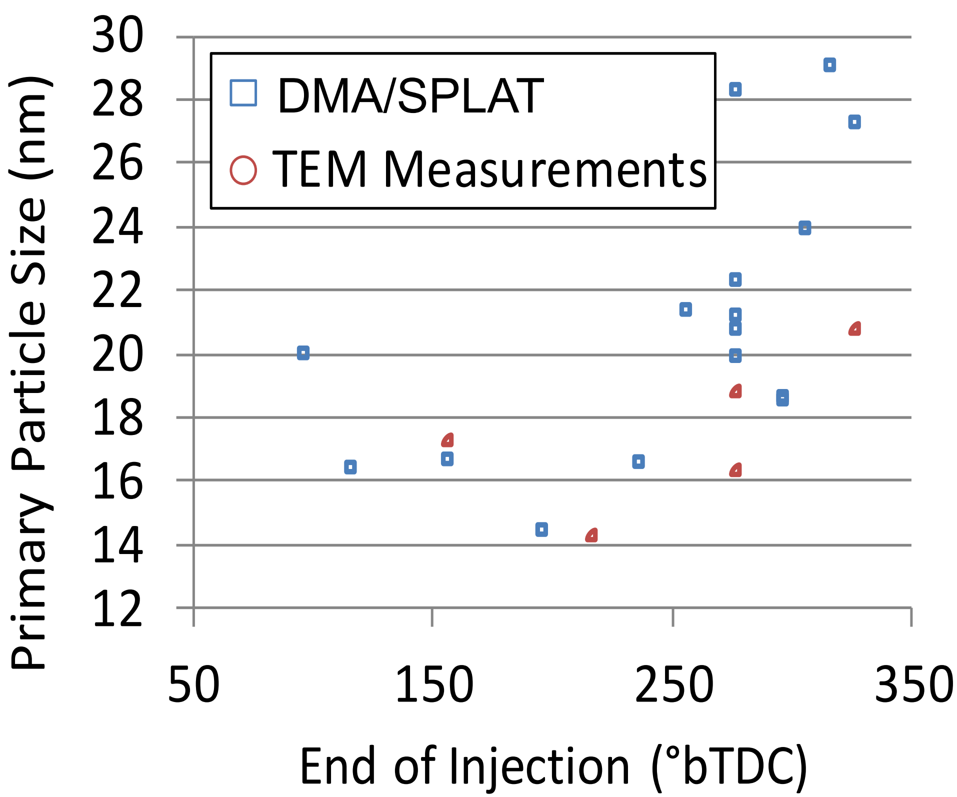
- Exhaust particulate characterization
  - 2 extensive cooperative campaigns with University of Wisconsin ERC
  - 4 fuel blends examined to date, including E20, E85
  - 3 engines: SIDI, RCCI, and GDICI
  - Dozens of experiments at various relevant engine operating points
  - Standard and advanced characterization techniques including SMPS, SPLAT-II, APM, TEM, HRTEM, FTIR-ATR, XPS – *used together for a more complete picture of particulate populations*

- Aftertreatment technology development
  - Effect of filter coatings on filtration performance
  - Application of improved unit collector modeling for high number efficiency
  - Pore-scale simulation

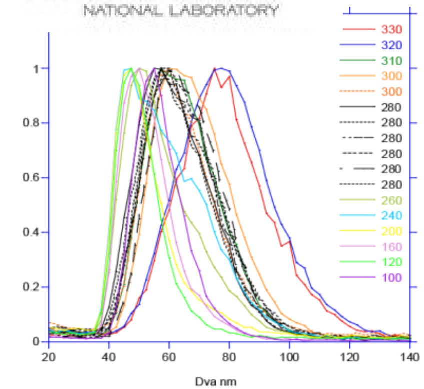
# Technical Accomplishments

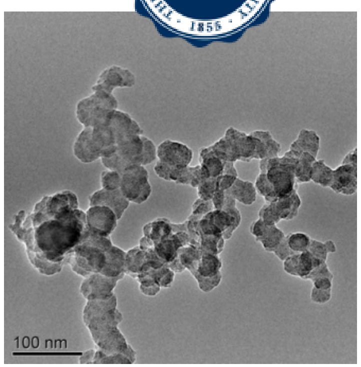


## Variation in avg primary particle size




- Developed and applied new method based on vacuum aerodynamic sizing of particles pre-selected by mobility size
- TEM observations by automated image processing of many aggregates



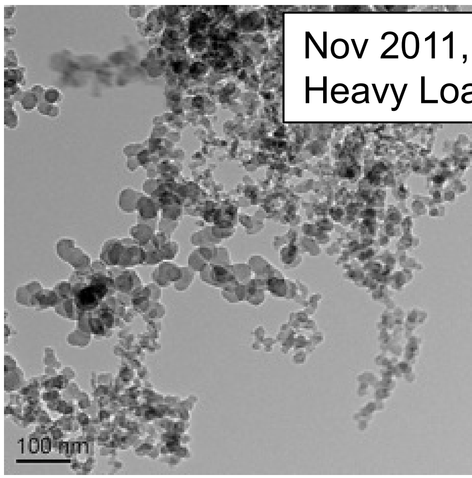

***<u>Remarkable agreement between two very different approaches</u>***

More variation in primary particle size than seen with diesel

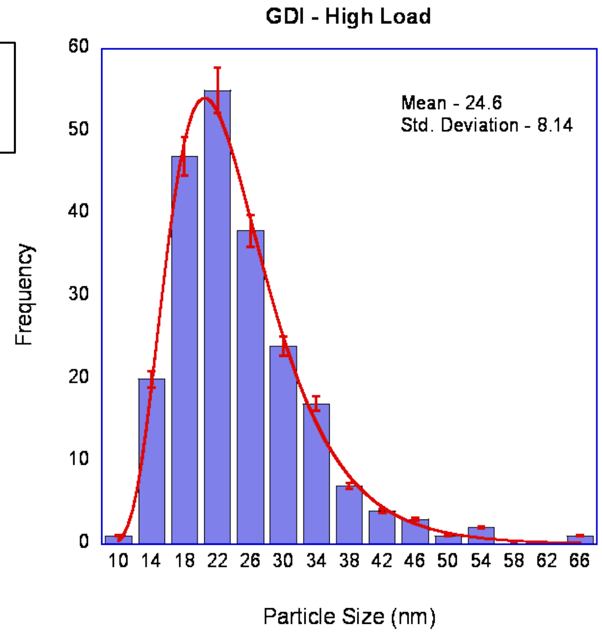
# Technical Accomplishments

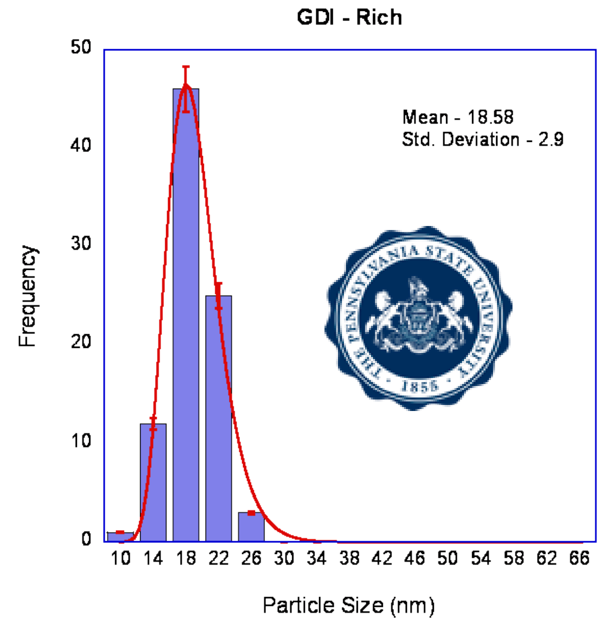


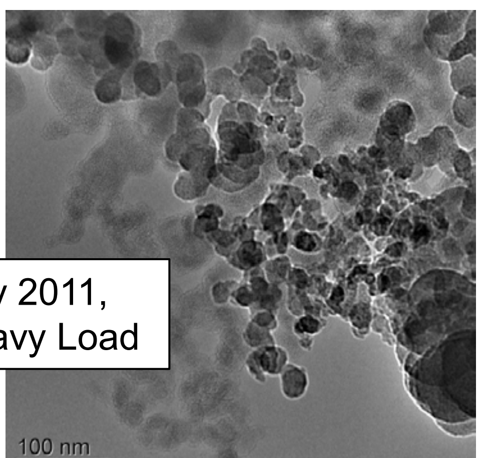
## TEM reveals dramatic variation in size of individual primary particles




