Particle free combustion:
What are the possibilities and tradeoffs?

M. Matti Maricq, Eric Kurtz
Chemical Engineering and Powertrain Engine Departments
Research & Advanced Engineering, Ford Motor Company

Introduction

With much attention over recent years focused on the application of diesel particulate filters (DPF) to control soot emissions from diesel engines, the question of particle free combustion conjures up visions of experimental new engine technologies that operate without PM formation (slide 2). Indeed, promising avenues of research in this direction exist. After a moments reflection however, one realizes that this goal has already been achieved by the port-fuel-injection (PFI) gasoline engine. Unlike diesel and early gasoline direct injection engines, PFI engines have been able to achieve PM emissions of <1 mg/km in US Federal Test Procedure and EU New European Driving Cycle tests, on the order of 10% of the current EU and US standards (slide 3). In this case, the question of particle free combustion is perhaps better interpreted in the context of eliminating PM emissions without compromises or tradeoffs. With rising concerns over energy independence and climate change, fuel economy is an important example. The PFI spark ignition engine provides the benefit of nearly PM free emissions, but it has lower fuel economy than other existing and developing engine technologies. The engineering task, therefore, is to innovate and optimize an engine with respect to emissions of PM, CO2, and other pollutants, as well as performance, safety, etc.

What we mean by "combustion particle"

Unlike gaseous emissions, such as CO and NO_x, particulate matter (PM) is chemically and physically heterogeneous (slide 5). It consists of solid as well as liquid particles. The solid particles comprise primarily of soot, but also small quantities of metals from lube oil additives. The liquid component includes condensed and nucleated heavy end hydrocarbons and sulfate. This composition, however, is not static. Rather it changes considerably from the time it exits the combustion cylinder, travels through the exhaust system, and exits the tailpipe (slide 6). Initially, PM in the hot exhaust leaving the engine consists only of solid particles. As the exhaust cools, hydrocarbons and sulfate can condense onto the soot. The extent of this depends on the presence of an oxidation catalyst and the sulfur content of the fuel. Nucleation of new particles depends also on these concentrations, as well as the extent and rate of dilution.

For the present discussion we will assume that "particle free" refers to soot particles. The issue of sulfate is one of fuel, not combustion technology. And since almost any combustion technology will require an oxidation, or three-way, catalyst, we will assume that this limits in addition the soluble organic fraction of PM.

Current technology (almost) particle free combustion: port-fuel spark-ignition engines

PM emissions from properly functioning, current model, PFI vehicles fall considerably below present emissions standards (slides 8 & 9). The PM emissions from these vehicles occur during vehicle accelerations. The PM levels are independent of fuel sulfur content (slide 8), but low sulfur fuel remains necessary for effective three-way catalyst operation. In this example, the PM consists of accumulation particles, presumably soot formed by transient rich operation or fuel impingement on piston or cylinder walls. Nucleation particles can be formed, but these are often present as storage/release artifacts from the transfer hose connecting tailpipe to dilution tunnel.

There are two principal reasons for very low PM emissions from PFI engines (slide 9): 1) the use of premixed combustion and 2) the accurate air/fuel ratio control that has evolved from efforts to improve performance and reduce costs of catalytic converters. During high acceleration, the relatively large injection of fuel may lead to incomplete volatization on the intake port and lead to soot formation. Also high load operation may require enrichment in order to protect the catalyst, which again increases soot. Low semivolatile PM emissions can also be achieved through catalytic removal of soluble organic fraction precursors and through low sulfur fuel.

Gasoline direct injection (GDI) engines have recently received considerable attention due to efforts to improve the fuel economy of spark ignition engines. The initial motivation was to use stratified operation in which fuel is injected late during the compression stroke. This provides the possibility to avoid the need to throttle the air intake and, thereby reduce pumping losses. However, late fuel injection reduces time for fuel vaporization and increases the possibility of fuel impingement on the piston, which in turn increase PM emissions (slide 10). Current efforts are focused on homogeneous operation GDI engines and derive energy efficiency improvements from downsizing and turbocharging.

Future technology approaches to particle free combustion

In diesel combustion, soot forms in the interior of the burning fuel spray. As oxygen diffuses in, especially after combustion is complete, most of the soot is oxidized (slide 12). In the lift-off region between injector and flame onset, oxygen is entrained into the fuel spray. Extending this lift-off region, for example by increasing dilution, decreasing temperature or reducing nozzle hole size, reduces the fuel per unit entrained air. This leads to leaner equivalence ratios at the lift-off length and reduces soot formation (slide 13).

Soot formation depends on the local air/fuel ratio and temperature. Soot forms at $\Phi > \sim 2.0$ and in the temperature range between about 1500 K and 2500 K (slide 14). Low temperature combustion (LTC) seeks to reduce soot by avoiding this region of equivalence and temperature via the use of high EGR rates, high fuel injection pressure, and early or late injection timing. There are many versions of LTC, including homogeneous charge compression ignition (HCCI), premixed charge compression ignition (PCCI), clean diesel combustion, etc. (slide 15). Engine dynamometer studies of LTC demonstrate lower smoke and NO_x emissions as compared to conventional diesel exhaust (slide 16). But hydrocarbon and CO emissions can be somewhat higher

.
Diesel fuel and gasoline represent, respectively, more and less reactive fuels. Higher reactivity is needed in diesel engines for compression ignition. Lower reactivity is required in spark ignited engines to avoid knock. Reactivity controlled compression ignition takes advantage of the different reactivities in a scheme that employs PFI gasoline and DI diesel to control combustion phasing and spread out heat release to achieve low emissions while improving efficiency. Use of both diesel fuel and gasoline leads to a staged combustion of the more reactive regions of the combustion chamber that have more diesel fuel, followed by the less reactive regions that have less diesel fuel. This extends combustion duration, lowers peak temperature, and can reduce NO_x and soot emissions to below current EPA Heavy Duty limits (slide 17).

The conventional view of HCCI is one of heat release via rapid chemical reactions rather than by flame propagation (slide 19). However, measurements of CO, CO_2, and HC emissions as a function of air/fuel ratio show a sudden transition from CO_2 to CO dominated exhaust as A/F increases above ~70 (slide 20). The high CO_2/CO and CO_2/HC ratios at A/F < 70 suggest flame propagation, although this may occur in separate domains within the cylinder. This possibility is consistent with a flammability limit that with compression heating extends to about A/F = 70. Above this value the mechanism shifts to rapid chemical oxidation.

PM emissions from HCCI combustion likewise exhibit two regimes. At A/F < ~70, PM is more soot-like, exhibiting primarily an accumulation mode (slide 21). With leaner combustion, the size distribution shifts to smaller particles reminiscent of a nucleation mode. High A/F ratios reduce soot, but nucleation mode particles arise from incomplete combustion and unburned fuel. A GDI engine was used for these HCCI emissions measurements. Thus, just as discussed in connection with stratified GDI combustion, fuel impingement could be responsible for the soot emissions observed at lower A/F ratios (slide 22).

The role of fuel

Fuel also plays an important role in PM emissions. The use of fatty acid methyl ester based biodiesel fuel, either neat or in blends with conventional diesel fuel, is generally found to reduce PM emissions from diesel engines. This reduction occurs in the accumulation mode of solid particles (slide 24). It is likely that the benefits of biodiesel blends could extend to the new combustion technologies discussed above and help them achieve "particle free" status. There are differences in the literature about the effectiveness of biodiesel blends to reduce particle number emissions, which likely arises from the effects of sampling methods, as well as fuel, on nucleation mode formation. The data in slide 24 were collected with hot dilution; thus, any effect on nucleation mode is suppressed.

The use of ethanol in gasoline likewise reduces PM emissions from PFI engines (slide 25), although these are already low with conventional gasoline fuel. This is generally linked to the presence of oxygen in the ethanol fuel. But other factors, for example aromatic content, may also play a role.

