ANL/ES/CP-101259

Particulate Emission Characteristics of a Port-Fuel-Injected SI Engine

Sreenath Gupta, Ramesh Poola, Kyeong Ook Lee, and Raj Sekar
Argonne National Laboratory
Argonne, IL 60439

ABSTRACT

Particulate emissions from spark-ignited (SI) engines have come under close scrutiny as they tend to be smaller than 50 nm, are composed mainly of volatile organic compounds, and are emitted in significant numbers. To assess the impact of such emissions, measurements were performed in the exhaust of a current-technology port-fuel-injected SI engine, which was operated at various steady-state conditions. To gain further insights into the particulate formation mechanisms, measurements were also performed upstream of the catalytic converter.

At all engine speeds, a general trend was observed in the number densities and mass concentrations: a moderate increase at low loads followed by a decrease at mid-range loads, which was followed by a steep increase at high loads. Within reasonable bounds, one could attribute such a trend to three different mechanisms. An unidentified mechanism at low loads results in particulate emissions monotonically increasing with load. At medium loads, wherein the engine operates close to stoichiometric conditions, high exhaust temperatures lead to particulate oxidation. At high loads, combustion occurs mostly under fuel-rich conditions, and the contribution from combustion soot becomes significant.

Estimates of the number of particles emitted per kilometer by a vehicle carrying the current test engine were found to be lower than those from a comparable diesel vehicle by three orders of magnitude. Similar estimates for mass emissions (grams of particulates emitted per kilometer) were found to be two orders of magnitude lower than the future regulated emission value of 0.006 (g/km) for light-duty diesel vehicles. Moreover, considering the fact that these particles have typical lifetimes of 15 min, the health hazard from particulate emissions from SI engines appears to be low.

Nomenclature

BSPM	Brake specific particulate mass (g/kWh)
BSPN	Brake specific particulate number (no. of particles/kWh)
CPC	Condensation Particulate Counter
EGR	Exhaust gas recirculation
M	Mass concentration, (g/cm^3)
N	Number density, (number of particles/cm^3)
WOT	Wide-open throttle
TEOM	Tapered element oscillating microbalance
ϕ	Equivalence ratio

1.0 INTRODUCTION

Because of stringent emission standards imposed by EPA and other such government agencies, particulate emissions from spark-ignited (SI) gasoline engines have come under close scrutiny. Numerous studies in the recent past have addressed various issues related to particulate emissions from these engines. Most successful attempts were those of Graskow and coworkers (1998) at the University of Minnesota, who observed intermittent spikes in the particulate emissions of a 1993 D-specification General Motors Quad-4 Engine. The spikes in number density were approximately two orders of magnitude higher than the baseline emissions and were found to consist mainly of volatile particles with sizes below 30 nm. Kayes and Hochgreb (1998) studied the dilution process and recommend a dilution ratio between 13 and 18. Shin and Cheng (1997), in their remarkable study of an SI engine with the Exhaust Gas Recirculation (EGR) defeated, identified that particulates under normal operating conditions (ϕ~1.0) result from the combustion of lubrication oil. The contribution from combustion soot was found to be significant only at high loads (ϕ >1.2).

Despite such studies, questions remain regarding the environmental impact of particulate emissions from the current-technology engines. The objectives of the present study are to characterize particulate emissions from one such engine and to assess the significance of their contribution to the environment. As engine-out