Nov 2011, Heavy Load



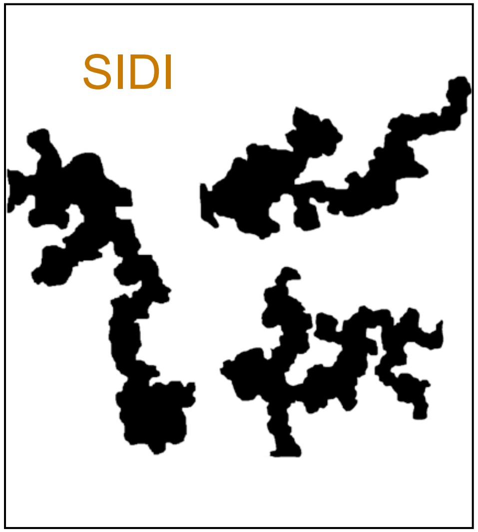



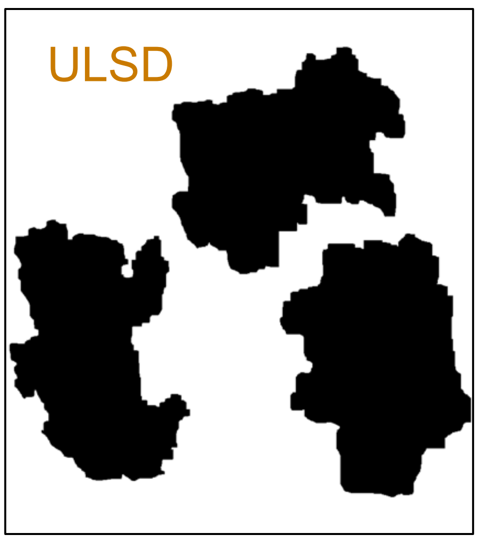

July 2011, Heavy Load

- High load condition may show some of the most variation in primary particle size, sometimes within what appears to be the same agglomerate
- Smaller particles seem more common at the periphery of aggregates – may indicate different trajectories
- Some instances of apparently fully-formed aggregates of smaller particles

# Technical Accomplishments

## Geometric analysis of aggregate morphology




- 2D outlines used for quantitative comparison of aggregate morphology
- Most common diesel soot shape is “fluffy ball” – SIDI soot is less compact
- SIDI morphology varied with operating conditions

| Type | Root Form factor | Fractal Dimension |
|---|---|---|
| ULSD | 0.62 | 2.39 |
| SIDI | 0.29 | 1.86 |

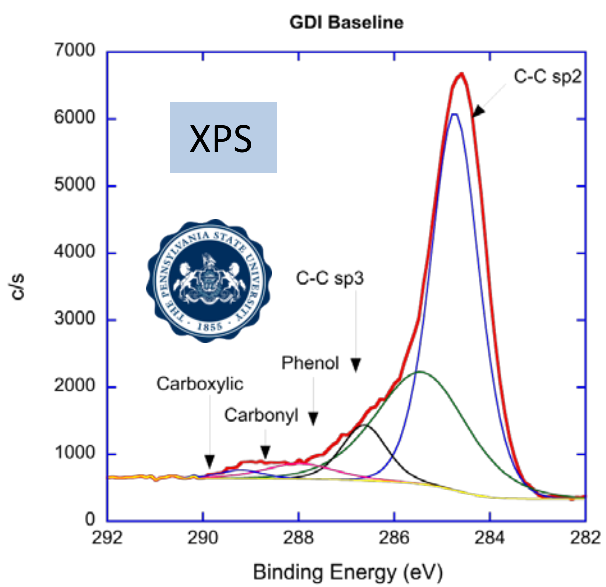
Root Form factor

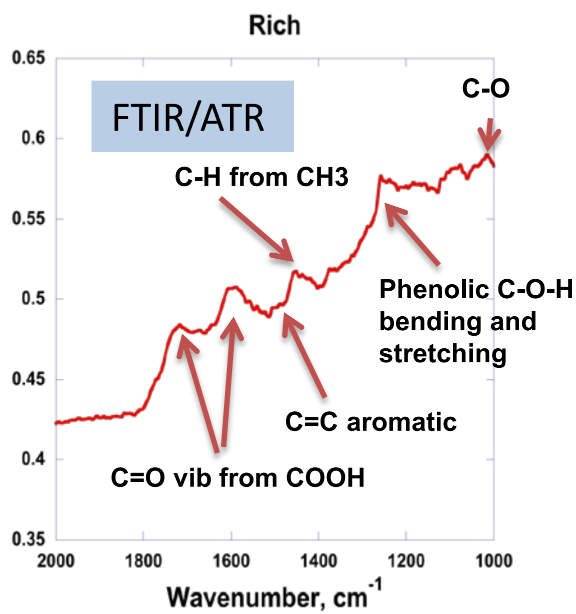
$$\sqrt{\frac{4\pi * Area}{(Perimeter)^2}}$$

July, 2011 SIDI

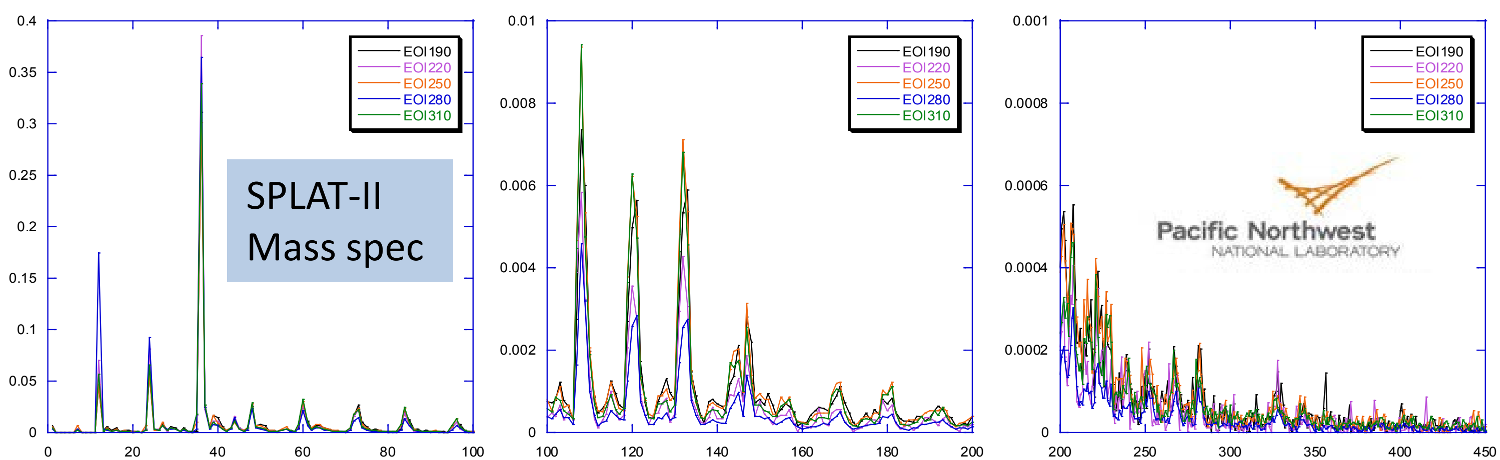
| Engine Parameter | Fractal Dimension |
|---|---|
| Late EOI | 2.20 |
| Rich | 2.37 |
| EOI 280 | 1.69 |
| Lean | 1.86 |
| Heavy Load | 2.06 |