Conclusion – tradeoffs

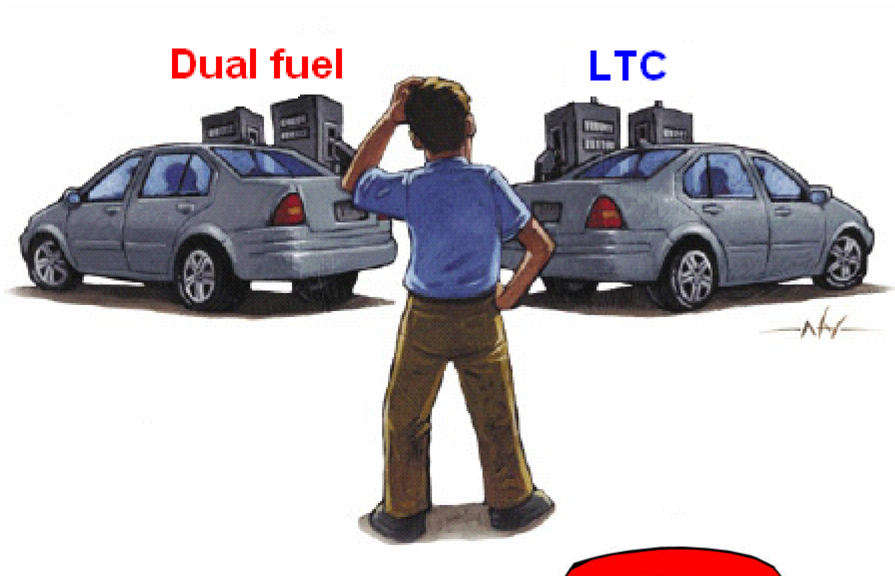
The answer to the question of particle free combustion is that: 1) On one hand this technology presently exists in the form of the PFI gasoline engine, but 2) Research continues to develop particle free engines that fulfill the goals of improved fuel efficiency and that provide the capabilities currently available from diesel engines (slide 26). While there are a number of attractive possibilities in this direction, there are also drawbacks / tradeoffs. As mentioned, PFI fuel economy needs to be improved to help meet most national and international CO_2 emissions targets. In the case of low temperature combustion, there is the need to overcome higher noise and higher CO and HC emissions. Extended lift-off combustion requires improvements in control strategies to deal with sensitivity to temperature, pressure, and fuel quality. Reactivity controlled compression ignition must overcome the drawback of needing dual fuel systems. HCCI and PCCI perform well, but have limited ranges of operation. And biofuels have tradeoffs with respect to fuel system durability and lower energy density. With continued research, however, hopefully one or more of these combustion technologies can overcome the drawbacks to provide "PM free" operation, good fuel economy and good performance.

Particle free combustion: What are the possibilities and tradeoffs?

Matti Maricq and Eric Kurtz
Chemical Engineering and Powertrain Engine
Research & Advanced Engineering
Ford Motor Company

14th ETH-Conference on Combustion Generated Nanoparticles

Particle free combustion?



Advanced combustion processes:

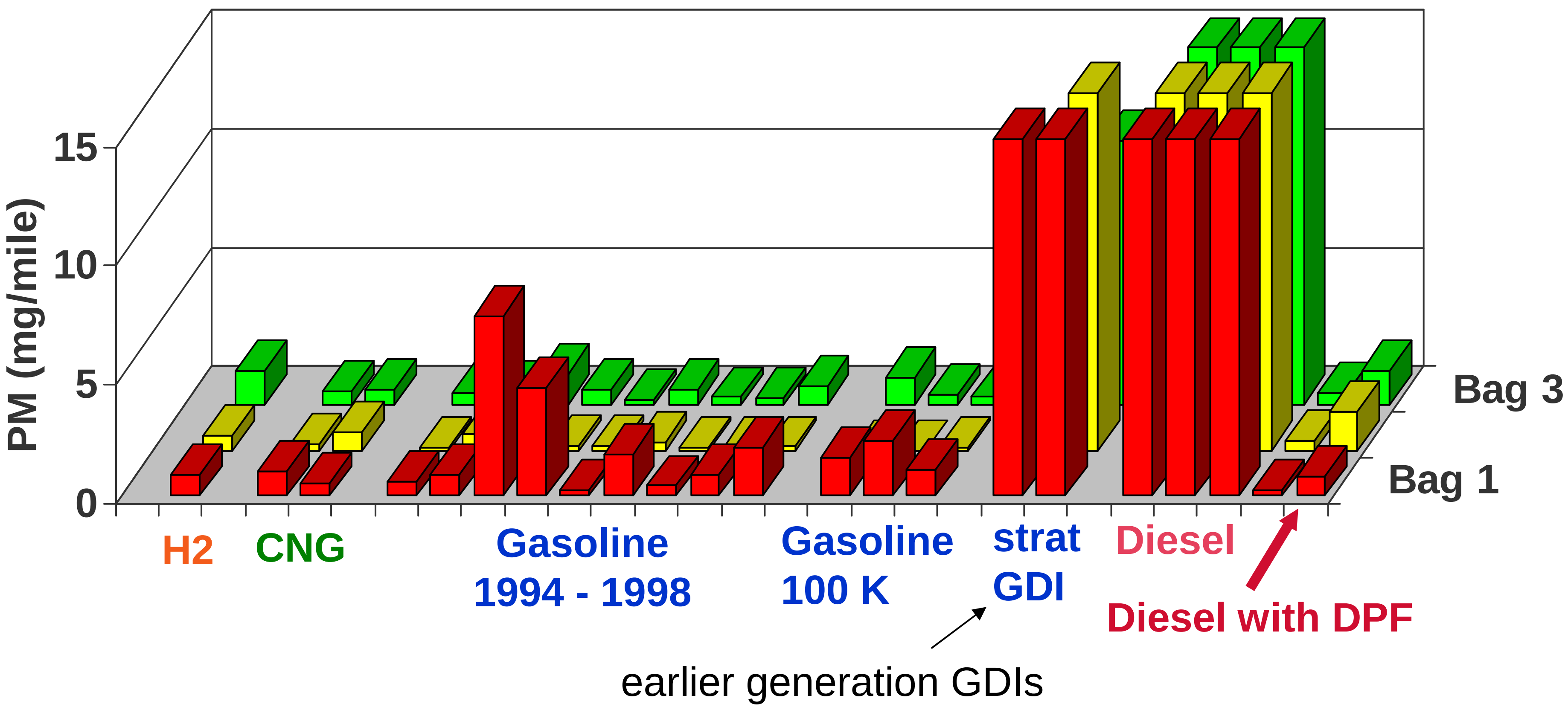
- Low temperature combustion
- Extended lift-off
- Homogeneous charge compression ignition
- Dual fuel

Simple answer:

- port fuel injection gasoline

PM mass emissions versus combustion technology

Gravimetric mass over FTP drive cycle



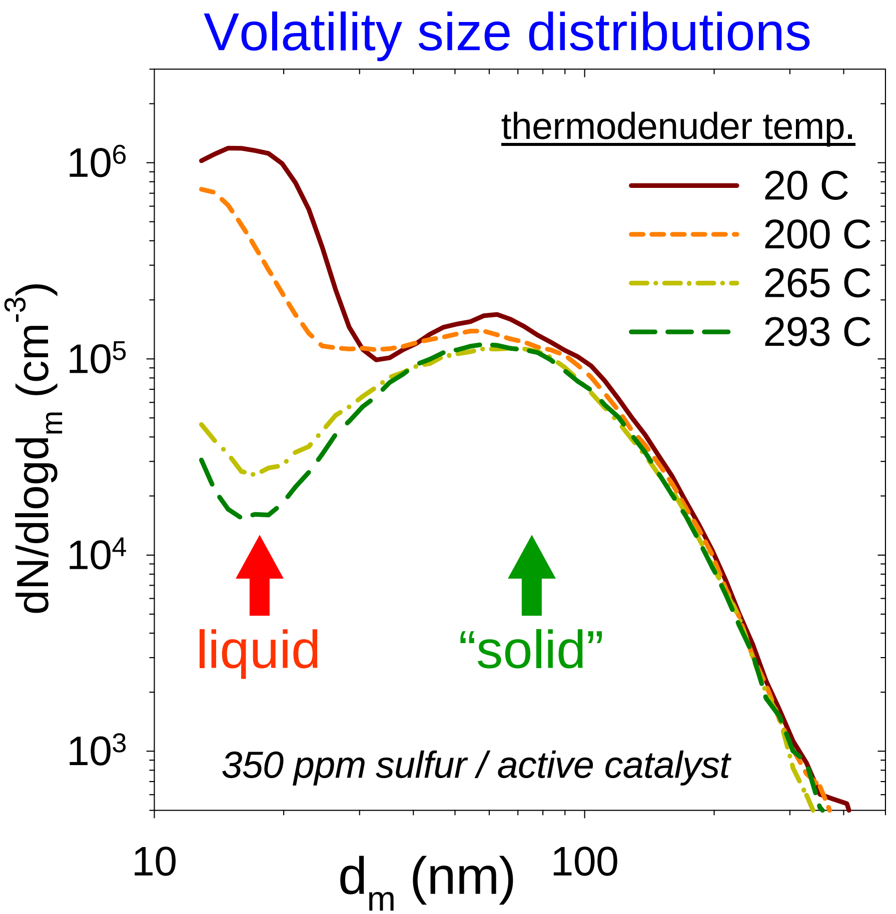
Environ. Sci. Technol. **1999**, 33, 1618

