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# Engine, Aftertreatment, and Fuel Quality Achievements to Lower Gasoline Vehicle PM Emissions: Literature Review and Future Prospects

*Executive Summary*

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# Executive Summary

## Motivation and Scope

Spark ignition gasoline vehicles comprise most light duty vehicles worldwide. They were not historically associated with PM emissions. This changed about 15 years ago when regulatory demands for fuel economy improvements and greenhouse gas emissions reductions prompted the widespread introduction of gasoline direct injection (GDI) engine equipped vehicles. Unfortunately, this change in engine technology was accompanied by an increase in PM emissions. Regulators worldwide responded with new or revised PM emissions standards. This review takes a comprehensive look at PM emissions from gasoline vehicles. It examines the technological advances that made it possible for GDI vehicles to meet even the most stringent tailpipe PM standards. These include fuel injection strategies and injector designs to limit fuel films in the engine cylinder that were pathways for soot formation and the development of gasoline particle filters to remove PM from engine exhaust. The review also examines non-exhaust PM emissions from brake, tire, and road wear, which have become the dominant sources of vehicle derived PM. Understanding the low levels of GDI tailpipe PM emissions that have been achieved and its contribution to total vehicle PM emissions is essential for the current debate about the future of internal combustion engines versus rapidly evolving battery electric vehicles. In this context, it does not make sense to consider BEVs as zero emitting vehicles. Rather, a more holistic framework is needed to compare the relative merits of various vehicle powertrains.

## Introduction

Due to potential adverse health and environmental impacts, particulate matter (PM) is one of the six criteria pollutants for which the Clean Air Act requires National Ambient Air Quality Standards to be established. Consequently, motor vehicle tailpipe emissions standards have included PM to some extent since their inception. Early on these addressed gross emissions from diesel vehicles, but had little impact on gasoline vehicles. The primary challenge for the latter was to meet hydrocarbon (HC) and $NO_x$ emissions standards. This was achieved by the development of the three-way catalyst, which removes HCs by oxidation to $CO_2$ and $NO_x$ by reduction to $N_2$. To accomplish both simultaneously requires tight control of the fuel / air ratio to remain near stoichiometric. Under this condition, PM emissions are synergistically reduced, since soot formation requires an equivalence ratio near 2, which is substantially removed from stoichiometric. Therefore, properly functioning gasoline vehicles easily met the 80 mg/mi California LEV and EPA Tier 1 emissions standards.

Beginning in the 2000's, this situation changed dramatically for two reasons: 1) Tailpipe PM emissions standards were tightened to the point that forced filtration of diesel engine exhaust and 2) Gasoline direct injection (GDI) technology was introduced to address requirements to improve fuel economy and reduce greenhouse gas emissions. The diesel particulate filter (DPF) was a rare example of engine aftertreatment technology that overachieved regulatory demands. The DPF reduced light duty diesel emissions sufficiently to meet the European Union (EU) 5b solid particle number standard of $6 \times 10^{11}$ particles/km. This is equivalent to <1 mg/mi, as compared to the 10 mg/mi California LEV II and EPA Tier 2 standards. Conversely, PM emissions from GDI vehicles increased relative to conventional port fuel injection (PFI) vehicles. As a result, gasoline vehicles became the "high emitters" in the motor vehicle fleet. Even though they met the existing 10 mg/mi PM emissions standard, concerns over "backsliding" resulted in a tightening of the standard to 3 mg/mi under Tier 3 and LEV 3.

In PFI engines soot emissions are low because fuel is injected onto the normally hot intake port, which vaporizes the fuel and provides good fuel air mixing as the mixture is drawn into the cylinder during the intake stroke. The direct injection of fuel into the combustion chamber in the GDI approach provides evaporative cooling and boosting opportunities that improve fuel economy, but also risks fuel spray impingement on the piston top and cylinder surfaces. The resulting presence of liquid fuel at spark ignition results in soot formation. Residual liquid fuel on the injector tip, which in GDI engines extends into the cylinder, presents another important soot source. Early GDI engines attempted to reduce pumping losses via a "stratified" combustion approach in which the engine was run overall lean, but with a local stoichiometric fuel / air ratio region at the spark plug. This is accomplished by fuel injection late in the compression stroke when the piston is close to the injector, which makes it difficult to avoid fuel impingement onto the piston. Later versions of GDI engines utilized the "homogeneous" combustion approach of early injection of a stoichiometric fuel air mixture.