# Particulate composition



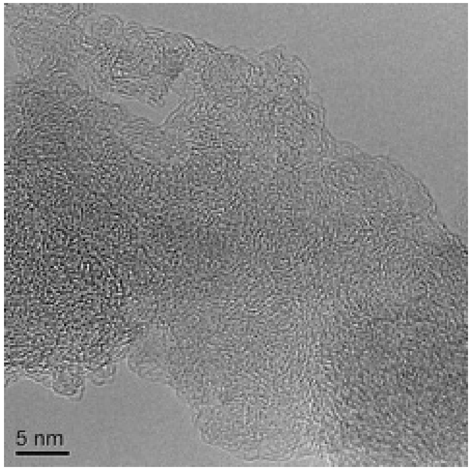

- Very high organic content in all particles
- Experiments with volatile stripping process had suggested tightly integrated organics
- XPS and FTIR/ATR data from Penn State gives a consistent picture, as does HRTEM (next slide)



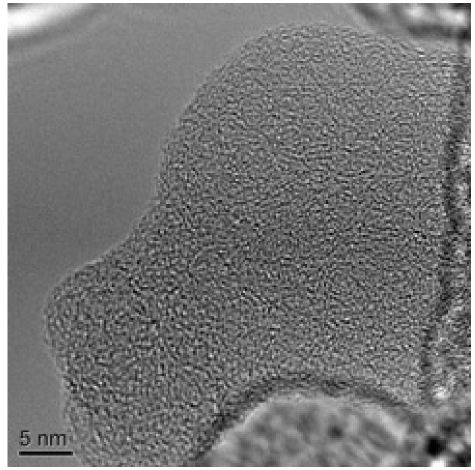

| Fraction of EC | |
|---|---|
| EOI190 | 0.42 |
| EOI220 | 0.54 |
| EOI250 | 0.40 |
| EOI280 | 0.63 |
| EOI310 | 0.46 |

# Technical Accomplishments



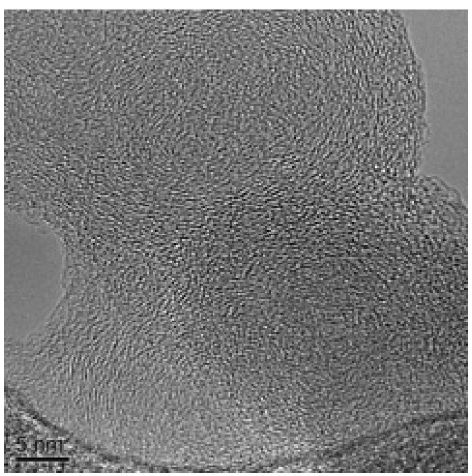



Late EOI
Highly curved, amorphous

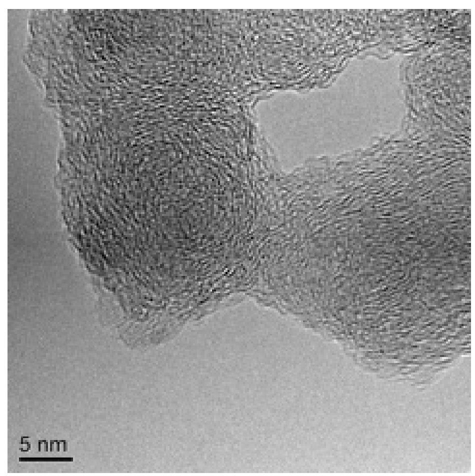



Rich
Amorphous

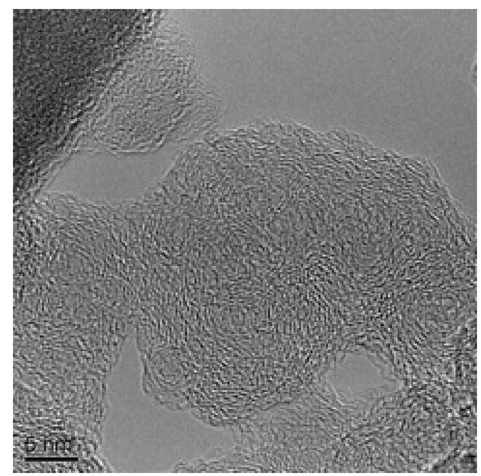
- Nanostructure was generally amorphous or lacked significant order
- Some variation in degree of order across operating conditions with EEE
- Amorphous nanostructure consistent with tightly integrated organics




EOI 280
Better order with short but recognizable lamella




Lean
Disorganized, but not chaotic




Heavy Load
Recognizable nanostructure

# Technical Accomplishments

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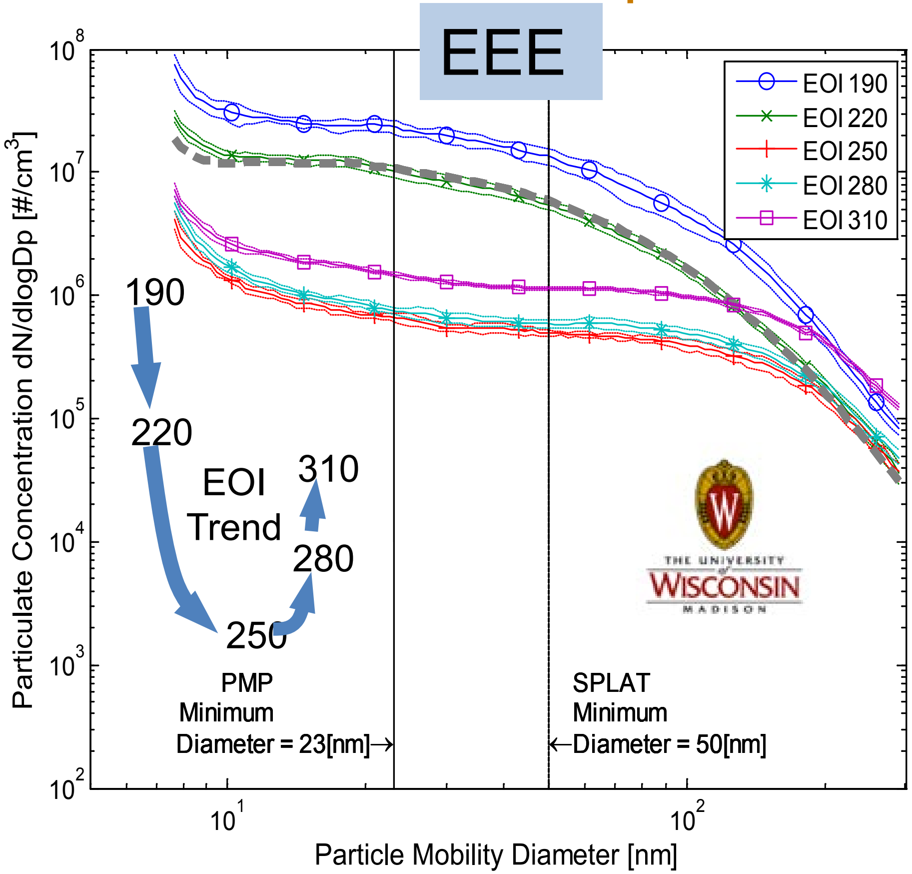
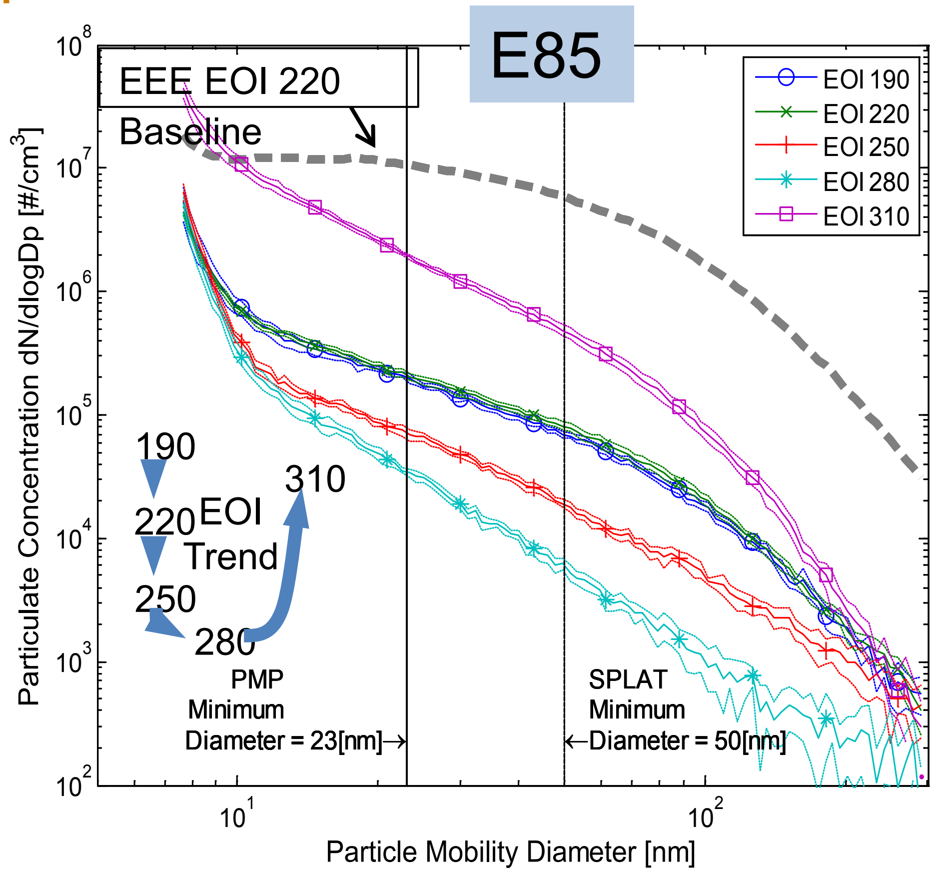
## Second round of cooperative experiments included E20, E85