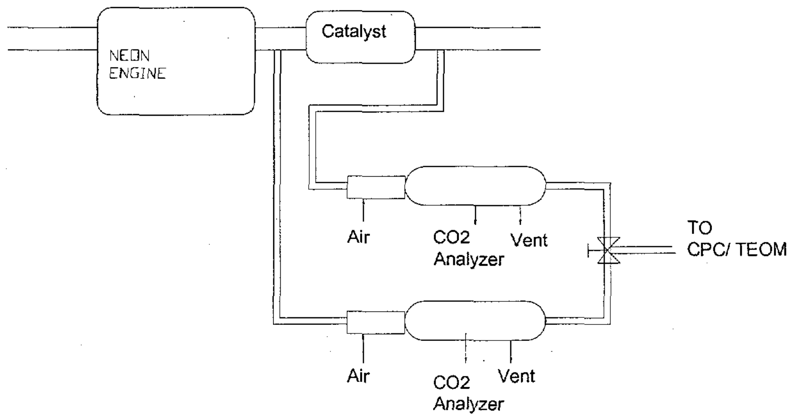

Fig. 1. A schematic of the exhaust sampling system.

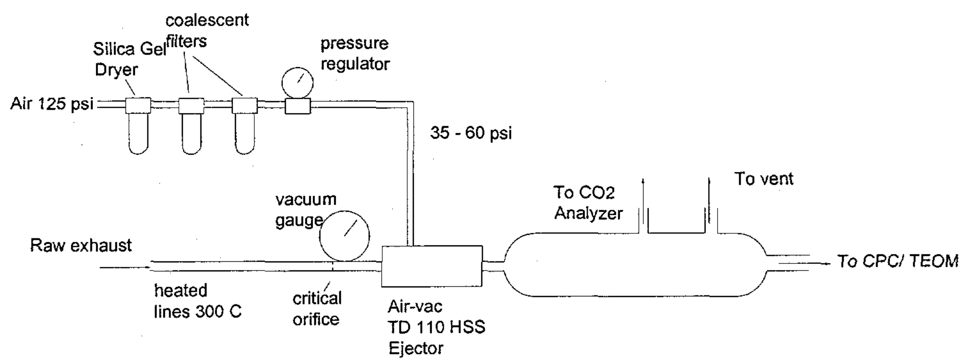

Fig. 2. Schematic of the ejector based dilution system

particulate emissions are key in identifying particulate formation mechanisms, emphasis is also placed in their characterization.

2.0 EXPERIMENTAL SETUP

A fully instrumented 1996 Chrysler Neon engine was coupled to a DC dynamometer. Details of this engine are provided in Table 1. Exhaust from this engine was partially sampled by two similar dilution tunnels, one before the catalytic converter and one after the catalytic converter (*cf.* fig. 1). The diluted exhaust streams were directed either towards a CPC or a TEOM instrument.

Table 1. Engine Specifications

1996 Chrysler Neon	
Engine Type	SI
Injection Type	PFI
Cam Configuration	SOHC
No. cylinders	4
Displacement (L)	2
Other	EGR

2.1 INSTRUMENTATION

Dilution Tunnel: The design of the dilution tunnel used for these measurements resembles the one previously used by researchers at the University of Minnesota (Graskow et al., 1998; Abdul-Khalek et al., 1998). Details of this system are given in fig. 2. It consists of an ejector pump driven by high-pressure (0.24–0.41 MPa) air supply. The air is dried by a silica gel dryer and is further filtered by two coalescing filters in series. These filters are capable of removing up to 95% of particulate matter above 0.3 μm. Although filtration efficiencies for particles below 0.3 μm are unspecified, one could reasonably assume high filtration via diffusion. Using such a system, the number density in the filtered air was measured to be between 400 and 800 (particles/cm^3), which is two orders of magnitude lower than the typical atmospheric concentrations. Further, a vacuum generated by an ejector pump facilitates partial sampling of the exhaust stream. A critical orifice placed in the sampling line meters the mass flow of the exhaust sample. Varying the orifice size and the air-supply pressure varied the dilution ratio, DR. CO_2 concentrations measured in the raw exhaust and in the diluted stream facilitated estimation of the dilution ratio. Kayes and Hochgreb (1998) provide the following relation for estimation of the dilution ratio, DR, which is based on mass flow rates:

$$DR = \left\{ \frac{X_{c,e} \cdot (1 - X_{w,e}) - X_{c,a} \cdot (M_a/M_e)}{X_{c,d} - X_{c,a} \cdot (M_a/M_d)} \right\} \cdot \left(\frac{M_e}{M_d} \right) \quad (1)$$

where,

X_c - dry mole fraction of CO_2;
X_w - mole fraction of water vapor estimated from fuel composition and air fuel ratio;
e,a,d - raw exhaust, air, diluted exhaust; and
M - molecular weight.