Outline

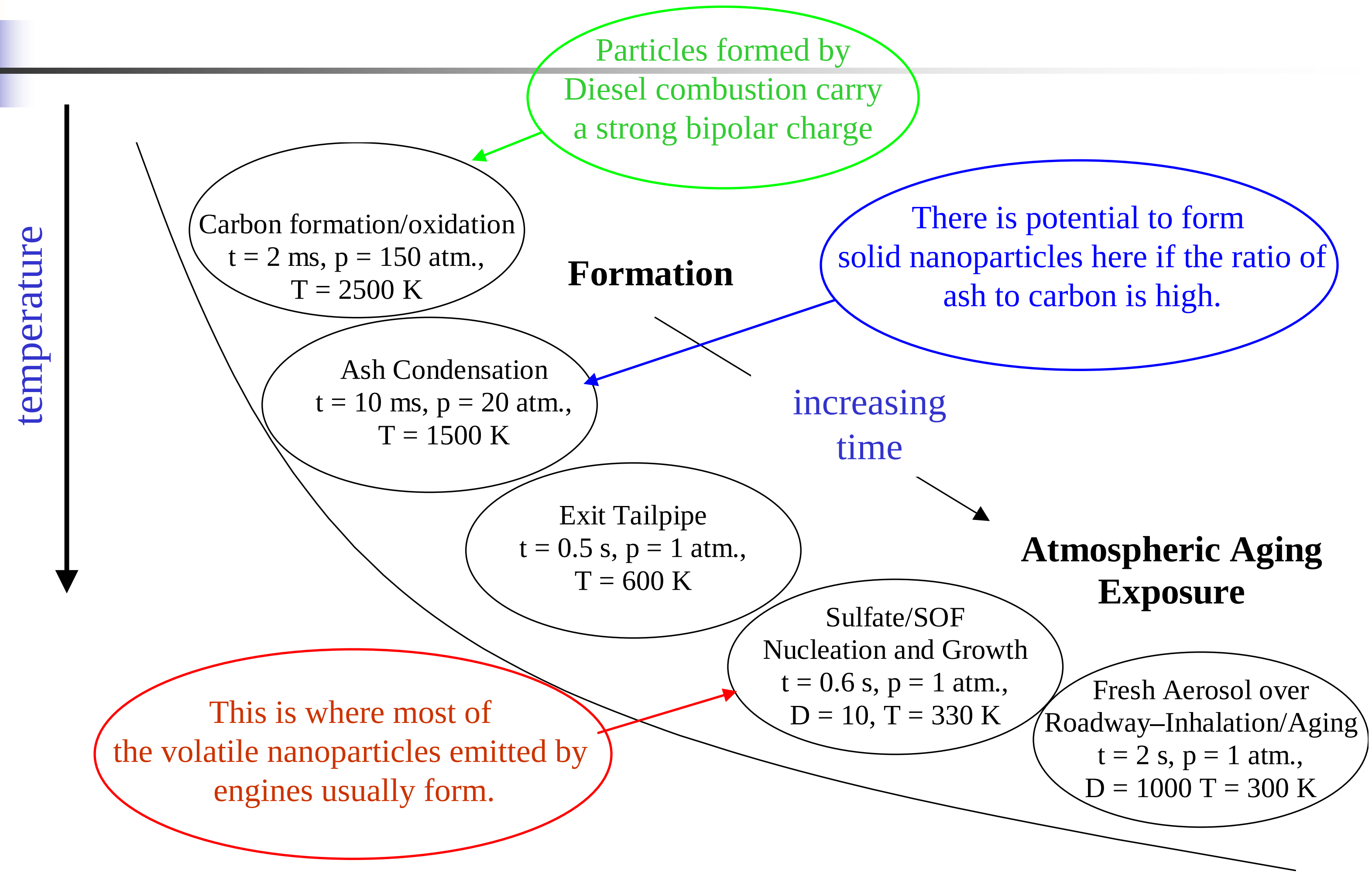
- What do we mean by “combustion particles”?
- Benefits / drawbacks of PFI
- Conditions for soot formation
- Low temperature combustion
- Dual fuel combustion
- Homogeneous charge compression ignition
- Fuels also play an important role

What does “combustion particle” mean?

- Engine exhaust PM is chemically & physically heterogeneous
- Soot with some metals from lube oil and fuel form solid particles
- Nuclei mode is usually semivolatile; disappears upon heating
  - Light duty diesel – high fraction sulfate
  - Heavy duty diesel – mostly lube oil (likely from higher oil consumption than LD)



Particle formation history by Prof. Kittelson: 2 s in the life of an engine exhaust aerosol

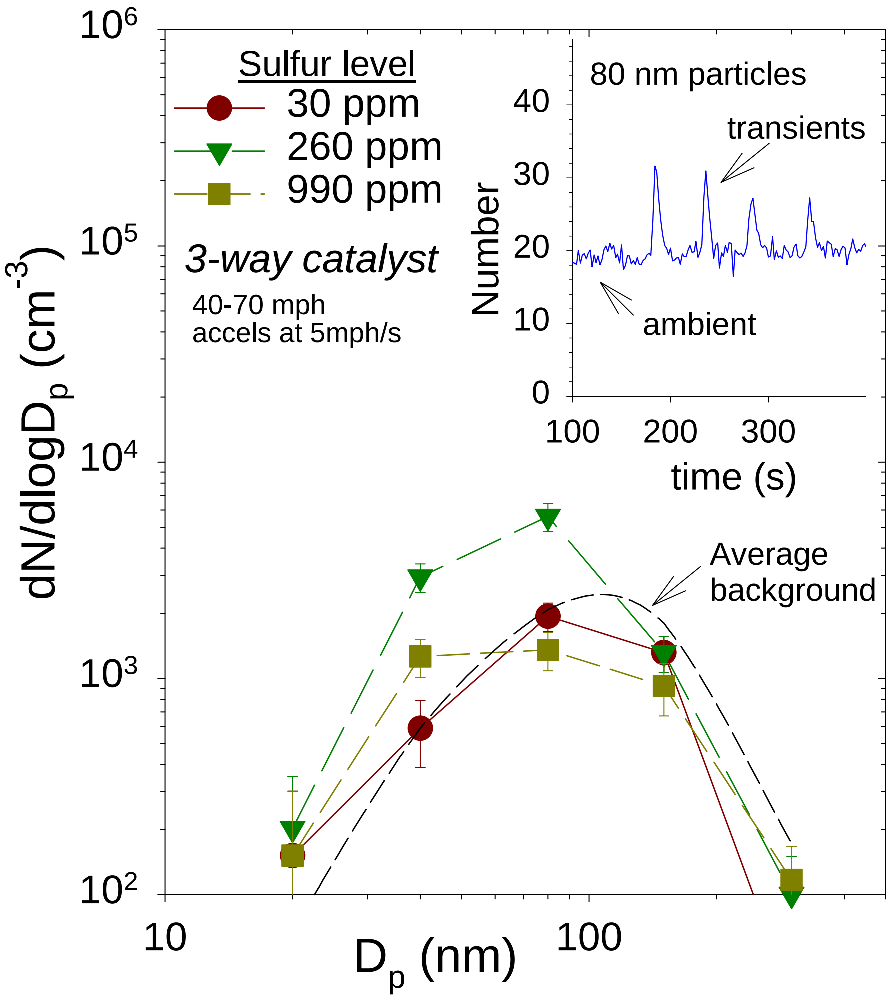


Kittelson, D. B., W. F. Watts, and J. P. Johnson 2006. “On-road and Laboratory Evaluation of Combustion Aerosols Part 1: Summary of Diesel Engine Results,” Journal of Aerosol Science 37, 913–930.

PFI gasoline engine PM

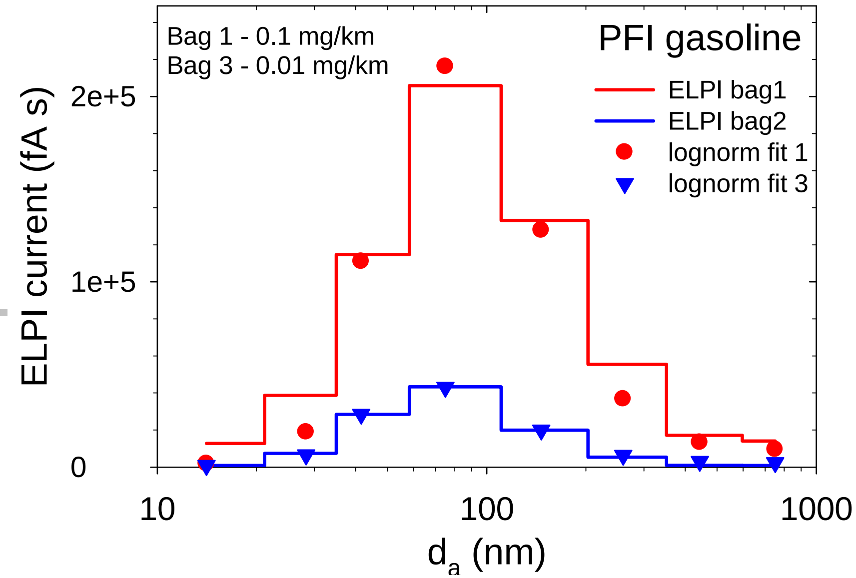
PFI vehicle in a wind tunnel

- Current gasoline vehicle PM near ambient
- Emissions occur during transients (inset)
- Distinct nuclei mode not observed, even at high sulfur levels