The body of the report, summarized below, examines the following topics in detail: 1) Advances in GDI engine technology and operation to reduce PM formation, 2) Gasoline engine exhaust aftertreatment, including development of the gasoline particulate filter (GPF) and impact of the catalyst, 3) Impact of fuel composition, including ethanol and aromatic effects, 4) Role of non-exhaust PM emissions from brakes and tire wear, and 5) Future prospects for gasoline vehicles in the era of electrification.

## Gasoline Engine Technology Advances

Efforts to reduce engine-out PM emissions centered on reducing the presence of liquid fuel in the engine cylinder, primarily from fuel impingement and injector tip wetting. These combined technology improvements, such as the introduction of high pressure injectors with optimization of operation, such as fuel injection timing and breaking injection into multiple fuel pulses. Optimum injection timing involves a balance between fuel impingement and fuel air mixing. Fuel injection early in the intake stroke provides the best mixing due to high intake charge motion and long time prior to ignition, but risks impingement due to proximity of the piston to the injector. Injection at the end of the intake stroke greatly reduces impingement, but at the price of reduced charge motion and mixing time. Thus, the optimum, which depends on engine speed and load, lies near the midpoint of the intake stroke.

Another way to reduce fuel impingement is to lower the liquid spray penetration. Injection pressure and temperature play strong roles in this. Raising injection pressure increases spray velocity, which would appear to be counterproductive. However, it also lowers droplet size and increases evaporation rate, which outweigh the velocity effect and reduce liquid fuel penetration. Optimization of injector nozzle design and spray targeting helped further lower piston impingement. Raising fuel and injector tip temperature reduce spray penetration by enhancing fuel evaporation. However, care is needed to avoid flash boiling. This increases evaporation, but can collapse the liquid spray and thereby enhance penetration. It also exacerbates injector tip wetting and deposit formation which increase PM emissions.

Direct injection affords the possibility to divide fuel injection into multiple pulses. Since spray penetration increases with pulse length, this directly reduces impingement. The challenge is to fit the individual pulses into the optimum injection timing window; thus, this strategy is not uniformly effective at all speed - load points. One situation where this can be especially beneficial is during cold starts, when the low fuel evaporation rate makes impingement especially problematic. An alternative split injection approach that effectively combats PM emissions is the combination of direct and port fuel injection. Here PFI is used at low loads for its fuel evaporation benefit, whereas DI is introduced at higher loads for its evaporative cooling benefit.

Advances in GDI engines also addressed PM emissions from injector tip wetting. This occurs due to fuel injector imperfections, such as needle bounce during closing and residual fuel dribbling from the sac after end of injection and from flash boiling. The latter is perhaps the most important factor. Flash boiling expands the spray angle in the injector nozzle counterbore, which increases contact between the liquid fuel and nozzle. Reducing injector tip temperature to lower the propensity for flash boiling and raising injection pressure to reduce the time available both decrease wetting. Increasing valve closing speed, reducing bounce, and reducing sac volume also help limit the amount of liquid fuel available for wetting.

Repeated cycles of tip wetting and subsequent boiling during the combustion stroke can lead to deposits that provide additional surface for wetting and, thereby, enhance PM emissions. In GDI engines these are primarily fuel derived and consist of a carbonaceous matrix covered by large polyaromatic hydrocarbons. Deposit formation is mitigated by injector design improvements that lower wetting, injector tip temperature control to reduce complete boiling of the wetted tip, and newly developed fuel additives to deter deposit formation.