Under the assumptions that the ratio of molecular weights = 1, and $X_{c,a} = 0$, the above relation reduces to

$$DR = \left(\frac{X_{c,e}}{X_{c,d}} \right) \cdot \left(1 - X_{w,e}\right). \quad (2)$$

CPC: Model 3022 CPC from TSI, Inc., was used for measuring the particulate number density. In this instrument, particles in the diluted stream act as nucleation sites for supersaturated butanol vapor. The particles quickly grow in size to a few micrometers in diameter and are easily detected by an optical detector. This instrument is capable of measuring number densities up to 10^7 (particles/cm^3) with a time response of 13 s. To keep the measured number concentrations within this range, the dilution ratio was fixed at ~ 20 by using a 0.8-mm orifice and reducing the air supply pressure to 0.41 MPa. Estimates from the measured CO_2 concentrations showed that the DR values were steady under all engine operating conditions, except at wide-open throttle, when a 5% decrease was observed.

TEOM: A tapered element oscillating microbalance (TEOM), model 1100 from Rupprecht and Patashnick company, was used in the measurement of mass concentration, M (g/cm^3). In this instrument, a micro-filter placed at the end of an oscillating element collects particles in the exhaust stream. The mass deposited on the filter is measured as a change in the oscillation frequency of the element. As the rated lower measurement limit of this instrument is 0.2×10^{-9} (g/cm^3), the dilution ratio was increased to ~10 to improve the measurement accuracy. This was achieved by using an orifice 1.2 mm in diameter and an air pressure of 0.24 MPa. Similar to the case above, the dilution ratio was observed to be steady under all conditions, except at wide-open throttle.

3.0 RESULTS AND DISCUSSION

3.1 EXPERIMENTAL MATRIX

To limit the scope of this study, measurements were limited to the most widely used operating conditions. The experimental matrix is provided in Table 2. The engine was "broken in" prior to the tests. Also, on a given day, it was ensured that the engine was warmed up for at least an hour before commencement of measurements. After a step change in the operating condition, a minimum of 15 min was allowed before commencement of measurements. Both the CPC and TEOM measurements were averaged over 10 min.

Table 2. Experimental matrix

	Load (Nm)				
RPM↓	20%	40%	60%	80%	100%
1500	29	58	87	116	149
2000	31	62	94	125	156
2500	31	62	94	124	156

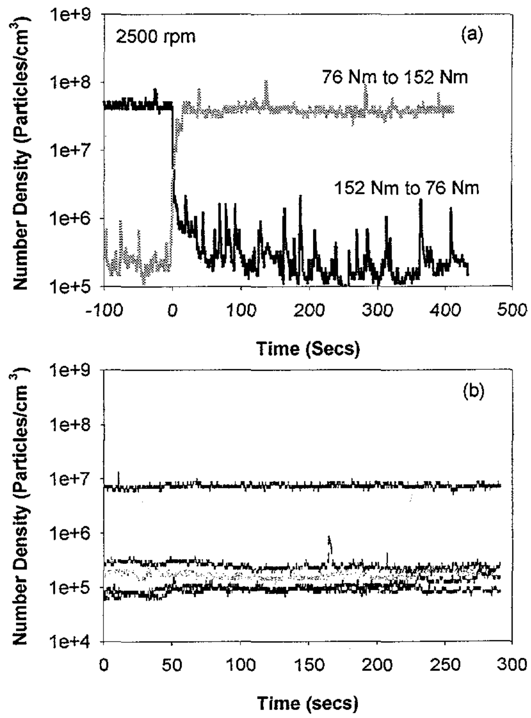

Fig. 3. (a) Transient nature of particulate emissions from an SI engine. Black - Number Density during a step transition from almost full load to half load at 2500 rpm. Grey - Number Density during a step transition from half load to almost full load at 2500 rpm. (b) Number density traces under various steady-state engine-operating conditions.

