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Effects of Barium-Based Additive on Diesel Exhaust Particulate

By H. William Zeller



UNITED STATES DEPARTMENT OF THE INTERIOR

Report of Investigations 9090

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**UNITED STATES DEPARTMENT OF THE INTERIOR
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UNIT OF MEASURE ABBREVIATIONS USED IN THIS REPORT

atm	atmosphere	lb/ft ³	pound per cubic foot
bhp	brake horsepower	lb/h	pound per hour
°C	degree Celsius	mg	milligram
cm ³ /m ³	cubic centimeter per cubic meter	mg/L	milligram per liter
mm ³ /mg	cubic millimeter per milligram	mg/m ³	milligram per cubic meter
cs	centistoke	mg/min	milligram per minute
°F	degree Fahrenheit	min	minute
ft·lbf	foot pound (force)	mm	millimeter
g	gram	mm ³ /m ³	cubic millimeter per cubic meter
gal	gallon	μm	micrometer
g/L	gram per liter	pct	percent
g/m ³	gram per cubic meter	ppm	part per million
h	hour	psi	pound per square inch
hp	horsepower	psig	pound per square inch, gauge
Hz	hertz	r/min	revolution per minute
in	inch	s	second
in H ₂ O	inch of water	scfm	standard cubic foot per minute
in Hg	inch of mercury	vol pct	volume percent
L	liter	wt pct	weight percent
L/min	liter per minute		

EFFECTS OF BARIUM-BASED ADDITIVE ON DIESEL EXHAUST PARTICULATE

By H. William Zeller¹

ABSTRACT

The Bureau of Mines performed laboratory research to determine the effects of a barium-based fuel additive on diesel particulate emissions. The test engine was typical of types used to power underground coal mining equipment. Test parameters consisted of baseline measurements without additive, three fuel additive concentrations, and five steady-state engine loads, all at 1,200 r/min. Additive effects on soot mass concentration, opacity, particle size distribution, volatile fraction, and NO_x emissions were determined.

Important findings are as follows: Using the manufacturer's recommended additive concentration increased the gravimetrically measured mass of particulate by 30 to 80 pct at four of the five steady-state load conditions. Soot measurements by optical methods did not agree with those by gravimetric techniques, for additive-treated fuels. The additive reduced volatile hydrocarbons adsorbed on filter deposits by up to 50 pct. At most engine loads, carbon particulate was also reduced. About 40 pct of the barium added to the fuel was accounted for in the exhaust.

The health implications for miners were considered, but no firm conclusions were drawn or recommendations made because the results are for steady-state conditions, which may not be representative of real-world operation of diesel-powered equipment underground.

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INTRODUCTION

Diesel-powered equipment is used extensively in underground metal and nonmetal mines, and its use in coal mines is increasing steadily. In many applications, diesels are more productive than electric-powered equipment, but they also have drawbacks. The engines used underground are often adjusted to produce less than maximum power (derated) to reduce emissions of gaseous pollutants (NO, NO₂, CO, CO₂, and SO₂), some of which require dilution with mine ventilation air to meet mandated standards. Other diesel exhaust components, such as particulate matter, aldehydes, and unburned hydrocarbons (HC), are also of concern but have no threshold limit value (TLV).

For many years, exposure to CO emissions from diesel engines was the primary health concern. Currently, the control of NO_x and particulate matter emissions from diesel engines is also considered important. Particulate emissions are important because of their penetration into and retention in the lungs. Ninety-five percent of the particles in diesel exhaust are typically less than 1.0 μm. They are made up of a carbon core surrounded by adsorbed organic compounds. The identification of carcinogens in soot extracts suggests that diesel exhaust particulates could cause or contribute to potentially serious health problems (17, 29).²

An additional problem is the fact that diesel soot contributes to the total airborne-dust load in mines. Consequently, some operators have difficulty meeting the standards for respirable dust, especially in coal mines. Reduction of soot from diesel-powered equipment would enable these operators to more easily comply with dust regulations.

Bureau of Mines diesel research activities were first reported in 1940 (16). The current goal of the program is to reduce occupational hazards associated with diesels by identifying, evaluating, and

improving exhaust control technology to reduce exposures. Additionally, upon request, assistance is provided to mine operators concerning recommendations for the safe use of diesels. The use of fuel additives is one approach some mine operators have tried in efforts to maintain acceptable air quality in sections where diesels are operating.

Each type of fuel additive on the market is designed to perform a particular function. "Pre-flame" additives correct problems that occur prior to burning (i.e., storage stability, flow in cold weather, water contamination) and include dispersants, pour-point depressants, and emulsifiers. "Flame" additives promote complete burning of fuel in the combustion chamber and include atomizers and combustion catalysts. "Post-flame" additives are designed to reduce engine deposits, smoke, and emissions.

The Bureau evaluated Lubrizol 565,³ a post-flame, barium-containing fuel additive, for its effects on diesel particulate emissions in a typical engine used in mining equipment. Its effectiveness as a smoke suppressant for heavy-duty, over-the-road vehicles has been reported by many investigators.

Using a smoke meter, Norman (25) measured large smoke reductions for an engine deliberately overfueled to produce black smoke. Tessier (31), who also used a smoke meter, determined that barium-based additives reduced smoke and asserted that the additive reduced odor. Using gravimetric methods, Turley (33) and Apostolescu (4-5) showed that fuel additives reduced smoke. Using both smoke meters and gravimetric methods, Miller (24) and Golothan (13) measured substantial smoke reductions when treated fuels were used in engines operated at full load.