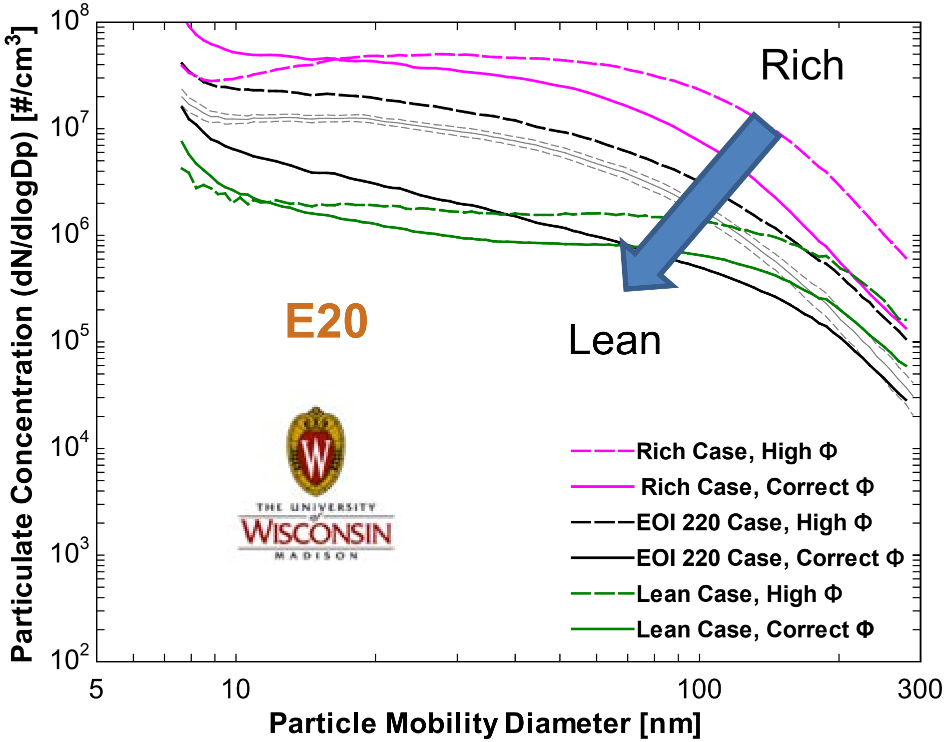

- Ethanol blends generally produced lower particle counts
- Different size distributions
- Somewhat different dependency on EOI
- Different composition: Ethanol blends generally produce particles with higher organic content (example, Med. Rail pressure)

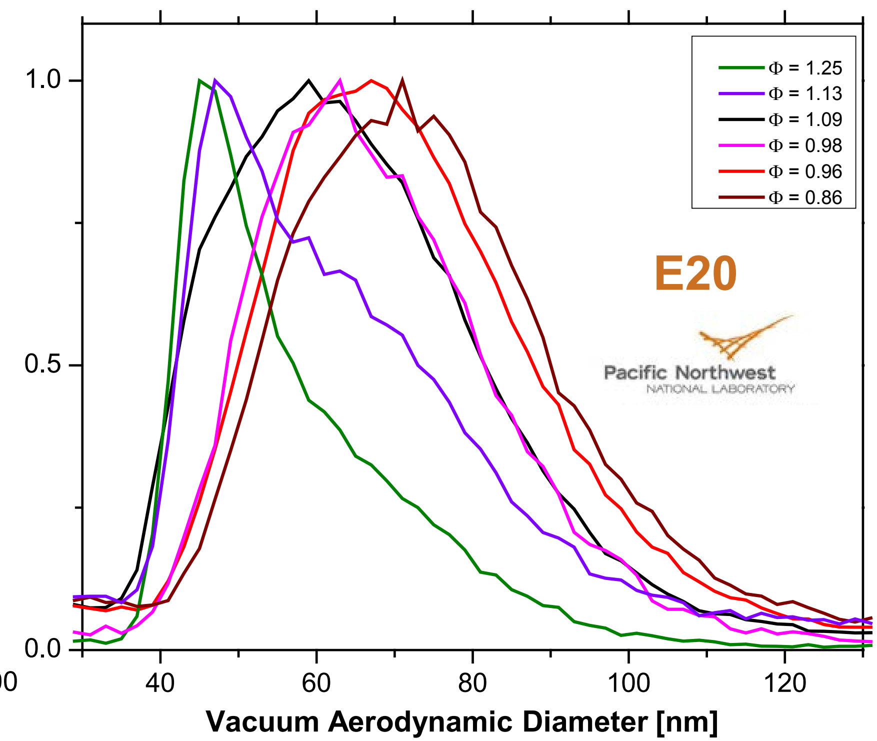
| Species | E85 | E20 | EEE |
|---|---|---|---|
| EC | 19.7 | 38.7 | 52.6 |
| Org | 72.8 | 54.3 | 43.5 |
| PAH | 3.9 | 2.6 | 2.0 |

# Technical Accomplishments



## Second round of cooperative experiments included equivalence ratio sweep for different fuels




- The E20 data shows a generally observed trend of particulate number decreasing with equivalence ratio
- Lower equivalence ratios produce particles with larger primary spherule diameters and broader distributions of shapes