3.2 RE-EQUILIBRATION TIMES

The measured number density signal for a step change in the engine load is shown in fig. 3 (a). While the re-equilibration time for a step decrease in load is about 200 s, that for a step increase is of the order of 25 s. Considering that the CPC instrument has a response time of 13 s, the re-equilibration times could actually be lower. Further still, they were found to be fairly independent of engine speed and load. To prevent errors due to such transient nature of the signal, settling times in excess of 10 min were allowed for changes between different steady-state conditions of the test matrix.

Previously, Graskow and coworkers (1998) had observed intermittent peaks in particulate number densities, which were found to be at least two orders of magnitude higher than the baseline conditions. In the present set of measurements, intermittent peaks were observed, but with magnitudes just about one order higher than the baseline.

Though their frequency was extremely high following a transition from high load to low load, they were seldom found to occur under steady engine operating conditions (*cf.* 3 [b]). Also, considering that researchers using a different dilution tunnel design (Kayes and Hochgreb, 1998) do not observe such peaks, it is plausible to attribute occurrence of such peaks to the present dilution tunnel design, wherein the exhaust and dilution air streams mix within the channels of an ejector pump.

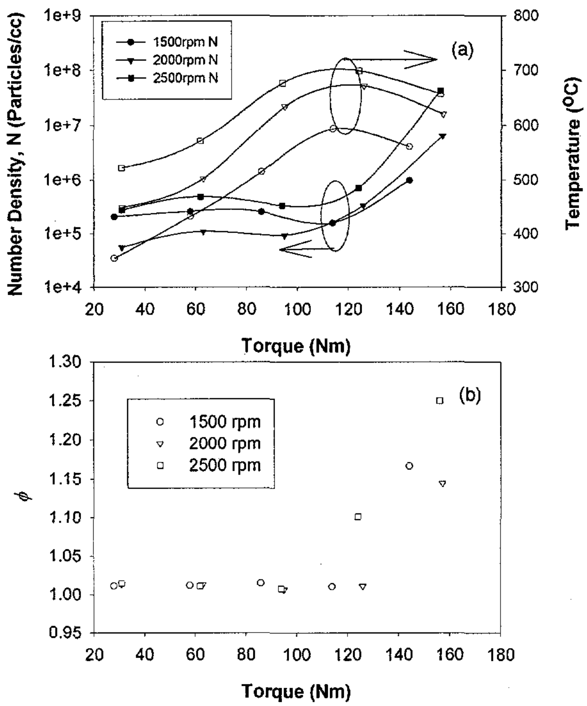

Fig. 4. Engine-out particulate number densities and exhaust temperatures at various engine-operating conditions

3.3 NUMBER DENSITY, N (particles/cm^3)

Particulate number densities at various engine-operating conditions corrected for dilution conditions per eqn. (2) are shown in fig. 4a. Most notably, the number densities are at a minimum for 2000 rpm and tend to increase for higher and lower speeds. Also, there appears to be a general trend with load at all engine speeds: A small peak at low loads, followed by a minimal point at medium loads, which is followed by a steep increase at high loads. While the increased emissions at high loads (torque >110 Nm) can be attributed to soot formation under fuel-rich conditions, i.e., ϕ>1.0, the trends observed at low loads (torque <110 Nm) cannot entirely be attributed to changes in the equivalence ratio. An effect due to EGR, which was functional in the current test engine, further complicates the picture. Readings from a lambda sensor in the engine exhaust as plotted in figs. 4b and 5 show the operation of the engine close to stoichiometry in a majority of the operating conditions.