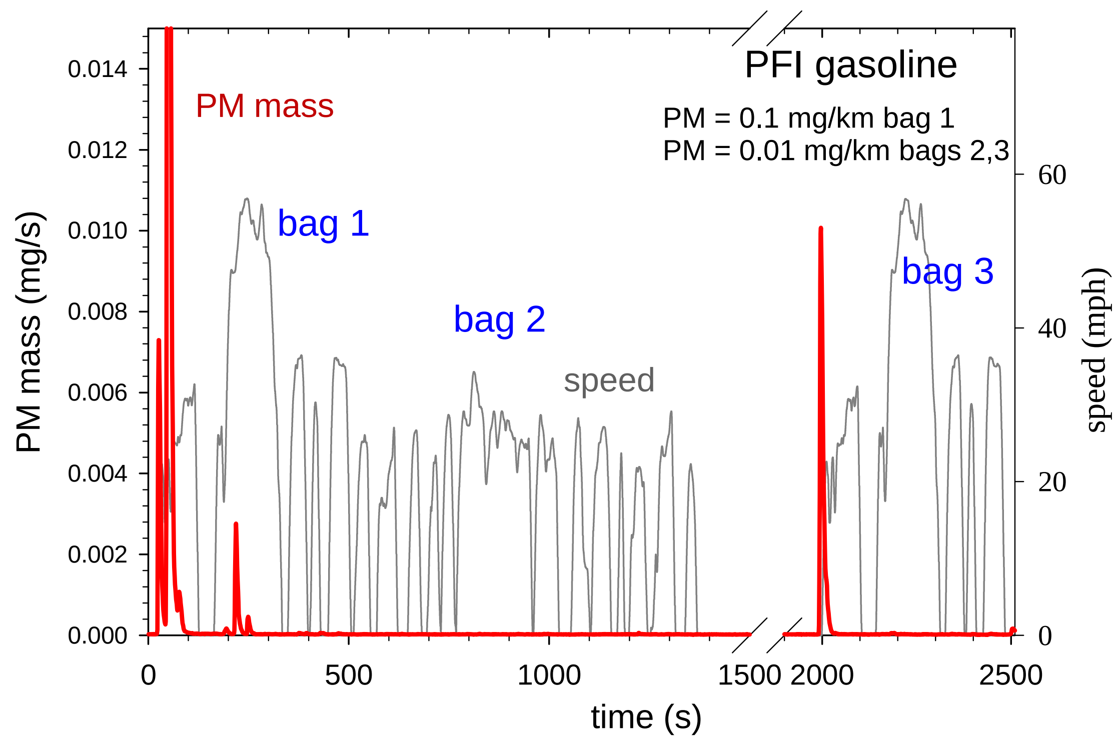
J. Air & Waste Manage. Assoc. 51, 174, 2001

Why is PM from PFI low?



Soot avoided by

- Efficient fuel volatilization
- Premixed combustion
- Very good air/fuel ratio control



Semivolatiles avoided by:

- Catalytic converter
- Low sulfur fuel

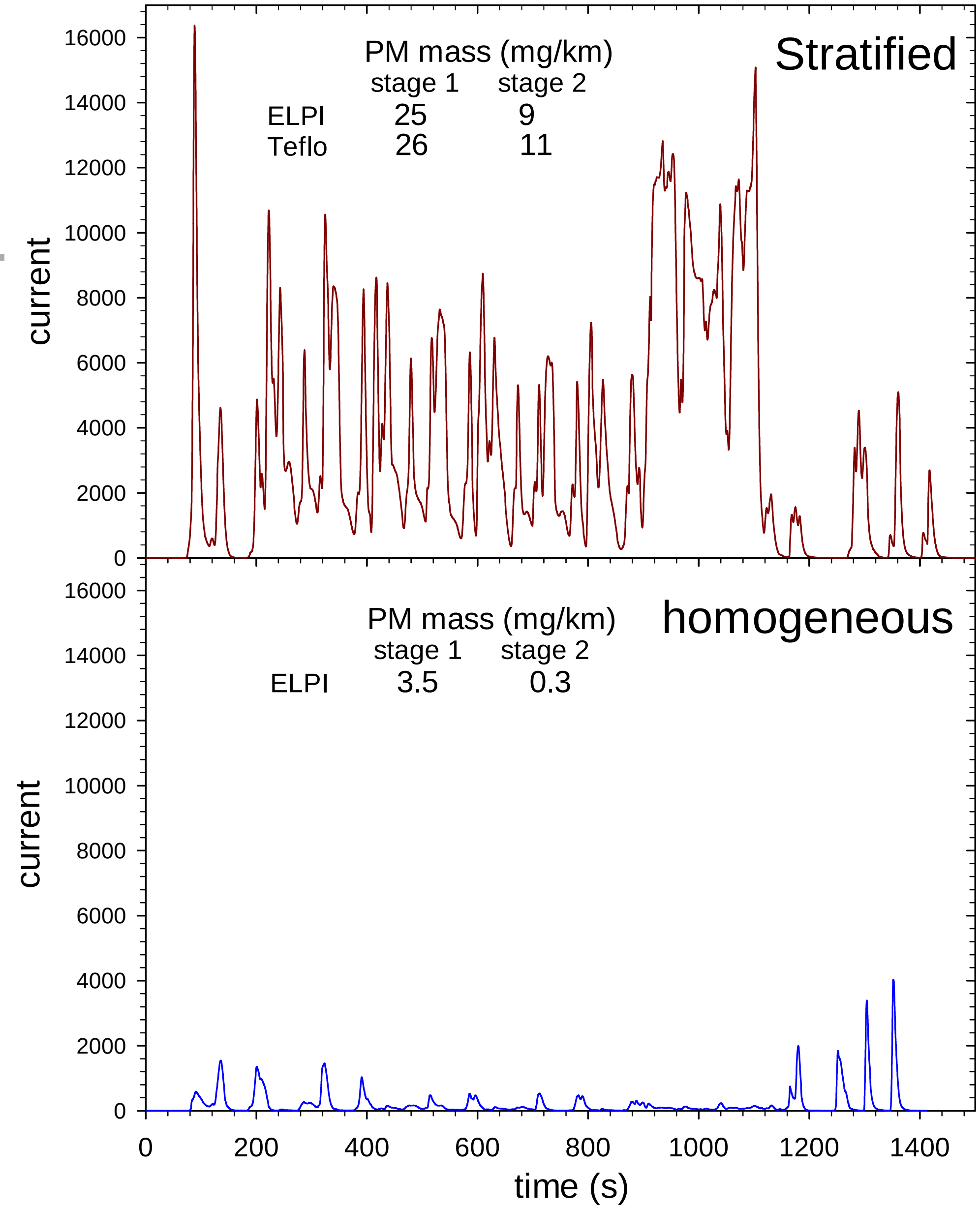
What's different about GDI?

Stratified operation (late injection) – soot from

- Piston wetting
- Lack of air / fuel homogeneity

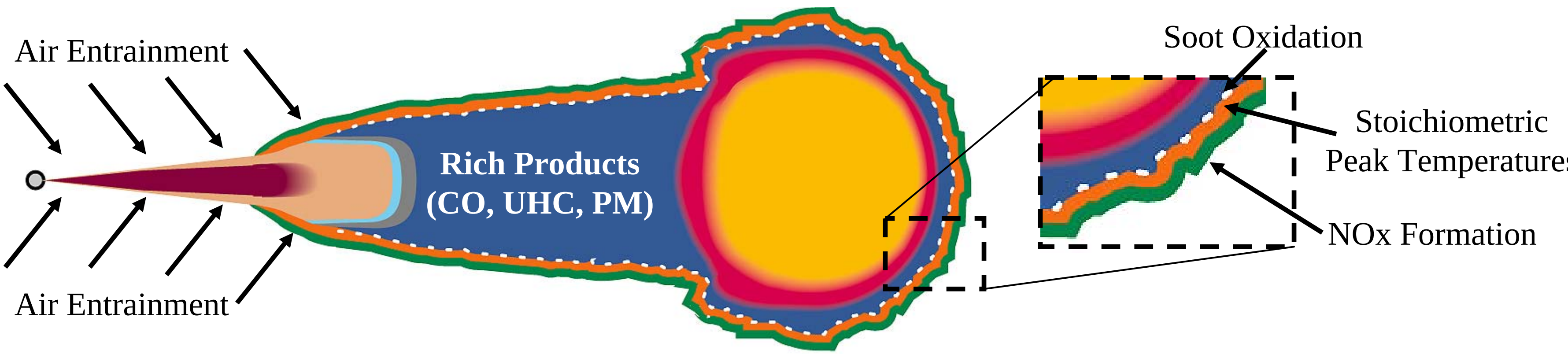
Homogeneous operation – soot can still arise from