The ability of current GDI vehicles to meet LEV III and Tier 3 PM emissions standards without additional exhaust aftertreatment attests to the effectiveness of GDI engine technology advances. While further improvements are possible, it remains uncertain if they alone will be sufficient to meet the very stringent 1 mg/mi LEV III or the European Union solid particle emissions standards.

## Gasoline Vehicle Exhaust Aftertreatment Advances

The success of DPFs prompted calls for GDI vehicles to be equipped with gasoline particulate filters (GPFs). However, there are two important differences between diesel and gasoline engine exhaust that impact GPF implementation: 1) Diesel engines continuously produce soot, whereas GDI engines do so primarily during cold start and 2) Gasoline engine exhaust is considerably hotter than diesel exhaust, sufficiently hot to oxidize soot. These factors limit soot cake formation in GPFs, which is a major factor in the high filtration efficiency of DPFs. In spite of stoichiometric combustion, there is sufficient oxygen during events such as deceleration fuel cuts for passive oxidation of soot collected in GPFs.

To ensure low backpressure and preserve the fuel economy benefit of GDI engines, GPF design aimed for high porosity, but this yielded filtration efficiencies too low to meet the stringent initial efficiency requirements under regulations such as imposed by the Real Driving Emissions test procedure. In addition, filter washcoating was observed to further lower efficiency. The evaluation of in-use GPFs revealed that ash accumulation led to an increase in GPF efficiency. The ash forms a dendritic layer on the GPF walls that mimics a soot cake. This increases efficiency with little added backpressure. Current research is exploring means to produce artificial ash layers in GPFs to produce this benefit in a controlled manner and without the need of mileage accumulation. However, GPFs are already available for production use that meet the most stringent PM emissions limits, namely the EU solid particle standard over on-road RDE testing.

The three-way catalyst also plays an important role in reducing the PM emissions burden of gasoline vehicles. It does little to limit direct particle emissions, such as soot; rather it removes gaseous precursors to PM. There are two types of precursors: those for primary and secondary organic aerosols (POA & SOA). Both comprise HCs that exist in the gas phase at exhaust temperatures and, hence, are not removed by filtration, but are oxidized by the TWC. POA includes HCs that condense during exhaust dilution and sampling and, thus, are included in the regulatory PM mass measurement. SOA consists of HCs that are photochemically oxidized in the atmosphere into the aerosol phase. Recent smog chamber measurements reveal SOA declines with increasing emissions stringency (pre-LEV to SULEV) that correlate with non-methane organic gaseous emissions reductions.

## Fuel and Lube Oil Impacts

Fuel composition has a significant impact on gasoline vehicle PM emissions. For hydrocarbon fuels this is nicely captured by a PM index (PMI) defined by the ratio of double bond equivalent to vapor pressure summed over all compounds in a given fuel. Though empirical, it captures the essence of soot formation in a gasoline engine. High double bond equivalence correlates with aromatics, which have a chemical propensity to form soot and low vapor pressure correlates with a low evaporation rate and, hence, a compound's propensity to remain liquid on the piston top or injector tip. Efforts to simplify this to just bulk fuel properties have had limited success due to the fact that the major sooting components in gasoline contribute little to the bulk fuel vapor pressure.

There has been considerable investigation into the impact of ethanol on PM emissions owing the market transition from gasoline to E10 fuel. Many studies of splash blended and pump E10 fuel show small declines in PM emissions, However, more careful studies of matched sets of gasoline versus E10 fuels on larger test fleets have shown that the ethanol containing fuels have slightly higher PM emissions, even though high ethanol blends exhibit very low PM. Laboratory studies have identified a mechanism for this, by which evaporative cooling of aromatic fuel species by ethanol leads to a PM emissions increase.

Lube oil is traditionally associated with PM emissions. While this remains an issue for older and poorly maintained vehicles, lube oil appears to present a small contribution to GDI PM emissions, even at current regulatory standards. The increase in oil change interval for light duty vehicles from 3000 to 10000 miles likely plays a role in this. Lube oil ash content has two effects: under modest mileage accumulation it increases GPF efficiency, but at high mileage it can impact GPF full useful life.