Considering all of the above factors, one could assume the trend observed above to be the net result of three different mechanisms. The primary contribution mechanism at low loads, the details of which are not clear from the present study, leads to number densities that monotonically increase with load. At medium loads, wherein the engine operates at close to stoichiometric conditions, high exhaust temperatures (*cf.* fig. 4a) lead to enhanced particle oxidation. At higher loads, as the engine operates under fuel-rich conditions (*cf.* fig. 4b), significant contributions from combustion soot lead to high particulate emissions. As shown in fig. 5, stoichiometry tends to be a strong governing factor for particulate number emissions under fuel-rich conditions

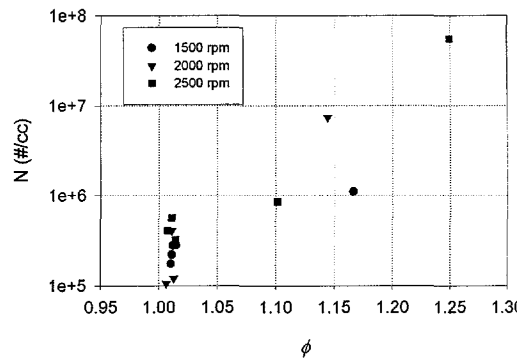

Fig. 5. Engine-out number density as a function of equivalence ratio

As observed in previous studies (Graskow et al., 1998; Shin and Cheng, 1997; Abdul-Khalek et al., 1995), most of the particles appear to be volatile. As a result, these particles are susceptible to oxidation in the exhaust catalyst. As shown in fig. 6, the number densities downstream of the catalyst decreased by an order of magnitude as compared with those at engine-out conditions. However, the extent of oxidation varies with the operating condition.

The brake-specific particulate number, BSPN (no. of particles/kWh), obtained upstream of the catalytic converter is shown in fig. 7. It should be noted that the trend in BSPN resembles closely the number density variation with load (*cf.* fig. 4). Also, this trend is different from that observed by Graskow and coworkers (1998), who have observed BSPN to vary linearly with load. This discrepancy may be due to the different design of the engine used by these researchers.

The brake-specific particulate number, BSPN (no. of particles/kWh), obtained upstream of the catalytic converter is shown in fig. 7. It should be noted that the trend in BSPN resembles closely the number density variation with load (*cf.* fig. 4). Also, this trend is different from that observed by Graskow and coworkers (1998), who have observed BSPN to vary linearly with load. This discrepancy may be due to the different design of the engine used by these researchers.

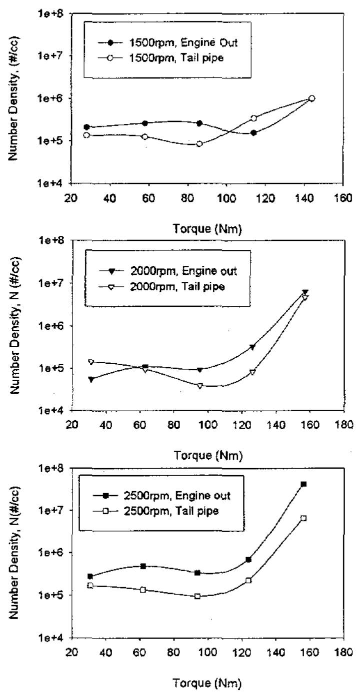

Fig. 6. Engine-out and tailpipe particulate number densities at various engine-operating conditions