However, other research studies have reported conflicting results. Truex (32) determined that a barium additive reduced

²Underlined numbers in parentheses refer to items in the list of references preceding the appendixes.

³Reference to specific brands is made for identification only and does not imply endorsement by the Bureau of Mines.

smoke opacity by 30 to 40 pct, but that particulate mass was relatively unaffected. Kittelson (20) tested Lubrizol 565 in a single-cylinder engine and found that smoke, measured with a smoke meter, was reduced by barium-treated fuels, but total particulate emissions, measured gravimetrically, increased. He noted that one effect of the additive was to reduce the particle size of diesel smoke. Because the response of optical sensors per unit mass of soot often decreases with decreasing size of submicrometer particles, the smoke meters underestimated the mass concentration of diesel particulate.

One objective of the current investigation was to confirm the findings by Kittelson but for an engine more representative of types used in mining. Because of the importance of instrument precision and accuracy in Bureau laboratory and field research, a second objective was to evaluate and compare available

instruments for measuring the mass concentration and size distribution of diesel particulate.

The experimental approach followed to accomplish these objectives was to measure the gaseous and particulate emissions from barium-treated and untreated fuels used in a diesel engine operated at different steady-state loads. The particulate emissions were monitored to determine mass concentration and particle size distributions. Limited chemical and physical analyses of the soot samples were performed to determine the major soot components. The data were analyzed to determine the effectiveness of different additive concentrations for reducing soot, to assess the effect of the additive on gaseous emissions, to help explain why certain types of mass concentration instruments furnished unreliable measurements for treated fuels, and to evaluate changes in emissions that might affect the health of miners.

ACKNOWLEDGMENTS

Several personnel of the Bureau's Twin Cities Research Center, Minneapolis, MN, contributed significantly to the research. The author gratefully acknowledges the contributions of Lito Mejia (now with MTS Systems, Inc.) and Kirby Baumgard, mechanical engineers, who were responsible for the design and construction of the laboratory facilities and

who assisted in the conduct of the research. The author also acknowledges the assistance of Carl Anderson, electrical engineer, Harland Kuhlman, engineering technician, and G. Robert Vandenbos, electronic technician, in setting up the laboratory, calibrating and maintaining equipment, and conducting the tests.

APPARATUS AND PROCEDURES

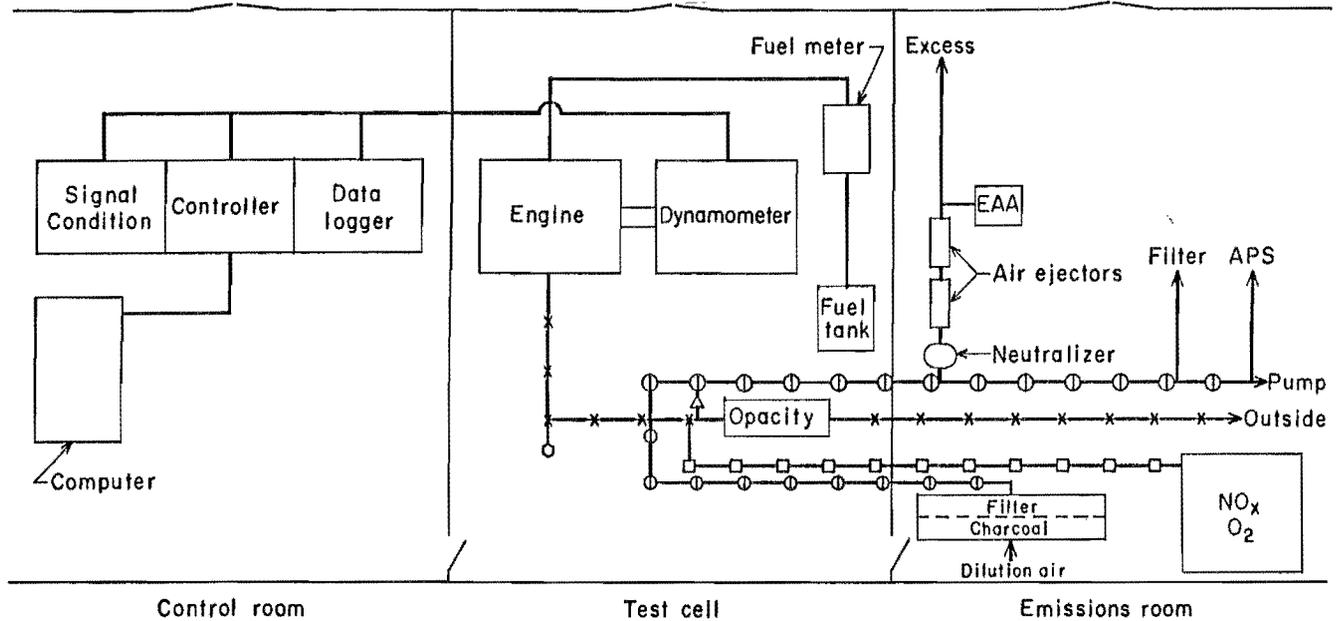
The diesel emissions research laboratory consists of three adjoining rooms: the engine test cell, the control room, and the emissions room. Figure 1 shows the general layout of the laboratory and identifies the major hardware used for this study.