Formulating an optimal fuel for PM emissions requires care. Reducing aromatic components aids in multiple ways: it reduces fuel PMI and also reduces injector tip deposits. Other properties can have opposing effects; thus, increasing T90 can help reduce flash boiling, but exacerbate PMI.

## Non-Tailpipe PM Emissions

As engine exhaust PM emissions have declined, non-exhaust contributions have become relatively more important. These sources include brake wear, tire wear, road wear and resuspended road dust. These emissions are more difficult to quantify since they are dispersed about the vehicle in contrast to engine exhaust which is localized to the tailpipe. Brake PM emissions depend solely on vehicle parameters, namely rotor speed, rotor temperature, and brake pressure. The PM is in general bimodal. The main contribution is wear debris from abrasion, which consists of particles in the 1 - 10 micron size range. At high brake temperatures a sub-micron mode of pyrolyzed HCs can form from the binders used in brake pads. This mode contributes little to the mass of brake PM, but dominates particle number when present. PM from tire wear depends both on vehicle parameters, speed, acceleration, tire pressure, etc. and non-vehicle parameters such as road surface characteristics. The emissions derive almost entirely from abrasion and can exhibit a very wide size range from 1 - 200 microns. Road wear and resuspended road dust particles exhibit a similar size range.

There has recently been considerable progress to develop a brake dynamometer approach to measure brake PM emissions both from the sampling and drive cycle perspectives. These reveal emissions rates of about 6 - 10 mg/mi, considerably higher than tailpipe PM levels. The ability to record tire wear emissions is more difficult and has lagged behind. However, estimates place tire PM emissions rates as comparable to brake PM. Road wear estimates are similar in magnitude. Thus, in total, light duty vehicle non-exhaust PM emissions amount to roughly 30 mg/mi as compared to the 3 mg/mi regulated for tailpipe emissions under LEV III and Tier 3.

A number of opportunities exist to mitigate non-exhaust PM emissions. Vehicle weight reduction is one major option that lowers emissions from all non-exhaust PM sources, brake, tire, and road wear. Synergistic benefits include improved fuel economy and lower greenhouse gas emissions. Additional means to reduce brake wear include the adoption of lower wear brake pads, ceramic rotors, and regenerative braking. Tire wear can also be reduced by suitable materials selection, but care is needed to ensure good traction over a wide range of road and weather conditions.

## Future Prospects for Gasoline Vehicles

The question of the gasoline vehicle's future prospects comes at a time of rapid progress in vehicle electrification. Given this progress, a number of regulatory decisions world wide are limiting or calling for the phase-out of internal combustion engines within the next decade or two. Much of the decision process is predicated on the notion that battery electric vehicles (BEV) are zero emitting. However, this appears to consider only tailpipe emissions. In the broader context, BEVs are non zero emitting. This is demonstratively the case in terms of PM emissions. In fact, comparisons of brake and tire wear emissions show them to be higher for BEVs than comparable model gasoline vehicles. The primary reason is vehicle weight; the battery increases BEV weight by about 20% relative to the gasoline vehicle. This is sufficiently large that the BEV's capacity for regenerative braking does not overcome the increase in brake wear, and no such compensating mechanisms exist for tire and road wear. Similarly, if life cycle energy costs are included, BEVs are not zero greenhouse gas emitting vehicles.

While a sustainable energy future is certainly an important goal, this can be achieved by multiple means, including battery electric, fuel cell, sustainable fuels, to name a few. It remains unclear if an immediate transition from internal combustion engine to battery propulsion is either environmentally or economically the most prudent path forward. The gasoline engine has made substantial progress in terms of emissions and fuel efficiency, meeting increasingly stringent regulatory requirements. With respect to PM emissions it is certainly competitive with BEVs and worthy of consideration in our transportation future.