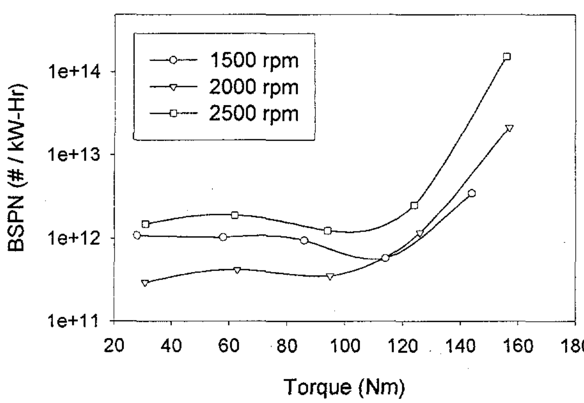

Fig. 7. Engine-out brake-specific particle number (BSPN) for various engine-operating conditions

3.4 MASS CONCENTRATION, M (g/cm^3)

Mass concentration of particulates in the engine exhaust, M (g of particulates/cm^3 of exhaust gases at STP), was measured by using a TEOM instrument. As this instrument relies on changes in the oscillation frequency of a long element carrying a filter, transmission of engine vibration to the instrument under certain engine operating conditions, especially at low loads of 2500 rpm, made the measurements difficult. Therefore, only data corresponding to 1500 and 2000 rpm are reported here. Also, under certain engine-operating conditions, the mass loading of the aerosol stream directed through the TEOM instrument was so low that reliable measurements were not possible.

As shown in fig. 8, the trend of a cusp followed by a steep increase at higher loads (as noticed in number densities) is also manifest in engine-out mass concentrations. An increase in mean particle size with load as noticed by Graskow and coworkers (1998) could account for the pronounced effect of such a trend. As expected, the particulate mass concentrations decreased because of oxidation by the catalyst. However, the extent of oxidation varied with operating conditions.

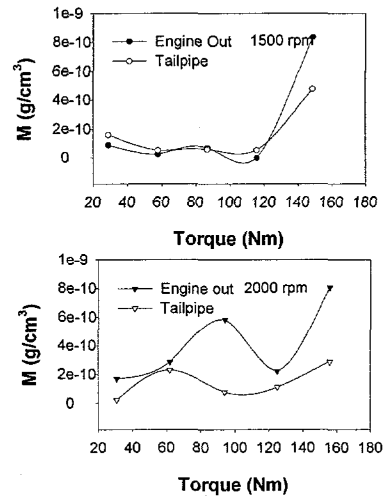

Fig. 8. Engine-out mass concentrations at various engine-operating conditions

Reasonably, one could assume that the cruising condition for a typical vehicle equipped with the current test engine would lie between the two operating conditions shown in table 3. From literature published previously (Graskow et al., 1998), average particulate emissions from a typical light duty diesel is about 1.62×10^{14} (particles/km), which is about three orders of magnitude higher than emissions from the current SI engine. Also, it should be noted that the particulate mass emissions (g/km) from the current SI engine are at least two orders of magnitude lower than the future regulated emission value of 0.0062 (g/km) for light-duty diesel vehicles (Walsh, 1999). Such estimates show that the contribution from SI engines to atmospheric particulate loading is significant only in densely populated areas or when these engines are operated at high loads.

With the probability of retention inside the lungs in excess of 50% for particles smaller than 50 nm (Luders, 1998), significant attention has been focused recently on particulate emissions from SI engines. Such health effects have been further magnified by the fact that most of the polyaromatic hydrocarbons (PAHs) that condense on the surface of these particles are carcinogenic. However, a recent review by Kittleson (1998) identifies the main mechanism of removal for particles smaller than 50 nm in the atmosphere to be coagulation with larger particles. Accordingly, estimated residence times of particles emitted from SI engines are typically 15 min. Considering such short residence times, the health hazard due to particulate emissions from SI engines appears relatively insignificant.

Table 3. Particulate emissions at two representative cruising conditions: 2000 rpm

Power (kW)	BSPN (No. of Particles/ kWh)	(No. of Particles /km)	(g/km)	BSPM (g/kWh)
6.49	7.5×10^{11}	5.03×10^{10}	0.81×10^{-5}	1.2×10^{-4}
13.2	3.6×10^{11}	4.91×10^{10}	1.24×10^{-4}	8.8×10^{-4}

4.0 CONCLUSIONS

Steady-state particulate number densities (no. of particles/cm^3) and mass concentrations (g/cm^3) both upstream and downstream of the catalytic converter in the exhaust of a current technology PFI gasoline engine were measured. Main conclusions drawn through such observations are summarized below.