# Technical Accomplishments

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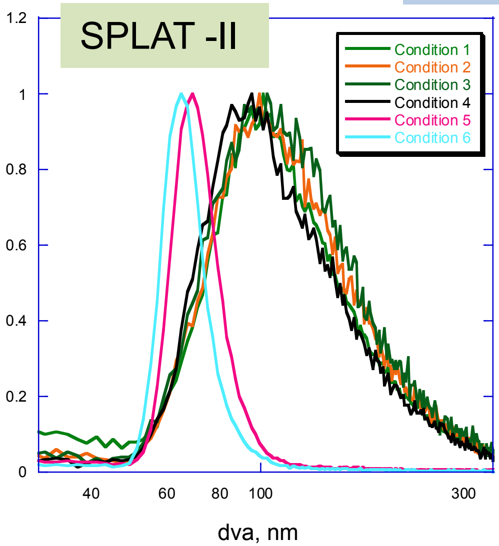
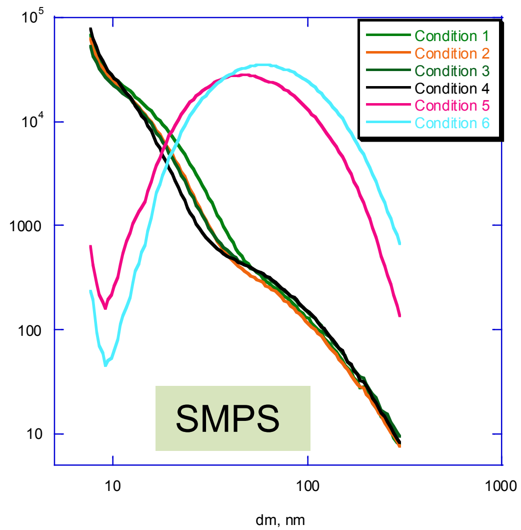
## Application to other advanced engine technologies

### GDICI




1: 5.5 bar IMEPn, 2000 rpm, SOIC2=39
2: 5.5 bar IMEPn, 2000 rpm, SOIC2=36
3: 5.5 bar IMEPn, 2000 rpm, SOIC2=33
4: 5.5 bar IMEPn, 2500 rpm, SOIC2=39
5: 14 bar IMEPn, 2000 rpm, SOIC2=21
6: 14 bar IMEPn, 2000 rpm, SOIC2=21 SOIC3=10

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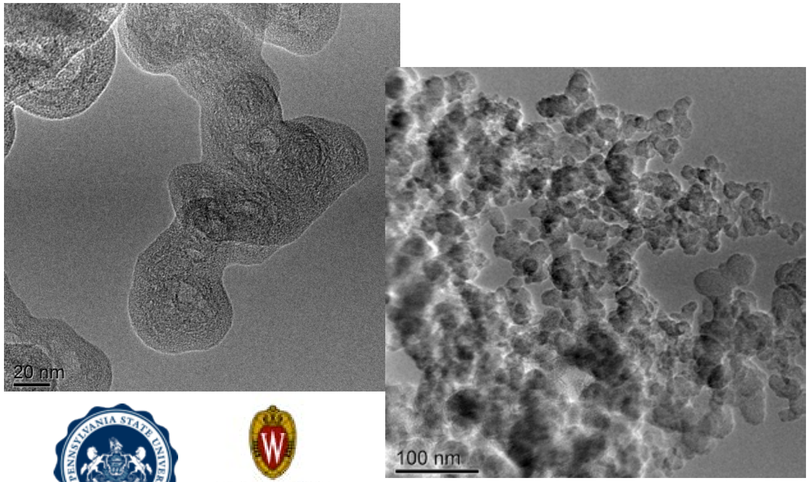
- GDICI runs fell into two groups
- Low load produced spherical, nucleation mode particles, dominated by organics
- High load produced fractal soot

### RCCI




THE PENNSYLVANIA STATE UNIVERSITY 1855

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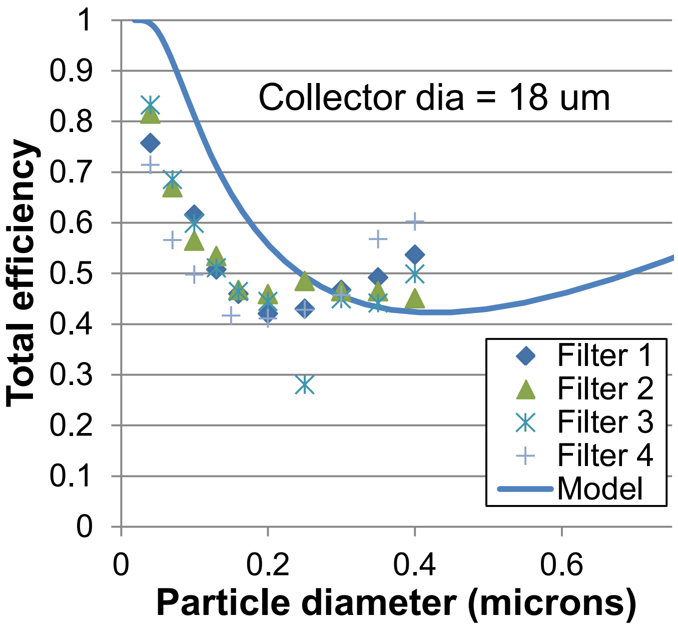
- Like SIDI, RCCI particulates had a wide range of primary particle sizes
- Generally, RCCI nanostructure was somewhat more ordered than SIDI
- Organic content varied somewhat between operating conditions
- One condition (upper left) showed unusual hollow structures

# Technical Accomplishments

## Modified unit collector model allows better fit of observed size-dependent filtration efficiency

### Classical diffusion efficiency model

$$\eta_D = 4.0 A_s \left( \frac{1}{Pe} \right)^{2/3}$$



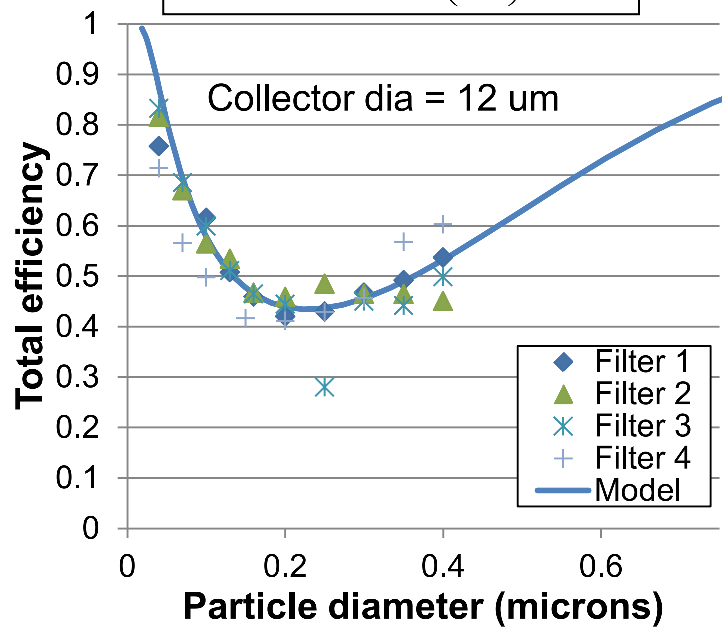

$$A_s = \frac{2(1-p^5)}{2-3p+3p^5-2p^6}$$

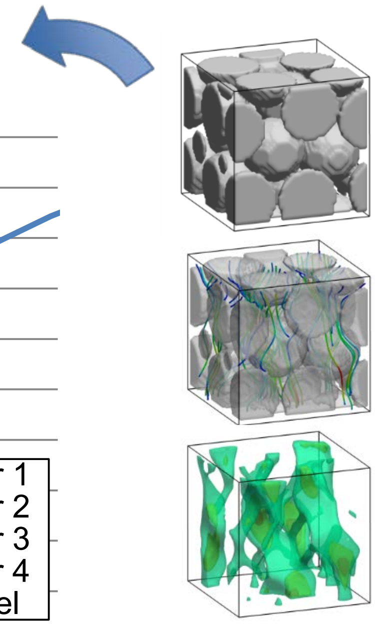
$$p = (1-\varepsilon)^{1/3}$$

### Model proposed by Long and Hilpert, 2009

(using micro-scale simulations)

$$\eta_D = 15.46 \frac{(1-\varepsilon)^2}{\varepsilon^3} \left( \frac{1}{Pe} \right)^{0.65} R^{0.19}$$