ENGINE CONTROL

The tests were conducted on a Caterpillar 3304 PCNA, four-cylinder, 7-L diesel engine rated for 85 hp at 1,800 r/min; it is a four-cycle, water-cooled, prechamber engine. Engines of this type, which have

been certified by the Mine Health and Safety Administration, U.S. Department of Labor, are used in underground coal mines. After a complete overhaul, the engine was operated in excess of 50 h at various speeds and loads to break-in new components. Baseline tests were conducted to assure compliance with factory-rated horsepower, fuel consumption, and emissions specifications.

Engine loads were applied by an eddy-current, universal dynamometer and were controlled by a microprocessor system which maintains precise speed (± 1 r/min) and load (± 0.1 pct). The engine and



KEY

x—x	Raw exhaust	△	Exhaust sample orifice
⊖—⊖	Dilution air	○	Bosch sample
⊕—⊕	Diluted exhaust	EAA	Electrical aerosol analyzer
□—□	Gas emissions sample	APS	Aerodynamic particle sizer

FIGURE 1.—Diesel engine emissions research laboratory.

dynamometer are mounted on a steel and concrete base that is suspended on six laterally stable coil springs. This spring mass provides 95 pct vibration isolation with a critical frequency less than 400 Hz to reduce resonance with an engine operated at low speeds.

Fuel consumption was monitored by a mass measurement system having an accuracy of ± 0.5 pct. Since not all the fuel feeding an injected engine is used, the portion normally returned to the supply tank is instead returned to a recirculating tank where entrained air, vapor, and combustion gases are removed. This recirculated fuel is then mixed with fresh fuel and returned to the engine.

Air flow into the engine was determined from pressure-drop measurements across the laminar flow element installed in the engine intake line shown in figure 2. Because the pressure drop across laminar flow elements is nearly linear with

volume flow, their accuracy is not affected by engine-induced pulsating flow.

A commercial data acquisition system records up to 64 analog inputs from thermocouples, pressure transducers, gas emissions monitors, and particle-measuring instruments. A built-in calculator chip permits limited data reduction before printing and storage in a microcomputer.

EMISSIONS MEASUREMENT

Gases

Measurements of nitrogen oxides (NO_x) and oxygen were made on undiluted exhaust transported through a line that was heated (350°F) to prevent loss of condensables. The NO_x meter was calibrated using commercial, standard gases; the oxygen meter was calibrated with atmospheric oxygen.