1) Number densities exhibited a re-equilibration time of ~200 s for a step decrease in engine load and of ~25 s for a step increase in the load. Following an initial transient, the number concentrations were observed to be quite steady, and intermittent peaks reported by previous studies were seldom observed. Also, the magnitudes of these peaks were about an order of magnitude higher than the baseline. These peaks appear to be caveats of the dilution system design rather than true representation of emission characteristics from the current engine.

2) At all engine speeds, a general trend in number densities was observed: a moderate increase at low loads followed by a decrease at mid-range loads, which is followed by a steep increase at high loads. This trend was also reflected in particulate mass concentration, M, and brake-specific particulate number, BSPN.

3) While effects due to exhaust gas recirculation make it difficult to draw conclusions regarding mechanisms leading to particulate formation, one could reasonably assume that the above trend is the net result of three different mechanisms. The primary mechanism at low loads, the details of which are unclear from the present study, leads to number densities that monotonically increase with load. At medium loads, wherein the engine operates close to stoichiometric conditions, high exhaust temperatures lead to particulate oxidation. At high loads, combustion occurs under fuel-rich conditions, and contribution from soot becomes significant.

4) Estimates of the number of particles emitted per mile by a vehicle carrying the current test engine were found to be lower than typical diesel values by three orders of magnitude. Similar estimates for mass emissions (grams of particulates emitted per mile) were found to be at least two orders of magnitude lower than the future regulated emission value of 0.0062 (g/km) for light-duty diesel vehicles. Considering that these particles have typical lifetimes of 15 min (Kittleson, 1998), the health hazard from particulate emissions from SI engines appears to be low.

5.0 ACKNOWLEDGMENTS

We greatly appreciate support from DOE Office of Advanced Automotive Technologies with Ken Howden as the program manager.

6.0 REFERENCES

Abdul-Khalek, I.S.; Kittleson, D.B.; Graskow, B.R.; Wei, Q.; and Brear, F., "Diesel Exhaust Particle Size: Measurement Issues and Trends," *SAE 980525*, 1998.

Abdul-Khalek, I.S., and Kittleson, D.B., "Real Time Measurement of Volatile and Solid Exhaust Particles Using a Catalytic Stripper," *SAE paper 950236*, 1995.

Graskow, B.R.; Kittleson, D.B.; Abdul-Khalek, I.S.; Ahmadi, M.R.; and Morris, J.E., "Characterization of Exhaust Particulate Emissions From a Spark Ignition Engine," *SAE 980528*, 1998.

Kayes, D., and Hochgreb, S., "Investigation of the Dilution Process for Measurement of Particulate Matter from Spark-Ignition Engines," *SAE 982601*, 1998.

Kittleson, D.B., "Engines and Nanoparticles: A Review," *J. Aerosol Sci.*, Vol. 29, No. 5/6, pp. 575–588, 1998.

Luders, H.; Kruger, M.; Stommel, P.; and Luers, B., "The Role of Sampling Conditions in Particle Size Distribution Measurements," SAE paper 981374, 1998.

McAughey, J.J.; Dickens, C.J.; Rickeard, D.J.; Bateman, J.R.; and Kwon, Y.K., "Exhaust Particulate Size Distribution: vehicle and Fuel Influences in Light Duty Vehicles," *SAE paper 961980*, 1996.

Shin, Y., and Cheng, W.K., "Engine-out 'Dry' Particulate Matter Emissions From SI Engines," *SAE 972890*, 1997.

Walsh, M.P., "Global Trends in Diesel Emissions Control – A 1999 Update," *SAE 1999-01-0107*, 1999.