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Research and Development

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Project Summary

Characterization of Emissions and Fuel Economy of In-Use Diesel Automobiles

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Exhaust emissions from twenty 1977-1980 in-use light-duty diesel vehicles were measured to determine the effects of driving cycle, mileage accumulation and test conditions. Hydrocarbons, carbon monoxide, carbon dioxide, nitrogen oxides and particulates were measured from the Federal Test Procedure (FTP), Highway Fuel Economy Test (HFET), Congested Freeway Driving Schedule (CFDS), and New York City Cycle (NYCC), 50 mph cruise (50C), and idle. Individual particulate samples were Soxhlet extracted with dichloromethane to partition the particulate into extract (soluble) and residue (insoluble). The extracts were tested for mutagenicity by the Ames *Salmonella typhimurium/microsome* method. Detailed chemical analysis and subsequent bioassay was performed on selected composite particulate samples.

Emissions (g/mi) and fuel consumption by driving cycle generally increased in the order 50C < HFET < CFDS < FTP < NYCC. Vehicles in the General Motors group generally had higher emissions than the Mercedes-Benz and Volkswagen groups and were more sensitive to driving cycle. The extract showed very little cycle dependence but the residue was very cycle dependent. NO_x emissions decreased with mileage accumulation, while other emissions increased or were unaffected.

Fuel economy was determined by the carbon balance method, by fuel meters, and by fueling records. Over-the-road fuel economy was always lower than carbon-balance fuel economy and was best approximated by the FTP.

A new method for real-time particulate measurements is described using a Tapered Element Oscillating Microbalance (TEOM). The TEOM mass agreed to within 10% of the gravimetric mass on average with a response time of 8-15s.

Short studies were performed on the effects of driving cycle sequence, the dilution tunnel, sub-FTP temperature and mutagenic artifact formation.

Bulk extract samples were fractionated and analyzed by GC, GC/MS and HPLC/UV. The highest specific activity was in the acidic fraction, but most total activity was in the neutral fraction which contained fluorenones and oxy-PAH's.

This Project Summary was developed by EPA's Environmental Sciences Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

The diesel passenger car has only recently become a significant contributor to automobile pollution. From 1975 to 1980 diesels increased from 0.05% to 0.64% of the light-duty fleet in New York State and penetration has been predicted to be as high as 25% by the year 2000.

This study grew out of a need for comprehensive emissions data from *in-use* diesel automobiles as opposed to certification data. Particulate emissions have been of special concern because they are a visible pollutant with a high potential for adverse health effects. Few data were available to assess the effects of non-FTP driving, vehicle aging and real-world use/abuse of light-duty diesel automobiles. The compounds responsible for the mutagenicity in diesel particulate extract have not been adequately characterized and the applicability of current emissions test methods to such areas as mutagenicity testing required further study.

Several short experiments were conducted to investigate special data interpretation situations. The topics investigated were the effects of driving cycle sequence on emissions of the dilution tunnel on emissions and mutagenicity, of filtered exhaust gas on particulate and extract mutagenicity, and of sub-FTP temperature soaking on particulate emissions. These experiments were conducted with a limited number of vehicles and, therefore, the results may not be generally applicable.

Experimental Approach

Table 1 lists the test vehicles by their car number designations which are used in subsequent data presentation. Cars 1 and 5 were purchased new to be used as loan cars for people whose private vehicles were being tested. Cars 2 and 3 were purchased new for use by the New York State Thruway Authority to serve as high mileage accumulation vehicles. Car 10 was also operated by the Thruway Authority. Except for Car 21, the other vehicles were privately owned and maintained. Vehicles were divided into groups according to manufacturer as follows:

GM: Cars 2, 3, 4, 5, 7, and 16
VW: Cars 1, 6, 8, 9, and 11
MB: Cars 12, 13, 14, and 19
Other: Cars 10, 15, 17, 18, and 21. (Car 18 was excluded from the GM group and Car 20 was excluded from all groups.)

The test protocol employed two or three replicate driving cycle sequences to test different fuel/lubricating oil test conditions. Each sequence of driving cycle was called a "Phase." The three phases were:

Phase 1 - vehicle tested as received;
Phase 2 - project control fuel, as received oil;
Phase 3 - project control fuel, fresh oil of manufacturer specifications.

Table 2 shows the phases and driving cycle sequences used.