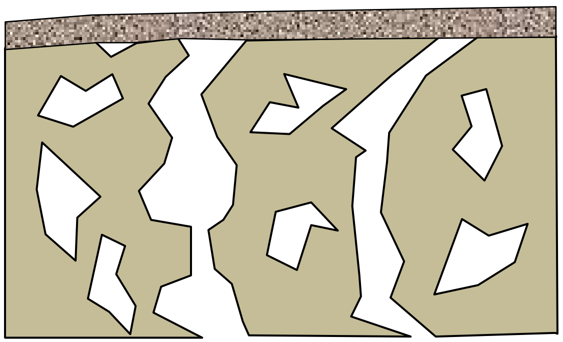



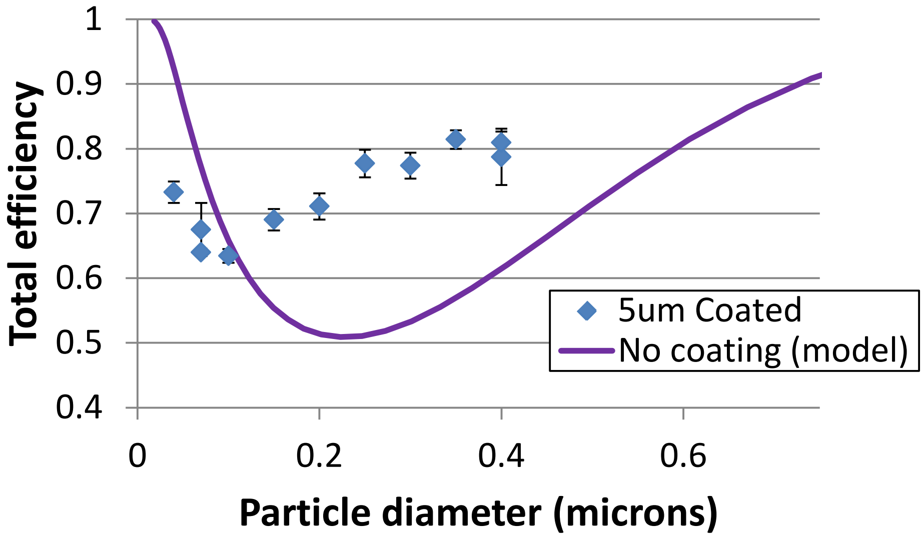
Long, W. and M. Hilpert (2009). "A Correlation for the Collector Efficiency of Brownian Particles in Clean-Bed Filtration in Sphere Packings by a Lattice-Boltzmann Method." ENVIRONMENTAL SCIENCE & TECHNOLOGY **43**(12): 4419-4424.

# Technical Accomplishments

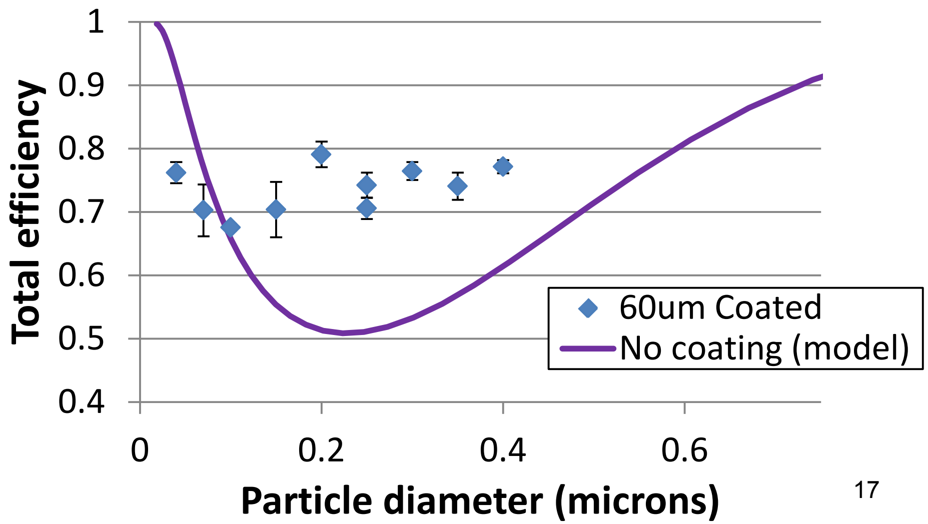


## Membrane coatings for high number efficiency without soot layer






- Membrane-coated filters are one candidate technology to maintain high number efficiency without a soot cake
- Several experimental coatings were applied to a standard DPF
- Coatings can improve number efficiency at the cost of higher backpressure
- Coating method is critical – may be difficult at industrial scale



# Collaborations



- General Motors Company (Industry): Provide hardware, expertise, and operational guidance for engine experiments at the ERC. Advise on project direction and priorities.
- Engine Research Center at University of Wisconsin, Madison (Academic): Operate test engine - including shakedown tests, independent experiments, and cooperative experiments. Assist in analysis and publication of data.
- Pennsylvania State University (Academic): Perform advanced analysis to characterize particulate nanostructure and composition.

# Future Work



- Complete analysis of second round data and publish findings
- Employ micro-scale modeling to explore possible approaches to achieve high filtration efficiency in high temperature exhaust
- Use improved unit collector models to explore tradeoffs between parameters such as number efficiency, backpressure, unit volume in SIDI applications
- Conduct experiments to evaluate candidate filter technologies for SIDI
- Apply advanced particulate characterization to subsequent generations of engines – leaner, higher fuel efficiency

# Summary



- This project represents a collaboration between PNNL; GM; University of Wisconsin, Madison; and Pennsylvania State University to characterize particulates from advanced combustion engines and develop suitable mitigation strategies.
- A second round of cooperative experiments with SIDI particulates has been completed. Very large sets of resulting data are currently being processed.
- Micro/nano-scale analyses from Penn State have been complimentary to advanced aerosol measurement techniques from PNNL.
- New efforts have confirmed previous conclusions and suggested several new ones:
  - Primary particle size and aggregate morphology in SIDI particulates are highly variable and depend upon engine operation.
  - SIDI soot aggregates tend to be less compact than diesel particles (relevant to filtration).
  - SIDI soot resulting from all fuels tested had very high organic content, which was tightly integrated with the inorganic carbon.
  - Ethanol blends generally produced lower particulate concentrations. E85 produced particles with even higher organic content than observed with EEE.
  - Nanostructure of SIDI particulates is generally less ordered than diesel soot. Nanostructure of particulates from ethanol blends tends to be even less ordered than those from EEE.
- Modified unit collector models appear to do a better job matching experimental data on size-dependent filtration efficiency.
- Experiments confirmed that number efficiency can be improved by filter coatings, but it is difficult to achieve consistent results without impacts to back-pressure.

Technical Back-Up Slides

(Note: please include this “separator” slide if you are including back-up technical slides (maximum of five). These back-up technical slides will be available for your presentation and will be included in the DVD and Web PDF files released to the public.)