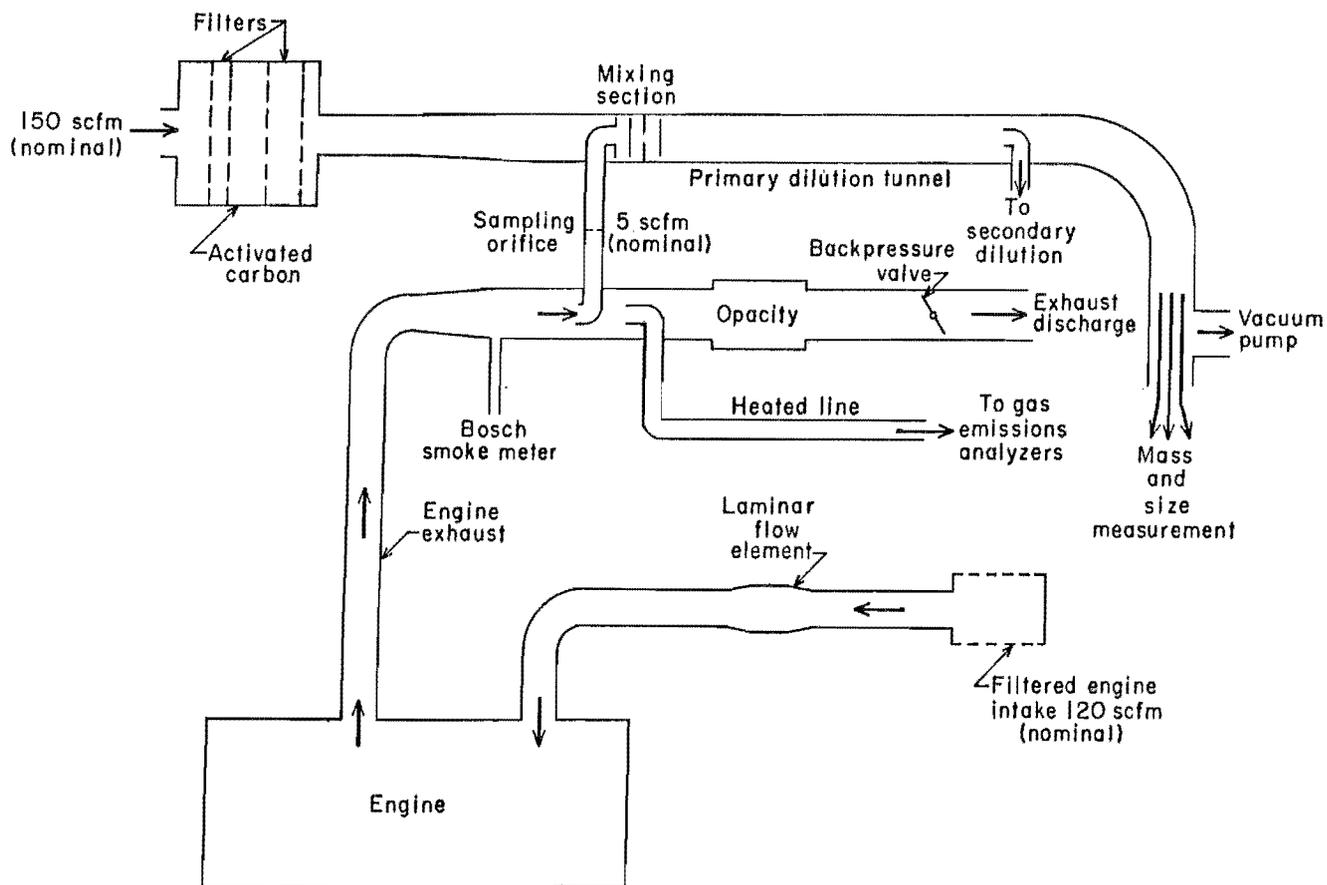


FIGURE 2.—Primary dilution and sampling system.

Particulate

Three different methods of measuring particulate mass concentration--gravimetric, Bosch number (30), and opacity--and one method for measuring particulate size distribution were used in the study.

Mass Concentration

Gravimetric measurements of filter (glass fiber filters coated with Teflon fluorcarbon polymer, 50-mm diam, nominal volume flow rate of 30 L/min) deposits were the reference standard for comparing and evaluating the data from other soot measuring instruments and for assessing additive effects. The filter samples were obtained from the primary dilution tunnel shown in figure 2. The data from

at least two and usually three filters were averaged for each run.

Ten filter samples from the undiluted exhaust were obtained for each steady-state test using a Bosch smoke meter. The Bosch sample line was connected directly to the exhaust pipe (fig. 2) from the engine. No special provisions for dampening engine pulsations were provided. It was discovered part way through the test program that engine pulsations were producing deposits on the Bosch filters even though the Bosch sampling plunger was not activated. In other words, during the period when the Bosch filter was exposed to the engine exhaust, the pressure fluctuations caused by pulsating flow increased the effective volume flow through the filter. For this reason, Bosch numbers reported here may

be too high by as much as a factor of two as discussed further in the analysis section.

The analog signal from the opacity meter (Celesco-Berkley model 107) in 6-in-diam exhaust pipe, located downstream of the Bosch smoke meter (fig. 2), was continuously recorded during the steady-state tests. The recorder traces were averaged to obtain the values used in this report.

Size Distribution

An electrical aerosol analyzer (EAA) was used to measure size and concentration of soot particles between 0.01 and 1.0 μm . Details on the use of the EAA for measuring diesel particulate have been reported by Baumgard (7), Dolan (11), Khatri (19), and Kittelson (20).

An aerodynamic particle sizing system (APS) was used to measure the size and concentration of particulate larger than the approximate 1- μm upper limit of the EAA. Soot particles larger than 10 μm , reported by Miller (24), were not detected by the APS. The most likely sources of large particles are heavily agglomerated soot deposits on engine and exhaust surfaces. Two possible explanations for not detecting these particles are that (1) resuspension of these deposits may have occurred when the APS was not being used, or (2) the APS was used in these tests with special dilution apparatus. The combined losses of the diluter and the APS itself may have seriously limited the potential of the APS to detect large particles.

EMISSIONS SAMPLING AND TRANSPORT

Dilution Systems

The general design and features of the exhaust sampling and dilution systems (figs. 2-3) are similar to those used by Kittelson (20) and Baumgard (7). The primary dilution system (fig. 2) had three design objectives: obtain a representative sample of the engine exhaust particulate, dilute the sample sufficiently to lower the temperature of the

mixture below 125° F, and transport the diluted, uniformly mixed sample to the secondary dilution system and to the filter sampling station with minimum, particle losses in the 0.001- to 1.0- μm range. The main components of the primary system are the tunnel intake (which is fitted with an activated-charcoal filter and an absolute filter), an exhaust sampling and mixing section, a secondary sampling probe, the large-particle and filter sample station, and the positive displacement pump.

The main components of the secondary dilution system (fig. 3) are the intake probe (0.5 in), the krypton-85 charge neutralizer, and the two air-ejector dilution stages. The purpose of the secondary system is to dilute the sample sufficiently to meet the requirements of instruments such as the EAA. The range of dilutions used in the secondary system was between 40:1 and 300:1. Combined primary and secondary dilutions of up to 8000:1 were sometimes required (table C-2).

The entire dilution system was designed and constructed to reduce particle losses. For example, the neutralizer was located near the intake at the primary dilution tunnel to reduce charge-related losses in the ejector dilution stages. Also, volume flow rates in the secondary system are as large as practical to reduce diffusional losses. Nevertheless, losses in the dilution system are likely, but the magnitudes are not known.

Flow Control

Various methods were used to measure or control air flows. All critical flows were calibrated using gasmeters having an accuracy of better than 1 pct. Flat plate orifices were used to monitor the exhaust sample (fig. 2), the diluted sample flow, and both secondary sample flows (fig. 3). The orifices were calibrated in place with gasmeters so that any influence of orifice-to-pipe diameter and pressure tap locations was accounted for. The sample dilution factors produced by the air ejectors were determined using gasmeters.