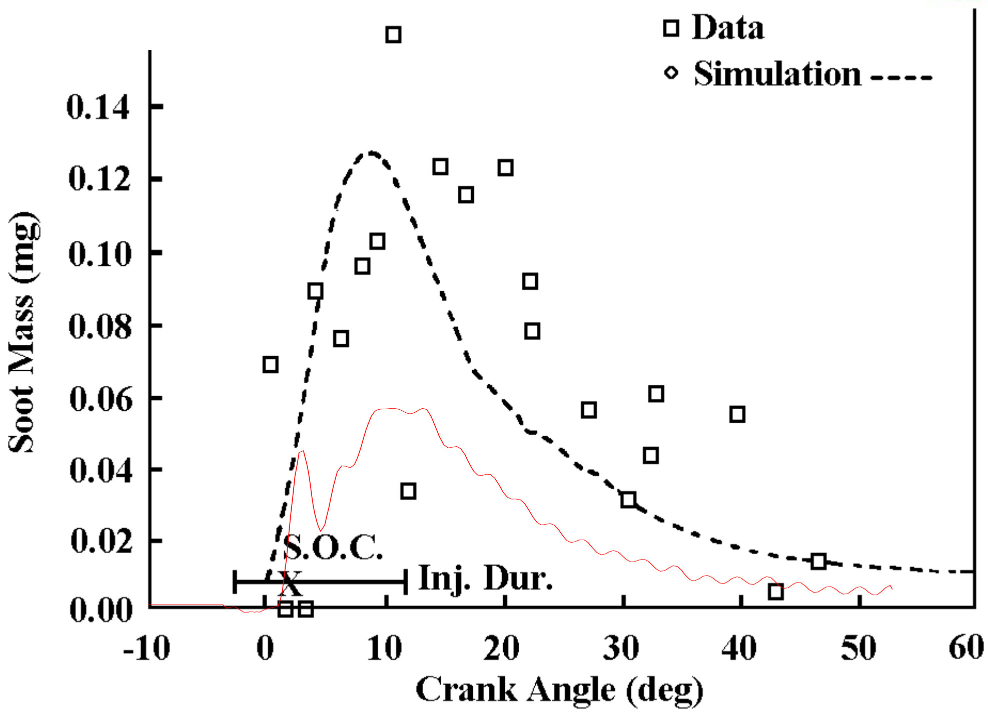
- Incomplete fuel volatization
- Piston / wall wetting



- What causes soot?
- Avoiding soot – low temperature combustion

Soot Formation and Oxidation





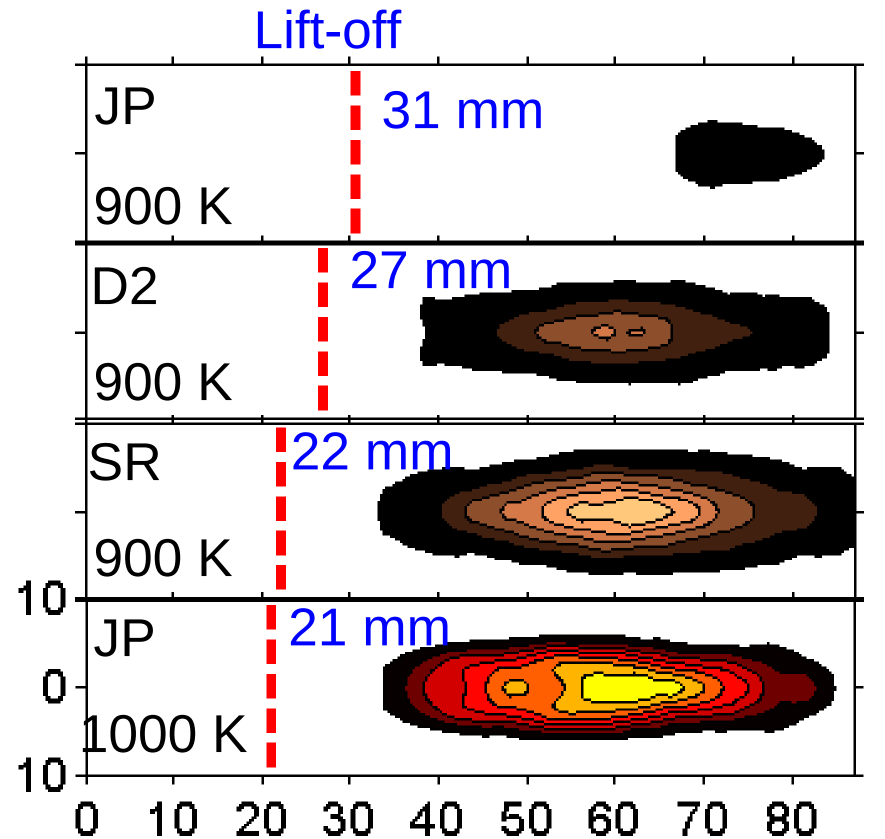
Kittelson et al. SAE 861569

- The bulk of the soot that is formed is oxidized before it leaves the engine
- Oxidation occurs as carbon finds oxygen
- Air utilization is a key for low soot emissions
- Air entrained upstream of the flame lift-off length can help reduce soot formation

e,g,, Flynn et al., SAE 1999-01-0509

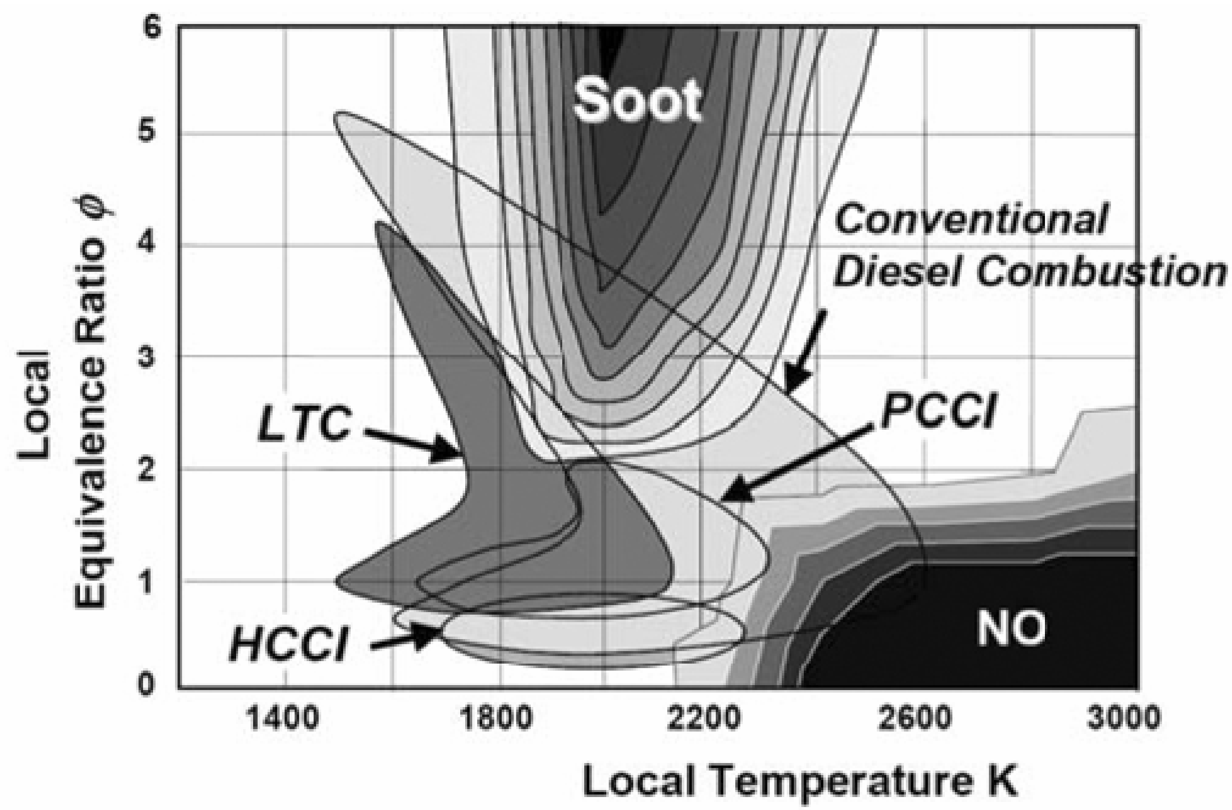
Reduce soot with extended lift-off

- Increase air entrainment into Fuel stream
- Increasing lift-off (e.g., by cetane) and decreasing orifice size entrains more air per unit fuel upstream of the lift-off length
- The added air decreases soot formation



Kook & Pickett, SAE **2009-01-2643**

Low temperature combustion



- In LTC soot formation is avoided by enabling fuel and air to mix prior to combustion. Increase ignition delay until equivalence < ~2.0
- Method: Early injection or late injection, high EGR rates, high fuel injection pressure