# ADS: APM/DMA/SPLAT

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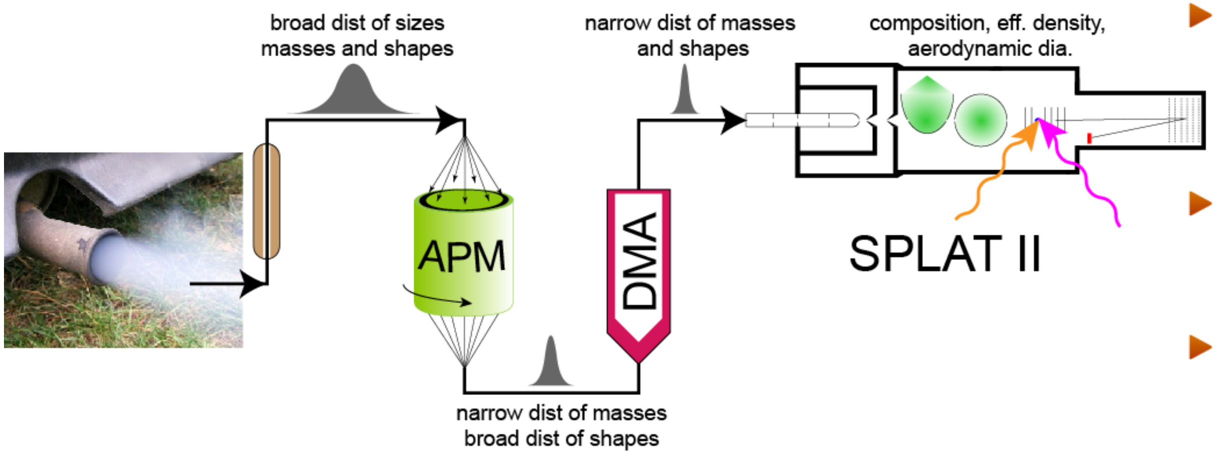
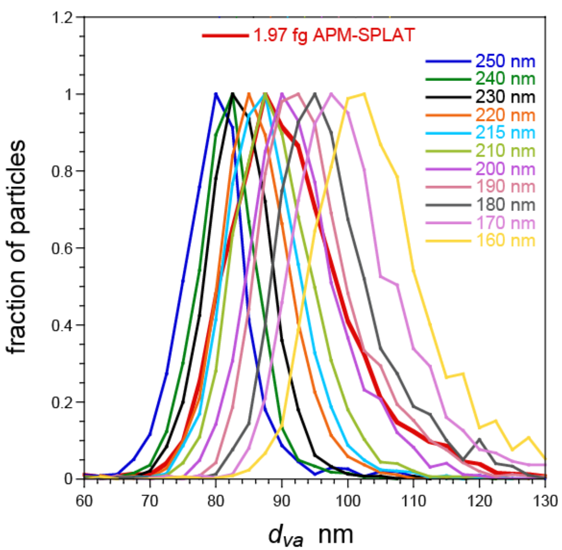
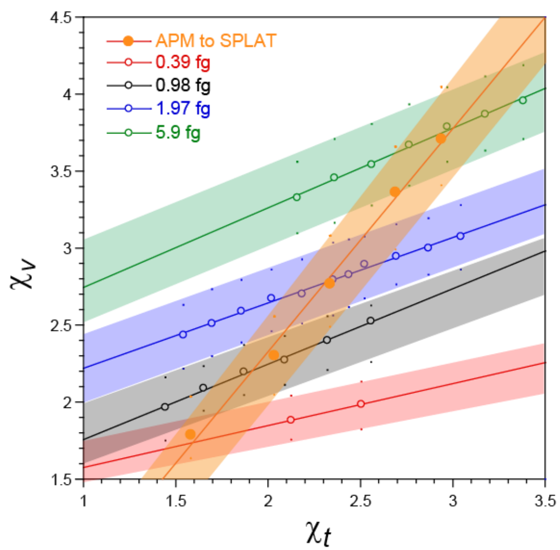
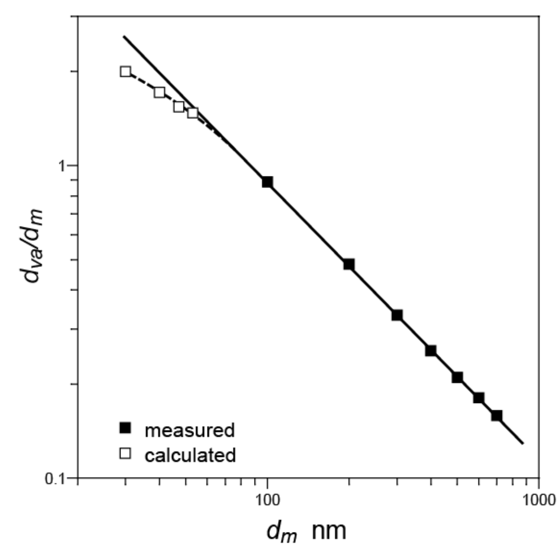



- ADS makes it possible to select particles with one shape, one mass, and one composition, and characterize their properties
- Applied to compact inorganic salt particles, soot produced by miniCAST, SIDI, and GDICI engines
- Data can be used to determine particle primary spherule diameter, number of spherules per aggregate, fractal dimension, aggregate void fraction, and dynamic shape factors in different regimes