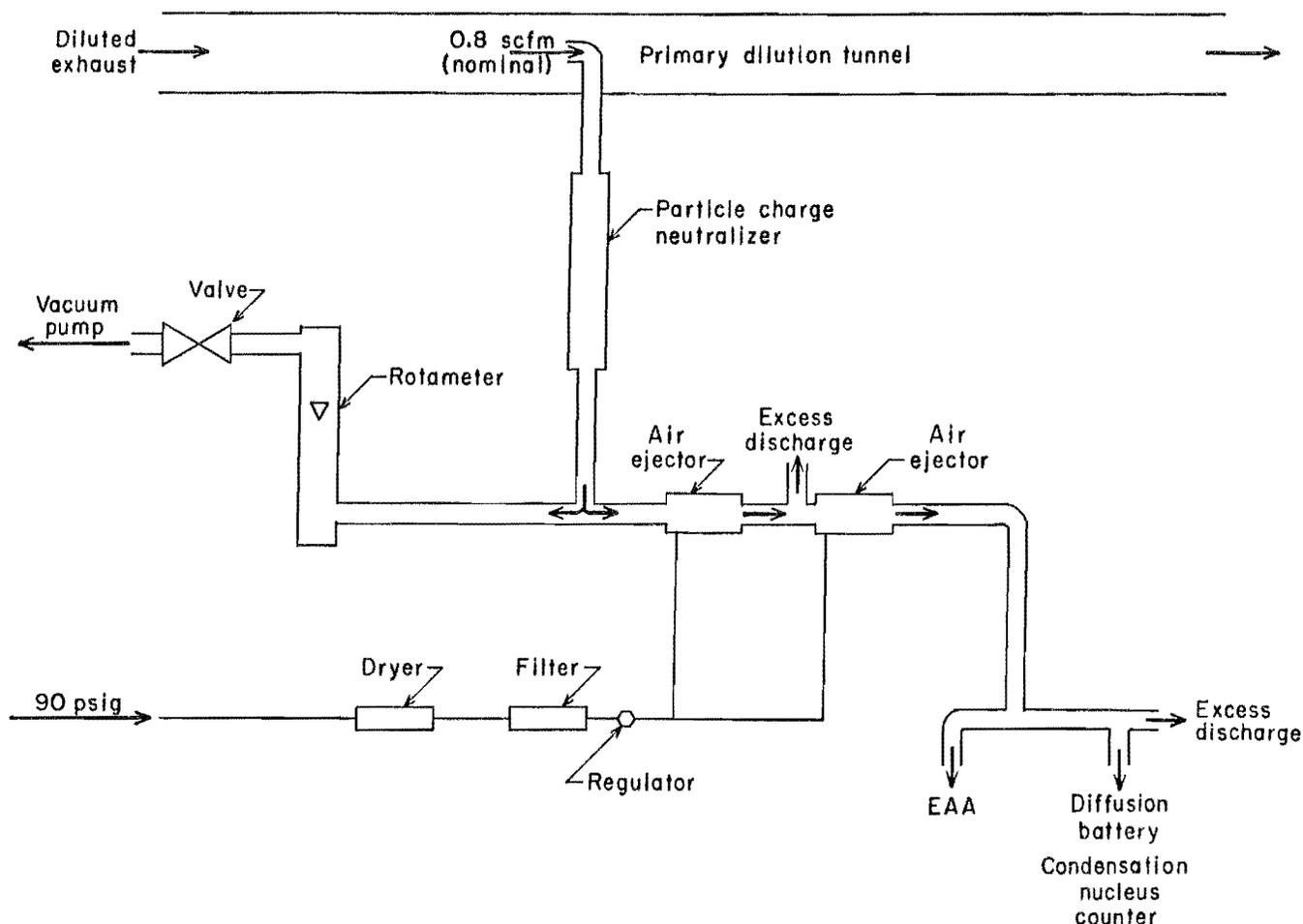


FIGURE 3.—Secondary dilution and sampling system.

The dilution-tunnel, flow control orifice could not be positioned in the preferred location just upstream of the exhaust sample line because the pressure drop produced was excessive for reliable APS operation; therefore, a downstream location was used. Accurate assessment of diluted sample flow at this location required adjustment for all the sample flows extracted upstream of the orifice.

Rotameters were used to monitor the filter flows (fig. 2) and to maintain the flow into the secondary sampling system (fig. 3). Because the rotameters were used in-line and were subject to considerable pressure drop, they were calibrated in place with gasmeters for pressure-drop effects up to 100 in H_2O . Since the manufacturer's pressure correction was found to be exact, it was used

to adjust all filter flow data. Because the pressure drop increased steadily with engine run time, the volume flow rate through the filters for each test was taken as the average of the initial and final flow rates.

PROCEDURES

Addition of Additive

The additive was premixed with the fuel in 55-gal drums prior to testing. The amounts used were 0.18, 0.36, and 0.72 wt pct of additive. According to the manufacturer, the recommended concentration is 0.36 wt pct and between 20 and 25 wt pct of the additive is elemental barium. Fuel and additive specifications are in appendixes A and B, respectively.

Between runs involving different fuel mixtures, the engine fueling system was drained to prevent the problem of fuel used in prior tests from influencing the results of subsequent tests. Any residual fuel in the system after draining was purged by operating the engine for up to 1 h at different loads and speeds with the new fuel mixture.

Test Procedures

A complete summary of engine and environmental parameters for all tests is in table C-1. A brief summary of selected averages is presented in table 1. The specific test modes were chosen to provide as much information as possible regarding the increase of soot particulate with increasing load. Each mode selected was based on only a few preliminary tests. In retrospect, and assuming that only five modes could be tested, better definition of the soot changes would have been obtained if a load between 90 and 100 pct had been selected instead of the 50-pct mode.