Hydrocarbons, carbon monoxide, carbon dioxide, nitrogen oxides, and particulates were measured from the Federal Test Procedure (FTP), Highway Fuel Economy Test (HFET), Congested Freeway Driving Schedule (CFDS), New York City Cycle (NYCC), 50 mph cruise (50C), and idle. Fuel economy was measured by the carbon balance method for dynamometer tests and by fuel meters and fueling records for over-the-road tests.

Table 1. *Vehicle Specifications and Dynamometer Test Conditions*

Car #	*Year*	*Make*	*Model*	*Engine Displacement*		*Trans-mission*	*Dynamometer*	
							H.P.	*I.W.*
1	*79*	*VW*	*Rabbit*	*I-4*	*1.5 L*	*M4*	*7.3*	*2250*
2	*79*	*Olds*	*Cutlass Cruiser*	*V-8*	*5.7 L*	*A3*	*12.5*	*4000*
3	*79*	*Olds*	*Cutlass Cruiser*	*V-8*	*5.7 L*	*A3*	*12.5*	*4000*
4	*79*	*Olds*	*98 Regency*	*V-8*	*5.7 L*	*A3*	*12.8*	*4500*
5	*79*	*Olds*	*Cutlass Cruiser*	*V-8*	*5.7 L*	*A3*	*12.5*	*4000*
6	*80*	*VW*	*Rabbit*	*I-4*	*1.5 L*	*M5*	*6.8*	*2250*
7	*79*	*Cadillac*	*Eldorado*	*V-8*	*5.7 L*	*A3*	*10.6*	*4500*
8	*78*	*VW*	*Rabbit*	*I-4*	*1.5 L*	*M4*	*7.3*	*2250*
9	*79*	*VW*	*Rabbit*	*I-4*	*1.5 L*	*M4*	*7.3*	*2250*
10	*78*	*Dodge*	*D-10 Mitsubishi*	*I-6*	*4.0 L*	*A3*	*14.4*	*5500 (+)*
11	*77*	*VW*	*Rabbit*	*I-4*	*1.5 L*	*M4*	*7.3*	*2250*
12	*77*	*M-B*	*240-D*	*I-4*	*2.4 L*	*M4*	*12.3*	*3500*
13	*78*	*M-B*	*300-CD*	*I-5*	*3.0 L*	*A4*	*13.2*	*4000*
14	*79*	*M-B*	*240-D*	*I-4*	*2.4 L*	*M4*	*12.6*	*3500*
15	*79*	*Audi*	*5000*	*I-5*	*2.0 L*	*M5*	*11.8*	*3000*
16	*79*	*Olds*	*Delta 88*	*V-8*	*5.7 L*	*A3*	*13.3*	*4500*
17	*79*	*Peugeot*	*504*	*I-4*	*2.3 L*	*M4*	*10.7*	*3500*
18	*80*	*Olds*	*Cutlass Cruiser*	*V-8*	*5.7 L*	*A3*	*12.6*	*4000*
19	*79*	*M-B*	*300-SD (Turbo)*	*I-5*	*3.0 L*	*A4*	*13.0*	*4000*
20	*78*	*Olds*	*Delta 88*	*V-8*	*5.7 L*	*A3*	*13.3*	*4500*
21	*78*	*Dodge*	*Tradesman 200*	*I-6*	*3.3 L*	*A3*	*12.0*	*4000*

Table 2. *Vehicle Test Driving Cycle Sequences*

A	*B*
Vehicle Tests 1-34	*Vehicle Tests 35-80*
*50C, 30 min**	*50C, 15 min**
50C, 30 min.	
CFDS	
HFET X 3	*HFET X 3*
SOAK, overnight	*SOAK, overnight*
FTP	*FTP*
CFDS	*CFDS*
HFET X 3	*HFET*
IDLE, 30 min	*NYCC*
	50C, 15 min
	IDLE, 15 min
Repeated for each of 3 fuel/ oil combinations= Phase 1, 2, 3	*Repeated for each of 2 fuel/ oil combinations= Phase 1, 3*