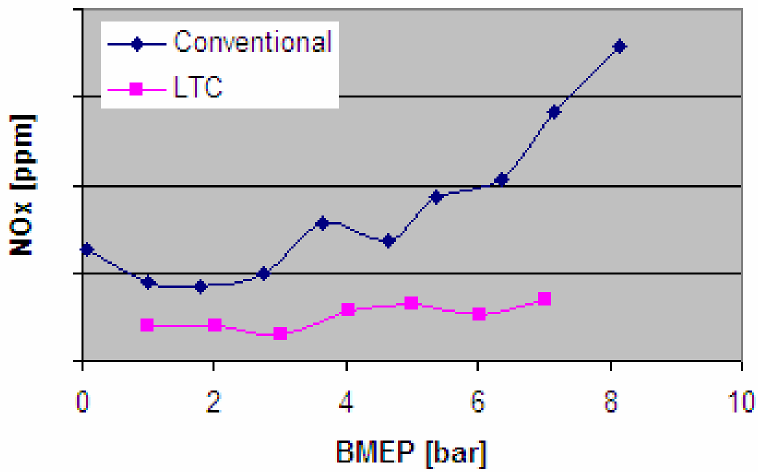
Hanson et al., SAE 2010-01-0864

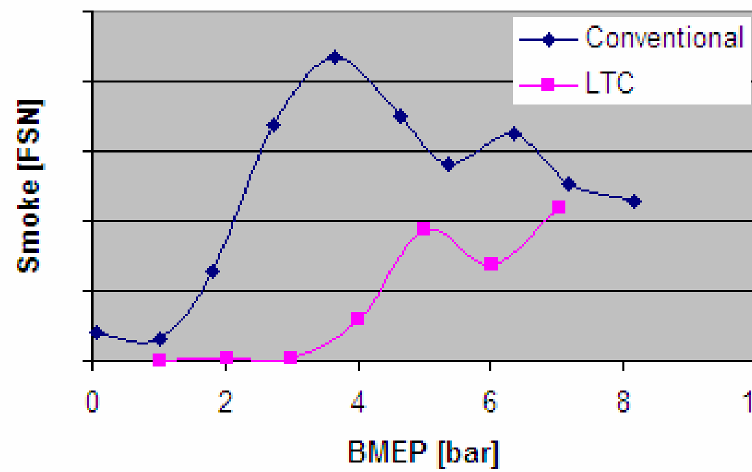
The many versions of LTC

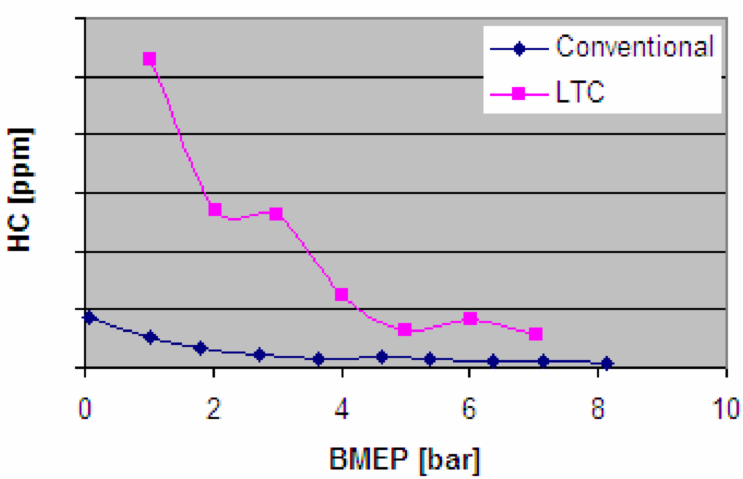
- Homogeneous Charge, Compression Ignition (HCCI)
- Modulated Kinetics "MK" Combustion
- Uniform Bulky Combustion (UNIBUS)
- Smokeless Rich Combustion
- Clean Diesel Combustion
- Premixed Charge, Compression Ignition (PCCI)

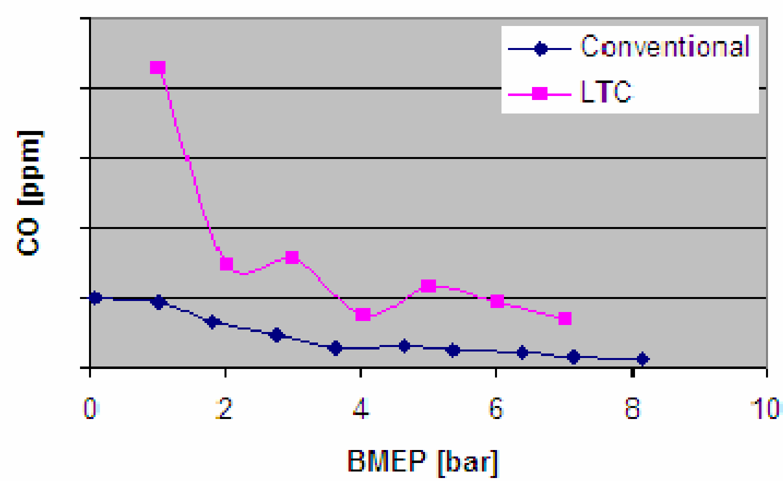
LTC vs. Conventional Combustion

- LTC has low NOx and low smoke vs conventional combustion
- Higher CO and HC are also normal
- Noise is a key tradeoff – expect higher noise in LTC



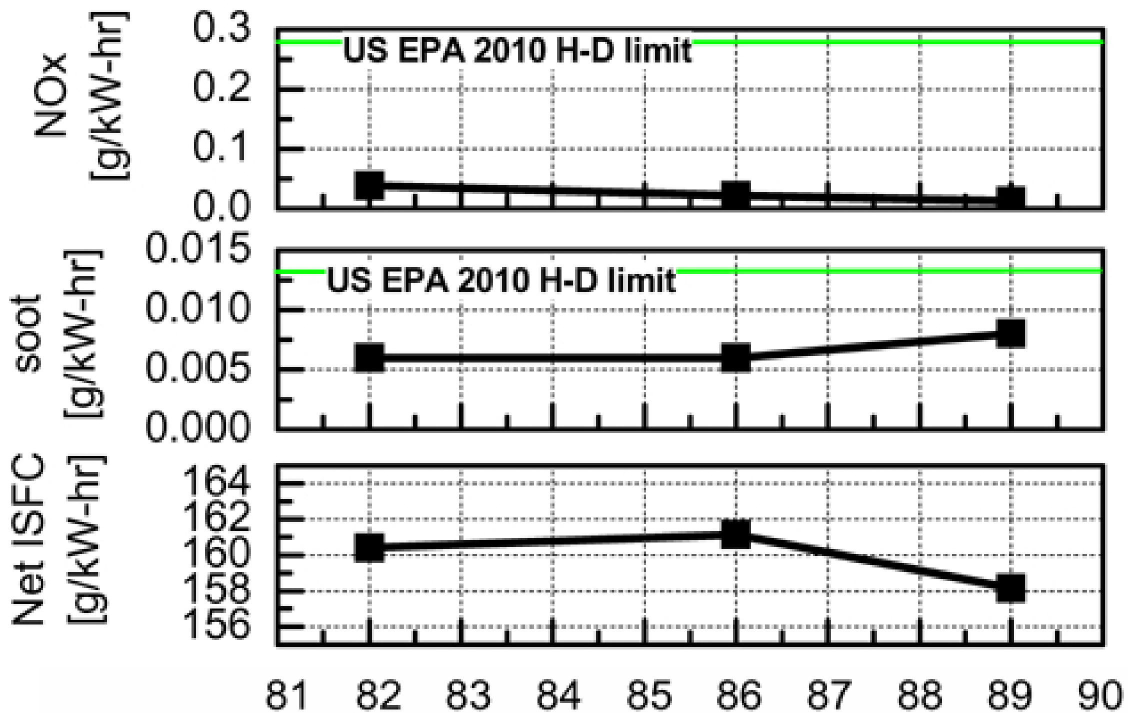






Dual fuel
Reactivity controlled compression ignition

- Use higher reactivity diesel and lower reactivity gasoline to control combustion phasing and heat release
- Retarded combustion lowers heat transfer, raises fuel economy
- Can lower NOx and PM below 2010 levels