Brake mean effective pressure (BMEP) is not physically measurable, but is instead a calculated parameter used to compare the performance characteristics among different engines. It is directly proportional to horsepower, and it is inversely proportional to engine displacement and the number of power strokes per minute. Because all the Bureau tests were conducted on one engine operated at a fixed speed, the BMEP values

in table 1 are directly proportional to horsepower.

The engine was started, brought to normal operating temperature, then operated at full load until engine and exhaust temperatures stabilized. A full test sequence consisted of the five engine loads at one fuel condition. The full load condition of 103.9 psi BMEP was run first, followed by runs at 90.5, 74.4, 49.0, and 7.5 psi BMEP. This order was primarily for convenience because temperatures stabilized more quickly than when tests were conducted starting with the minimum load first. A butterfly valve (fig. 2) in the system exhaust was used to create a backpressure on the engine ranging between about 10 and 30 in of water depending on engine load. It was judged that this was representative of actual operating conditions for this engine when fitted with typical exhaust hardware.

Run lengths varied from about 20 min at full load to as much as 60 min at idle (7.5 psi BMEP). The maximum test interval was determined by the time required to deposit approximately 1 mg of soot on the filters for accurate weighing on a quartz crystal balance. The 10-min minimum interval was needed to provide at least five samples for the EAA to average for the steady-state runs. A complete EAA cycle requires about 2 min. These run times permitted completion of a full test sequence, consisting of the five engine loads at one fuel treatment condition, within an 8-h day.

TABLE 1. - Nominal engine parameters for five test modes at 1,200 r/min

Test mode	BMEP, ¹ psi	Load, pct of full	Power, hp	Torque, ft·lbf	Fuel rate, lb/h	Air-to-fuel ratio
1...	7.5	7	4.6	20	5.72	86
2...	49.0	50	33	145	13.3	37
3...	74.4	75	49	217	19.2	26
4...	90.5	90	60	261	22.2	22
5...	103.9	100	66	290	17.9	18

¹Brake mean effective pressure.

RESULTS AND ANALYSIS

All of the data presented in this section are summarized in appendix tables C-1 through C-5. Between two and six replicate runs were conducted for each test condition. Except where noted, the plotted points are not averages but are individual test results. The emissions concentration data are adjusted to a temperature and pressure of 68° F (20° C) and 1 atm.

ADDITIVE EFFECTS ON PARTICULATE

Gravimetric Measurements

Figure 4 shows the effects of both additive concentration and engine load on gravimetric measurements of soot mass. The lines fitted to the data are drawn through the averages of replicate tests and are the basis for the plots in figure 5 where the percent change (increase or decrease) in soot concentration is as follows:

$$\text{Change} = 100 \times \left(\frac{\text{Treated fuel soot concentration}}{\text{Untreated fuel soot concentration}} - 1 \right). \quad (1)$$

Much of the scatter observed in figure 4, especially at full load, is not random error but is caused by small variations of atmospheric oxygen concentration into the engine. The results of linear regression analysis (fig. 6) show that 90 pct of the variability in measured soot concentration for the untreated fuel is accounted for by the concentration of oxygen in the engine intake air. However, this observed linearity for untreated fuel may be limited to the data range shown and to the particulate component of soot. Ahmad (1), for example, observed exponential increases in hydrocarbons with decreasing oxygen into the engine.

The general trend of the treated-fuel data suggests that the effect of the additive on reducing soot is diminished as oxygen concentration increases. The single trend line drawn through the treated fuel data intersects with the untreated fuel regression line at an oxygen concentration of about 0.0156 lb/ft³. This concentration might be interpreted as an upper limit beyond which little or no benefit from this additive is expected.

The data for the other engine operating modes were also examined for possible correlations of soot levels with oxygen concentration. Only for untreated fuel at 74.4 and 90.5 psi BMEP were weak trends observed.

The soot level increase (fig. 4) with increasing engine load for both untreated and treated fuel agrees with the findings of other investigators (5, 20, 27). For untreated fuel, the overall range of mass concentrations between about 15 and 300 mg/m³ is consistent with the findings of Baumgard (7) using a similar engine but for different engine speeds (1,400 and 1,800 r/min) and load ranges (37 to 100 pct of full load).

The plots in figure 5 clearly show that particulate emissions increase with increasing additive concentration except at full load and at 90.5 psi BMEP and 0.18 wt pct. This result agrees with the findings by Kittelson (20), who determined that, at low to medium (0 - 60 psi BMEP) engine loads, barium-based additives increased mass emissions and only at engine operation approaching full load were emissions lowered for treated fuel compared with untreated fuel.