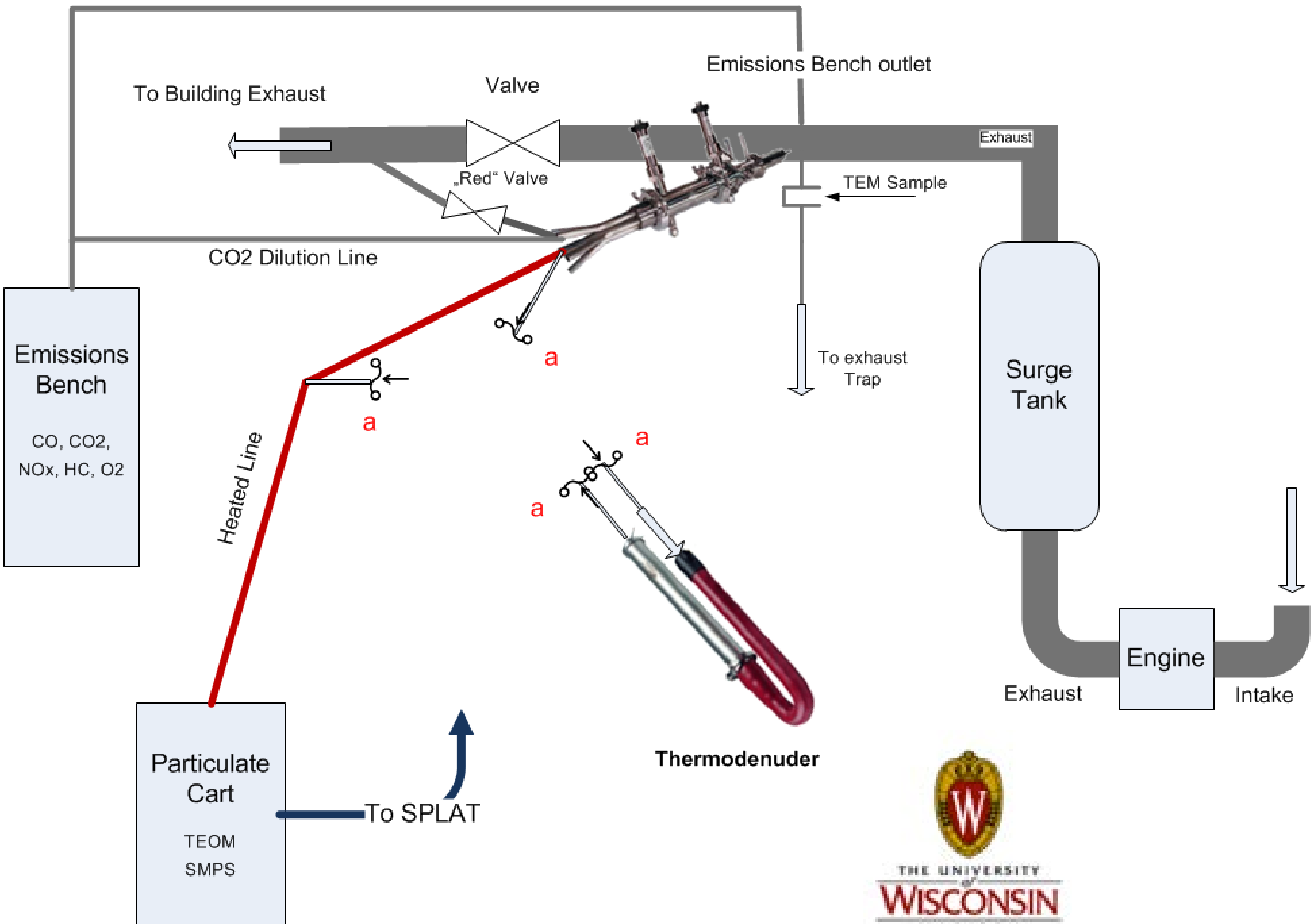


# Engine Research Center laboratory setup





# EEE Matrix

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| | Speed | Injection Timing | Fuel Quantity | AF | Φ | Spark Advance | Injection Press. | Intake Temp. | Oil Temp. | Coolant Temp. | Intake Manifold Press. |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | [RPM] | [°bTDC] | [mg/cyc] | [-] | [-] | [°bTDC] | [MPa] | [°C] | [°C] | [°C] | [kPa] |
| EOI 220 | 2100 | 220 | 11 | 15 | 0.975 | 25 | 11 | 45 | 90 | 90 | 40 |
| EOI 280 | 2100 | **280** | 11 | 15 | 0.975 | 25 | 11 | 45 | 90 | 90 | 40 |
| Heavy Load | 2100 | 220 | **21** | 15 | 0.975 | **18** | 11 | 45 | 90 | 90 | 66 |
| Rich | 2100 | 220 | 11 | **13** | **1.125** | 25 | 11 | 45 | 90 | 90 | 36 |
| MBT-15 | 2100 | 220 | 11 | 15 | 0.975 | **10** | 11 | 45 | 90 | 90 | 39 |
| Lean | 2100 | 220 | 11 | **17** | **0.860** | 25 | 11 | 45 | 90 | 90 | 53 |
| Medium Press. | 2100 | 220 | 11 | 15 | 0.975 | 25 | **8** | 45 | 90 | 90 | 38 |
| Cold Start | 2100 | **80** | 11 | 15 | 0.975 | 25 | 11 | **30** | **30** | **30** | 38 |

# E85 Matrix

| | Speed | Injection Timing | Calc. Fuel Quantity | AF | Φ | Spark Advance | Injection Press. | Intake Temp. | Oil Temp. | Coolant Temp. | Intake Manifold Press. |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | [RPM] | [°bTDC] | [mg/cyc] | [-] | [-] | [°bTDC] | [MPa] | [°C] | [°C] | [°C] | [kPa] |
| **EOI 220** | 2100 | 220 | 16.23 | 10.2 | 0.958 | 25 | 11 | 45 | 90 | 90 | 37 |
| **EOI 280** | 2100 | **280** | 16.23 | 10.2 | 0.958 | 25 | 11 | 45 | 90 | 90 | 37 |
| **Heavy Load** | 2100 | 220 | **32.55** | 10.2 | 0.958 | **18** | 11 | 45 | 90 | 90 | 61 |
| **Rich** | 2100 | 220 | 16.23 | **8.84** | **1.106** | 25 | 11 | 45 | 90 | 90 | 33 |
| **MBT-15** | 2100 | 220 | 16.23 | 10.2 | 0.958 | **10** | 11 | 45 | 90 | 90 | 35 |
| **Lean** | 2100 | 220 | 16.23 | **8.83** | **0.846** | 25 | 11 | 45 | 90 | 90 | 41 |
| **Medium Press.** | 2100 | 220 | 16.23 | 10.2 | 0.958 | 25 | **8** | 45 | 90 | 90 | 37 |
| **Cold Start** | 2100 | **80** | 16.23 | 10.2 | 0.958 | 25 | 11 | **30** | **30** | **30** | 36 |

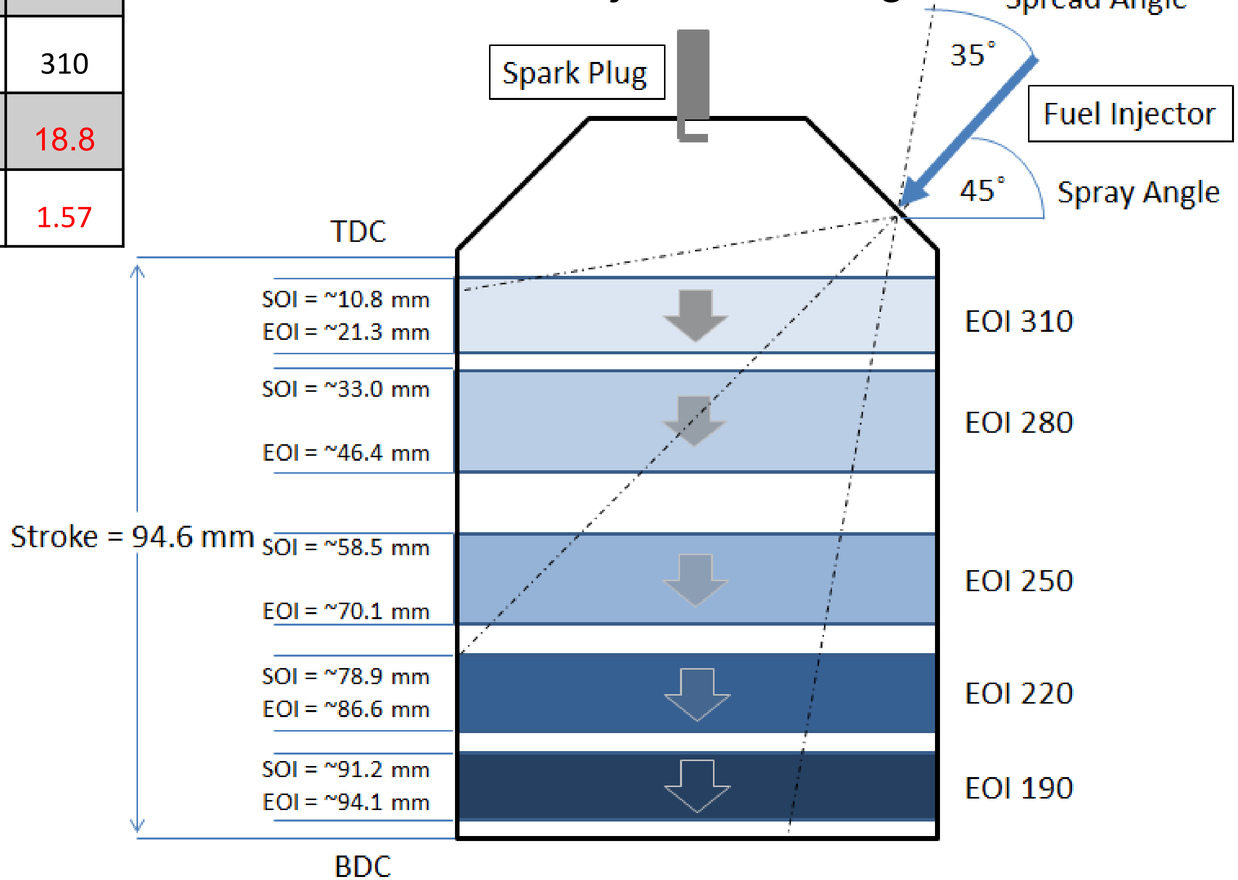
# Effect of EOI: Possible Piston/Wall Wetting

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

| Fuel | EEE | E20 | E85 |
|---|---|---|---|
| SOI [ºbTDC] | 324.8 | 325.4 | 328.8 |
| EOI [ºbTDC] | 310 | 310 | 310 |
| Injection Duration [ºCA] | 14.8 | 15.4 | 18.8 |
| Injection duration [ms] | 1.23 | 1.28 | 1.57 |

EOI 310 shows potential for piston and wall wetting

Spray Targeting Diagram
EEE Injection Timing

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