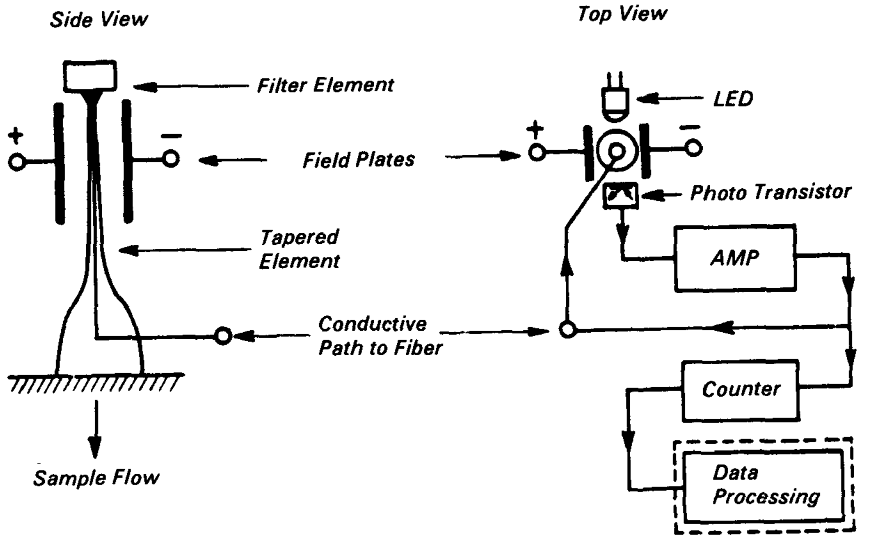
**Pre-Test conditioning, no data taken.*

Particulate samples were collected on 50- x 50-cm Pallflex T60A20 Teflon-coated fiberglass filters. The particulate was Soxhlet extracted with dichloromethane for 24 hours. The resultant extract was dried, weighed and an aliquot tested in dimethylsulfoxide for mutagenicity by the Ames *Salmonella typhimurium*/microsome method. Most work was performed with tester strain TA98 without metabolic activation. Duplicate plates were run at extract doses of 0, 10, 20, 30, 40, 50, 75, 100, and 200 μg. Linear correlation coefficients and their significance levels were computed for bioactivity and extract/residue parameters.

Real-time diesel particulate mass measurements were made for the first time using a Tapered Element Oscillating Microbalance (TEOM). The TEOM is a hollow glass rod, fixed at a wide base, with a removable filter element attached to the narrow top, and oscillating in an electric field (Figure 1). The TEOM has been shown to behave as a harmonic oscillator with a frequency dependent upon the mass collected by the filter element.

Physical, chemical and bioactive characteristics of the particulate were measured to assess the effect of the dilution tunnel on the particulate. The effects of sub-FTP temperature soaks on FTP emission were investigated by cold-soaking a vehicle outdoors during the winter.

Large samples of particulate were collected to provide large amounts of extract (1-27 g) for chemical fractionation and identification by GC, GC/MS and HPLC/UV. The Ames assay was used as a biological monitor to identify important chemical fractions. The extracts were first fractionated into acidic, basic, and neutral compounds by liquid-liquid partitioning. The neutral fraction (about 90-95% by weight) was further divided into seven subfractions by silica gel adsorption chromatography. All subfractions were bioassayed with tester strains



TEOM Operation

1. *Electric field is set up between field plates.*
2. *Image of tapered element is projected on phototransistor.*
3. *Oscillation of element initiated electrically or mechanically produces an AC voltage output from phototransistor.*
4. *AC voltage is amplified and applied to conductive path on element which maintains the oscillation due to interaction with field set up in Step 1.*
5. *Frequency of oscillation and hence mass on filter element is determined by frequency counter.*

Figure 1. *Schematic representation of TEOM instrumentation.*

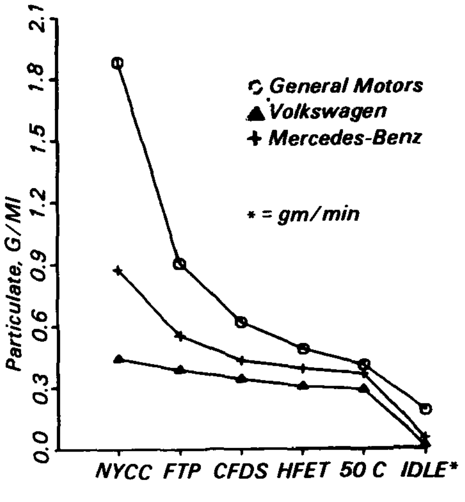


Figure 2. *Cycle variations of particle by vehicle group.*

TA98 and TA100 with and without S9 activation.

Results and Discussion

Gaseous and Particulate Emissions

Table 3 summarizes the mean values of particulate and gaseous emissions and related parameters for the three groups of vehicles for Phase 3. Particulate refers to the material collected by the EPA procedure while residue and extract are respectively the insoluble and soluble fractions of the particulate after extraction.