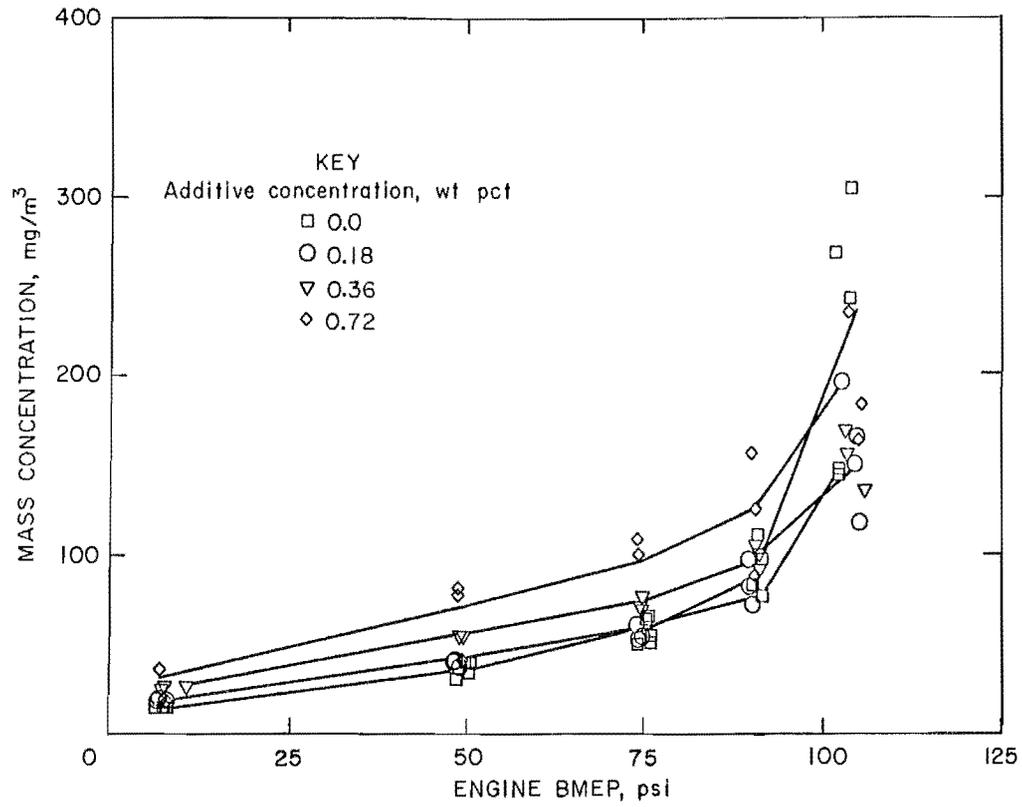


FIGURE 4.—Engine load and fuel additive effects on soot mass concentration.

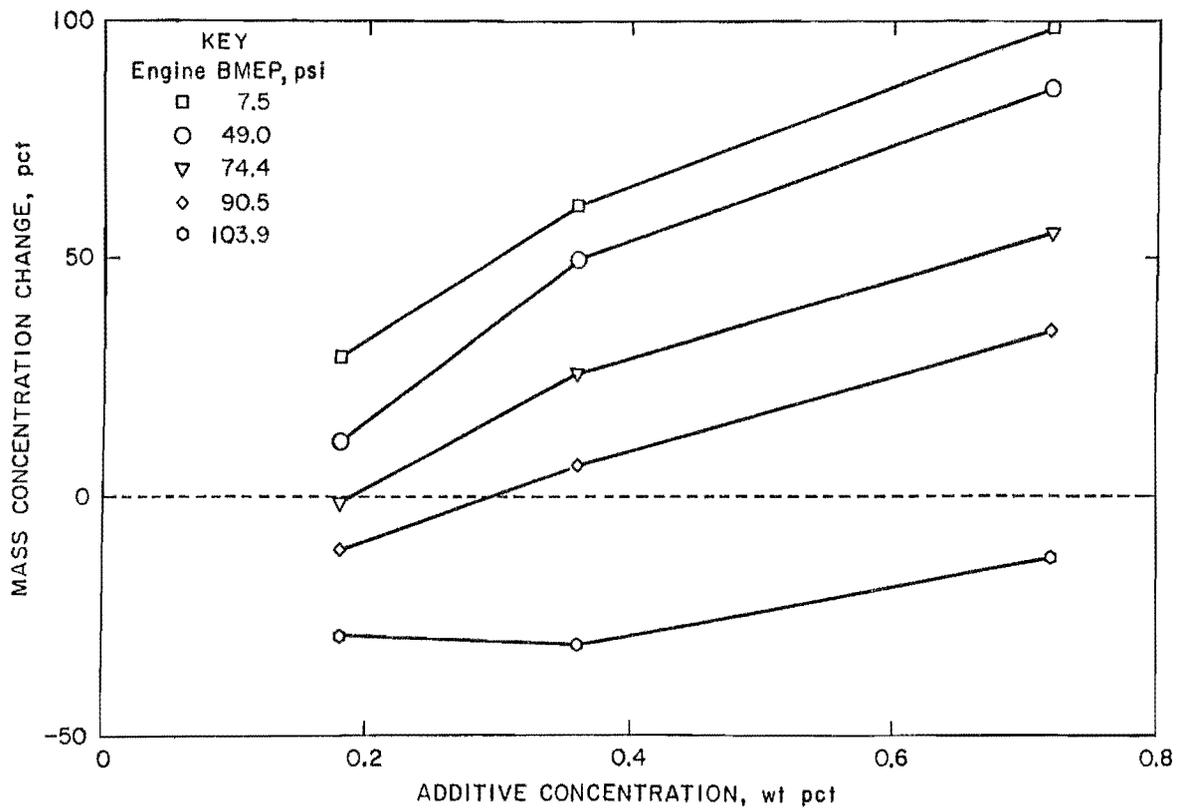


FIGURE 5.—Change in soot levels for different additive concentrations and engine loads.