Hanson, Kokjohn, Splitter, Reitz, SAE 2010-01-0864

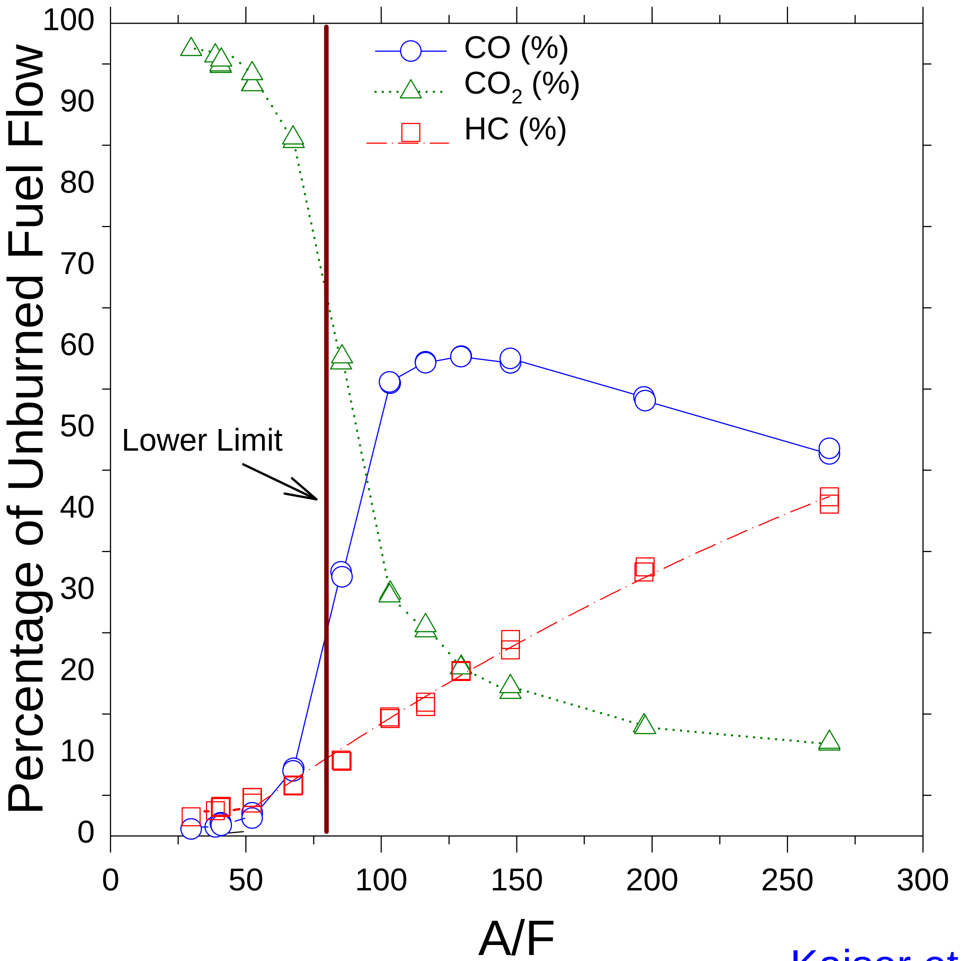
Homogeneous charge compression ignition

How does HCCI work?

Acknowledgements to Bill Kaiser, Jay Yang, Jason Cardinal, Tom Kenney

- *Conventional wisdom*.
  - Energy released by rapid chemical reactions – no flame propagation.
  - Naturally low PM and NOx emissions.
- *Speciated gaseous and PM measurements suggest that:*
  - Flame propagation might well occur up to a threshold A/F ratio of about 70:1.
  - PM emissions are low, but not negligible, and vary in nature with A/F ratio and injection timing.

HCCI engine gaseous emissions versus A/F

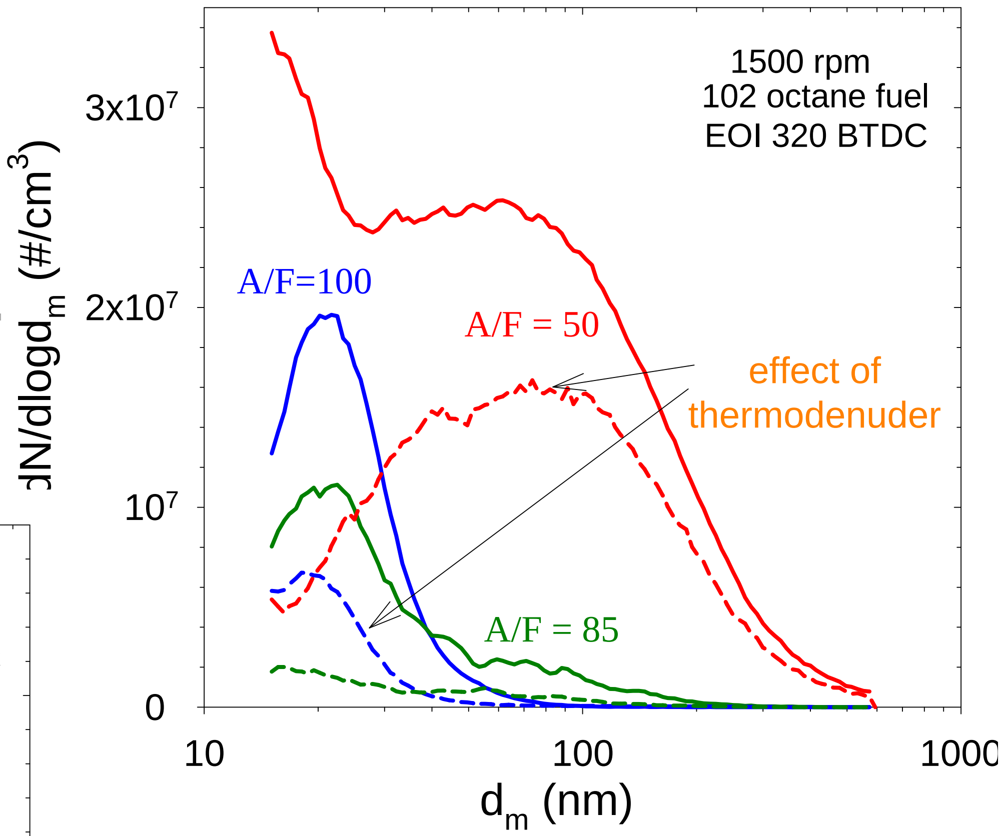


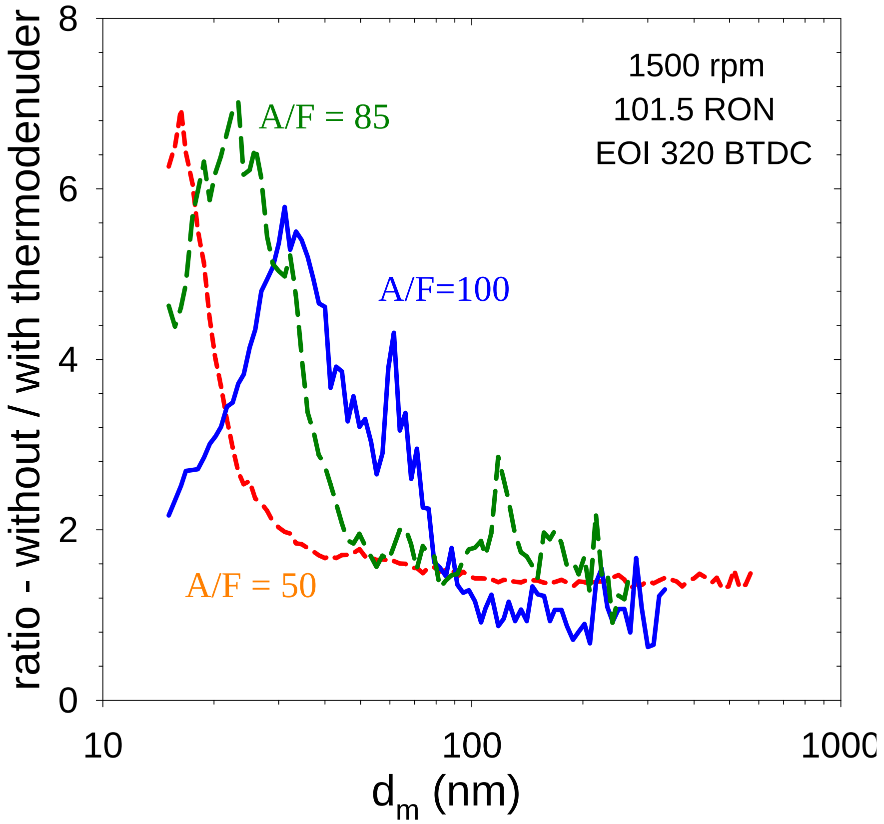
- Stable engine operation achieved by heating intake air (e.g., 160 C)
- With additional compression heating, flammability limit extends to A/F= ~70.
- At A/F= ~70 CO_2 rapidly decreases and CO increases.
- Suggests transition in combustion mechanism from flame propagation to rapid oxidation.

Kaiser et al. Int. J. Engine Research, **3**, 185 (2002)

HCCI: PM vs A/F

- PM emissions intermediate between diesel and gasoline

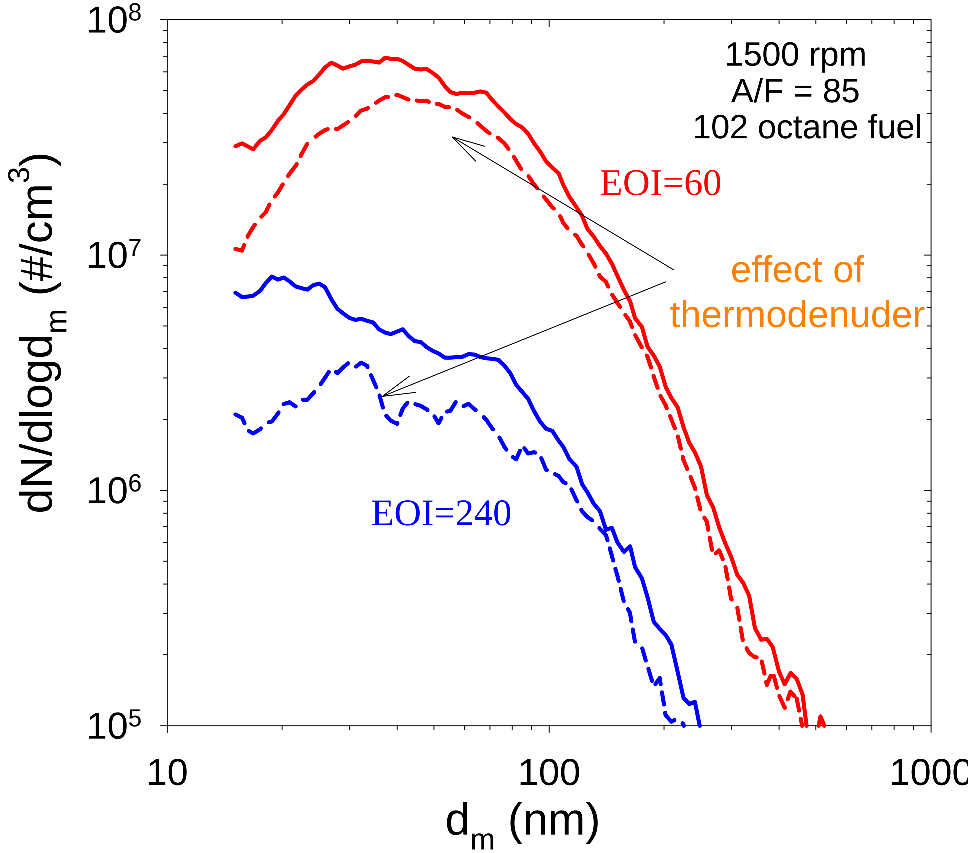




- Nature of PM changes at A/F = ~70.
  - More “soot” like at A/F < 70.
  - More semivolatile at A/F >70.