Figure 2 illustrates the effect of driving cycles on particulate emissions. In general the other emissions (g/mi) followed the same general trends of 50C < HFET < CFDS < FTP < NYCC and VW < MB < GM. The most prominent aspects for all emissions were the very large increase for the NYCC (about double the FTP) and the large decrease for the IDLE relative to the driven cycles. The General Motors group's emissions were greater and much more sensitive to driving cycle than those of the Volkswagen and Mercedes-Benz groups. The cycle variation of the particulate emission rates were principally due to variations in the residue emission rate for all vehicle groups. Residue emission rates were very cycle dependent for the General Motors group but only slightly cycle dependent for the Volkswagen and Mercedes-Benz groups. The extract emission rate had little cycle dependence except for a very large increase for the NYCC for all vehicle groups. Extract as a percent of the particulate showed no cycle variation for the Mercedes-Benz group, few variations for the Volkswagen group, and strong cycle dependence for the General Motors group.

Changes in FTP particulate emissions with mileage accumulation are illustrated in Figure 3 for the General Motors group. Other groups and parameters did not usually show such large changes. Some of the large increases noted here coincided with mechanical repairs to the vehicle's engine. FTP particulate and hydrocarbon emissions did not show a mileage related increase for the Volkswagen and Mercedes Benz group, but showed large increases for several General Motors vehicles due to large increases in extractable material. The General Motors and Volkswagen groups showed increases in FTP carbon monoxide with mileage but the Mercedes-Benz group did not. There was a decrease in FTP nitrogen oxides for the General Motors and Mercedes-Benz groups, but no trend for the Volkswagen group.

Table 3. *Fuel Economy and Particulate and Gaseous Emissions Summary All Cycles - Phase 3*

	FTP			*CFDS*			*HFET*			*50C*			*NYCC*			*IDLE*		
	GM	*VW*	*MB*	*GM*	*VW*	*MB*	*GM*	*VW*	*MB*	*GM*	*VW*	*MB*	*GM*	*VW*	*MB*	*GM*	*VW*	*MB*
Particulate, g/mi	0.89	0.36	0.51	0.62	0.34	0.43	0.49	0.30	0.39	0.41	0.29	0.37	1.88	0.44	0.87	0.188	0.017	0.053
Residue, g/mi	0.65	0.31	0.48	0.39	0.26	0.37	0.29	0.24	0.34	0.21	0.23	0.31	1.33	0.30	0.75	0.145	0.007	0.045
Extract, g/mi	0.25	0.08	0.08	0.22	0.08	0.06	0.20	0.07	0.06	0.20	0.06	0.06	0.55	0.14	0.13	0.042	0.011	0.008
% Extractable	26.3	21.0	13.8	33.1	23.3	15.2	37.7	23.2	14.6	43.2	21.4	15.4	27.9	31.8	15.1	22.5	52.5	15.0
Hydrocarbons, g/mi	0.65	0 29	0.28	0.41	0.26	0.17	0.33	0.20	0.13	0.41	0.21	0.14	1.69	0.49	0.47	0.230	0.058	0.040
Carbon Monoxide, g/mi	1.69	1 11	1.25	1.12	0.92	0.90	0.95	0.77	0.85	0.94	0.83	0.82	4.02	2.06	2.27	0.549	0.186	0.153
Nitrogen Oxides, g/mi	1.71	0 97	1.58	1.38	0.83	1.35	1.36	0.85	1.34	1.30	0.90	1.2	2.82	1.53	2.32	0.160	0.099	0.128
Fuel Economy, mpg	20.0	43.1	24.7	27.4	53.6	31.1	30 1	57.2	32.7	32.2	59.5	34.4	11.1	29.4	17.0	16.3	56.1	34.6

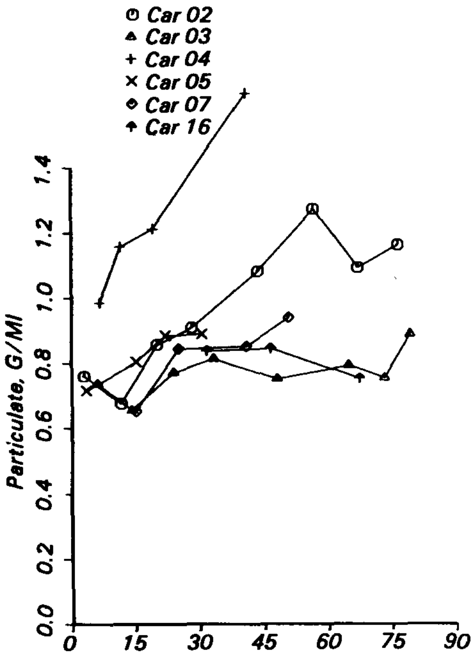


Figure 3. *Mileage accumulation effects for particulate from the General Motors group.*

Fuel Economy

Fuel economy was measured by the carbon balance method and by underhood flow totalizing meters. Figure 4 shows the variations of carbon-balance fuel economy with driving cycle. For each vehicle group, the fuel economy increases as the average speed of the cycle increases. Fuel economy for a given cycle was always highest for the Volkswagen group, followed by the Mercedes-Benz group. Mileage accumulation generally had no effect on the FTP fuel economy.