What causes PM in HCCI?

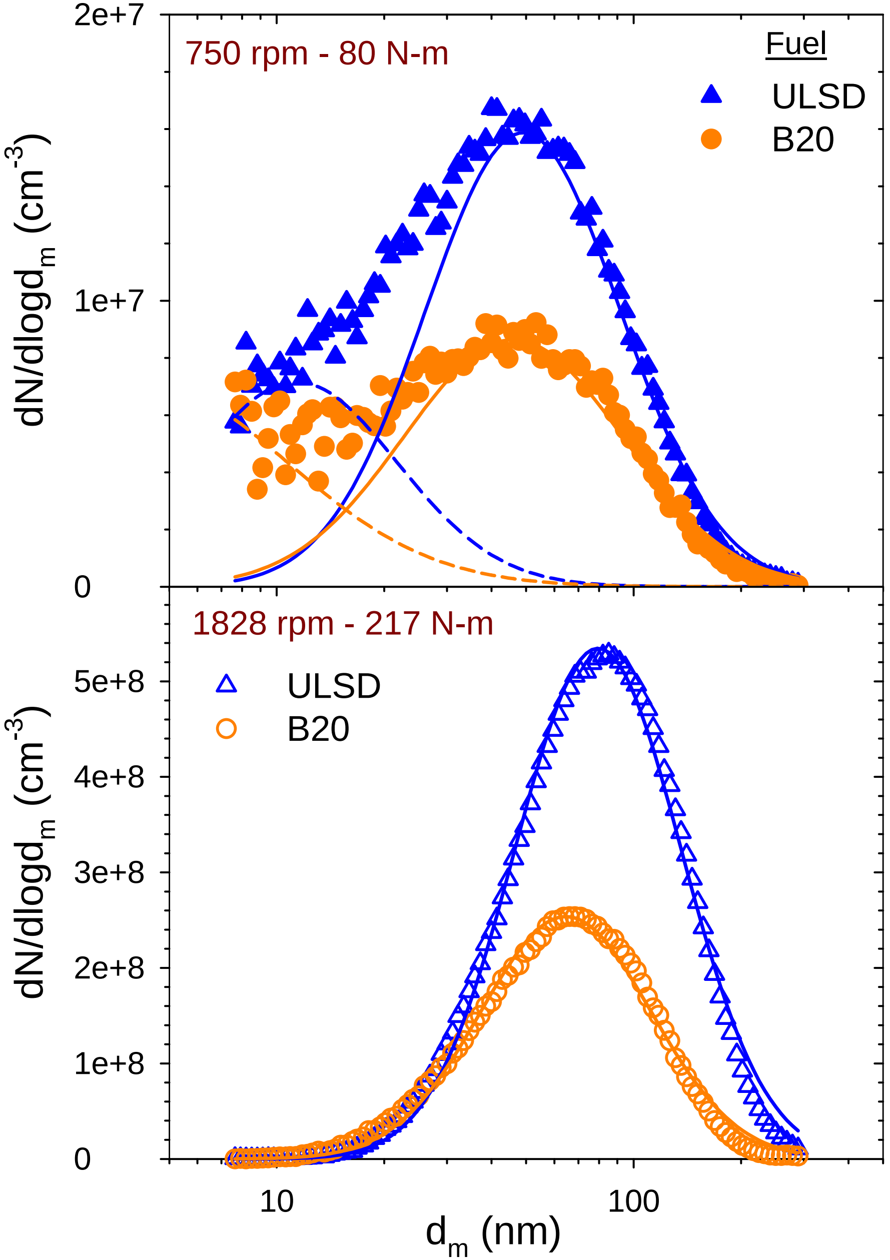
- Test engine was based on direct injection gasoline technology
- At low A/F soot is from piston inpingement and incomplete fuel volatization
- At high A/F semivolatile PM is from incomplete combustion and fugitive heavy end hydrocarbons



PM formation is a combination of engine technology and fuel

Petroleum vs biodiesel

- Size distributions show ~2x lower PM with B20
- Small, 0 – 20 nm decrease in size for B20
- Mostly single lognormal accumulation mode
- Small nucleation mode at idle and low load (hot diltuion)

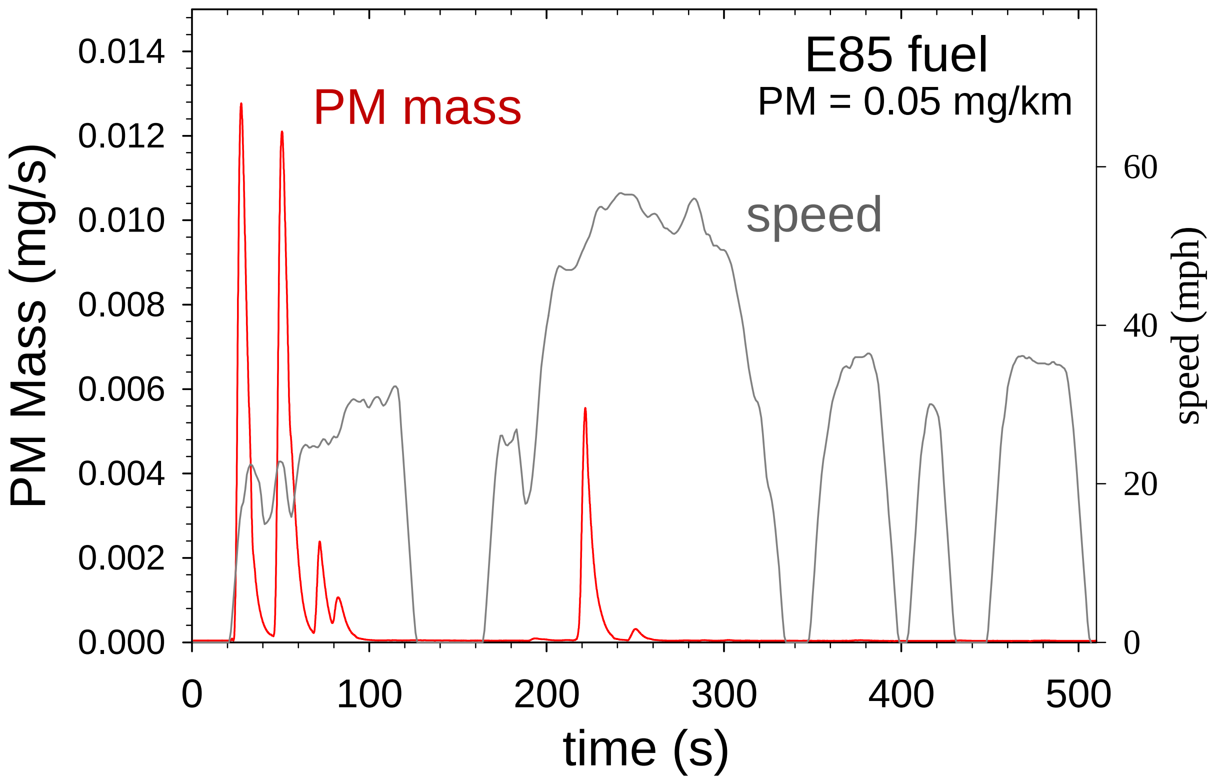


SAE 2010-01-0790

Ethanol blends vs gasoline

Mass emissions

- ELPI data converted to real time PM mass
- PM emissions occur during cold start – almost no PM after 300s
- Ethanol & biodiesel have lower energy density; so fuel economy and range is lower



Conclusions

Particle free combustion & tradeoffs

- PFI – needs improved fuel economy
- LTC – noise, HC and CO emissions
- ELOC – sensitive to pressure, temperature, and fuel properties
- Dual fuel – complexity of two fuel systems
- HCCI – limited operating range, can emit PM
- Biofuels – tradeoffs include availability, fuel system durability and energy density