Over-the-road fuel economy was most closely approximated by the FTP economy even though over-the-road useage was at a higher average speed. At comparable average speeds, over-the-road fuel economy was about 15-20% less than carbon-balance fuel economy. In general, the fuel economy for the CFDS very closely approximated the maximum over-the-road fuel economy.

Bioassay Characterization

The Ames activity of the extract itself generally increased in the order GM< MB < VW for all test cycles and all methods of expressing activity. Figure 5 shows the Ames activity in terms of revertants/μg extract as a function of cycle. The activity generally increased as the average speed of the cycle decreased except that the NYCC generally showed less activity than the other driven cycles.

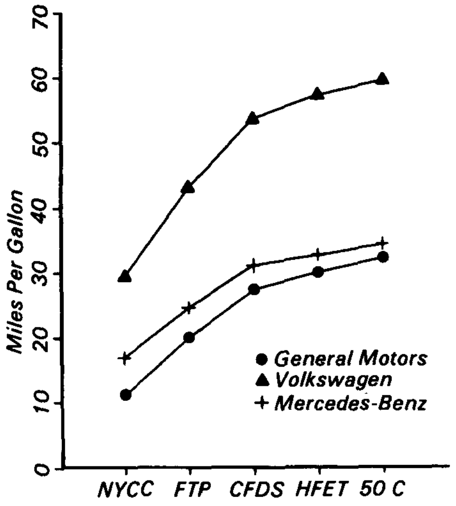


Figure 4. *Cycle variations of fuel economy, miles/gallon, by vehicle group.*

The FTP or the CFDS usually had the highest activity. Some large differences in activity were noted with fuel/lubricating oil changes and with mileage accumulation, but no definite trends could be established.

Linear correlations of activity parameters and exhaust parameters were generally very weak (r values typically ranging from 0.2 to 0.4), although often statistically significant. Some parameter pairs showed a consistency in the sign of the correlation coefficient which was independent of vehicle type and driving cycle. The bioactivity parameter which most frequently yielded a statistically significant correlation coefficient was revertants per μg extract and it was usually correlated negatively with extract and correlated positively with residue.

Real-Time Particulate Measurements

The TEOM (Figure 1), as tested, could respond to dilution tunnel concentrations as low as 1 to 2 mg/m^3 with a response time on the order of 8 to 15 seconds. Cars 1 and 5 were driven over the FTP Bag 3 and NYCC using both standard 47-mm filter collection and the TEOM. On average (29 tests) the TEOM mass was within 10% of the gravimetric determinations. Figure 6 shows a trace of particulate mass emissions rate *vs.* time and the corresponding acceleration *vs.* time trace for the FTP Bag 3. Periods of high particulate emissions corresponded to periods of rapid acceleration while the particulate emissions decreased very rapidly during deceleration. Portions of the curve which seemingly indicate negative particulate emission rates are thought to be caused by the desorption of water held on the particulate and filter.

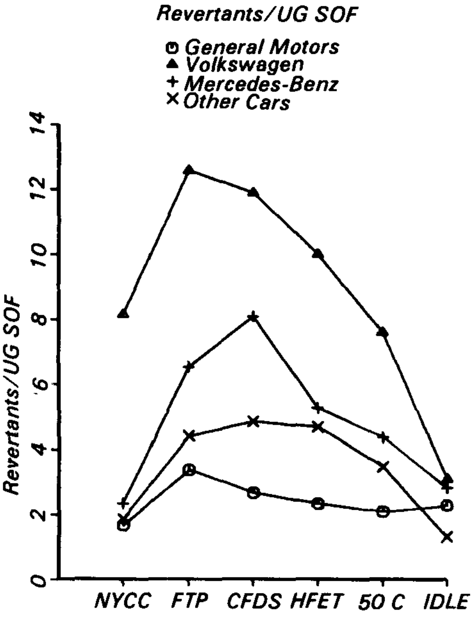


Figure 5. *Cycle variations of specific Ames activity.*

Chemical Characterization of Extracts

The acidic fraction had the highest specific activity, but was a small fraction of the total mass, and therefore, was not the main contributor to the mutagenicity of the extract. The neutral fraction (about 90 to 95% by weight) had a lower specific activity; but due to its mass was the main contributor to the mutagenicity of the extract. The subfraction containing PAH's showed very little activity. Twenty-one PAH's were identified by gas chromatography. The highest specific mutagenic activity was found in the fourth subfraction which contained about 2-4% of the mass of the neutral fraction, but contained 42-52% of the direct-acting mutagenicity of the neutral fraction. GC/MS and HPLC/UV showed this fraction to contain alkylfluorenones with benzo(a)fluorenone the major constituent. The fifth subfraction comprised 4 to 5% (predominantly oxy-PAH's of the mass and 13-20% of the activity of the neutral fraction.

For experiments conducted only with Car 5 the cycle driven prior to the FTP cold soak had no effect on gaseous emissions and little effect on particulate emissions except when that cycle was an idle. Idling prior to a driven cycle always increased particulate, but did not affect gaseous emissions. Previously driven cycles had no effect on gaseous emissions. When a cycle was repeated consecutively during

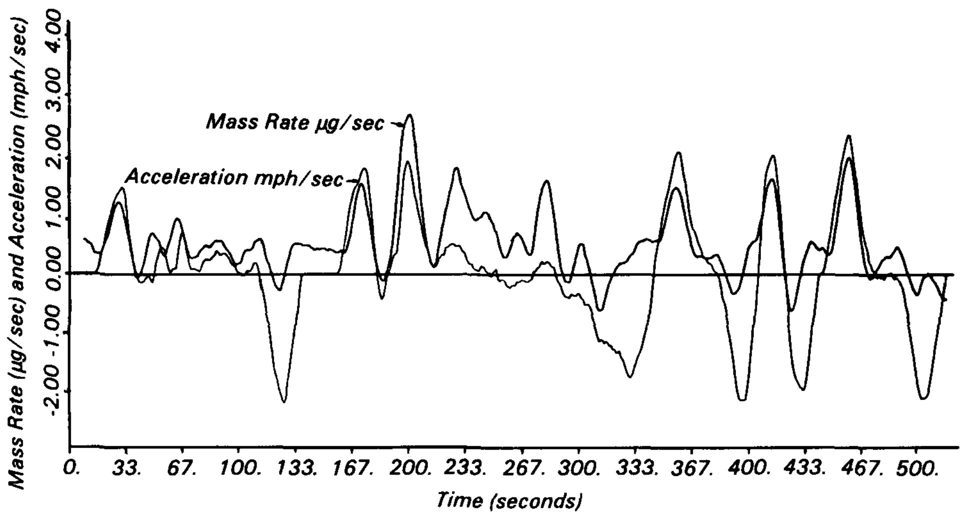


Figure 6. *TEOM mass rate vs. acceleration for Car 5 and Phase 3 of the FTP.*

a day, the fuel economy gradually increased.

The day-to-day variability of particulate and gaseous emission measurements over a period of six or seven test-days usually did not exceed 0.04 g/mi (standard deviation). Multiple repetition of a cycle in a single day had ranges in the order of 0.01 to 0.04 g/mi. The ranges for fuel economy over a six test-day period were 0.5 to 0.8 mpg for Car 5.

The dilution tunnel collected particulate of varying physical, chemical, and biological character. Proceeding from the warmest to the coolest section, particulate matter decreased in bulk density, increased in soluble organic content and decreased in Ames specific activity. The specific activity of the particulate extract from the tunnel walls was higher than that of most of the particulate matter collected with filters.

No effect on specific activity or total mutagenic activity of the extract was observed when particulate or its extract alone were re-exposed to filtered dilute exhaust. The effect of cold ambient temperature on FTP emissions was investigated by cold soaking a vehicle outdoors during the winter. Comparison of FTP data for soaks at 0°C as opposed to 20°C showed an increase in particulate of 18-75%, a decrease in percent extractable of 13-36%, little change in the mass of the extract, and an increase in specific bioactivity of 230-400%.

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***Peter Gabele** is the EPA Project Officer (see below).*

The complete report, entitled "Characterization of Emissions and Fuel Economy of In-Use Diesel Automobiles," (Order No. PB 83-262-071; Cost: $17.50, subject to change) will be available only from:

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