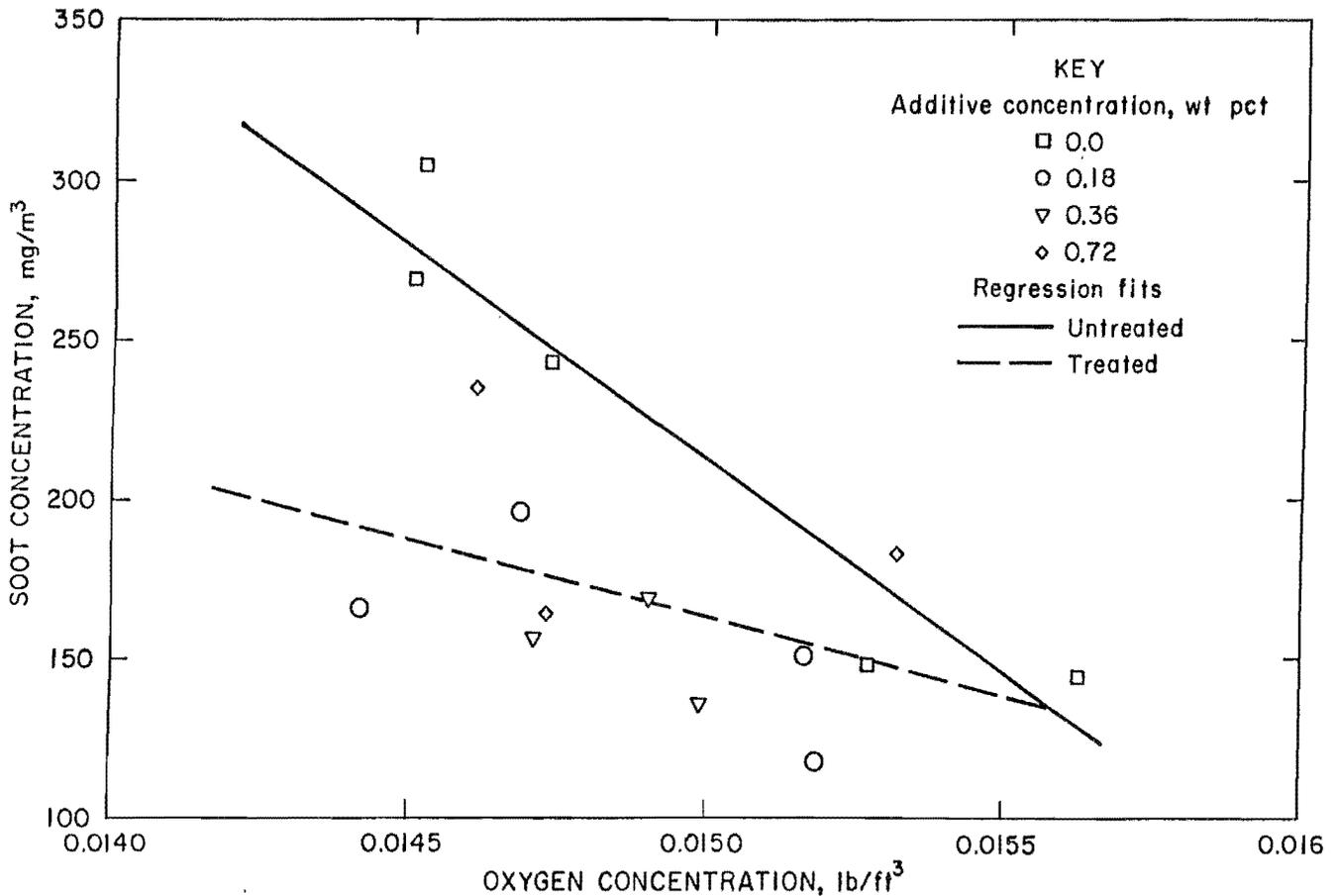


FIGURE 6.—Dependence of soot levels on oxygen concentration into engine at full load.

These results appear to conflict with gravimetric measurements by Miller (24) and Apostolescu (4-5), both of whom found that particulates were reduced with barium-treated fuels. However, Miller's results are for full load only where there is no disagreement with our findings in figure 5. Apostolescu (5) found reduced soot levels for treated fuels over a range of engine loads. A possible explanation is based on his use of a membrane-type filter of 0.8- μ m pore diameter. Tests by Liu (22) on similar filters indicate that collection efficiency decreases with decreasing particle size to a minimum of about 50 pct in the 0.05- to 0.15- μ m-diam range, depending on face velocity. Our findings and those by Kittelson (20) show that mean soot sizes for treated fuels decrease substantially from those for untreated fuels at all engine loads, a result that could account for the apparent soot reduction observed by Apostolescu for treated fuels.

Optical Methods

Bosch Number

Despite a probable bias in the Bosch data (discussed in the Apparatus and Procedures section), the results are included here in order to estimate the magnitude of the bias and to make comparisons with other measurements in this study and with the results of others.

Figure 7 shows that the Bosch number increases with increasing engine load for all treated-fuel conditions. The lines fitted to the data are drawn through the averages of replicate tests and are the basis for figure 8, which shows that treating the fuel with the barium additive reduced the measured Bosch numbers at most of the engine test conditions. Furthermore, the manufacturer's recommended concentration of 0.36 wt pct was optimum; both lower and higher additive concentrations generally produced larger Bosch numbers.

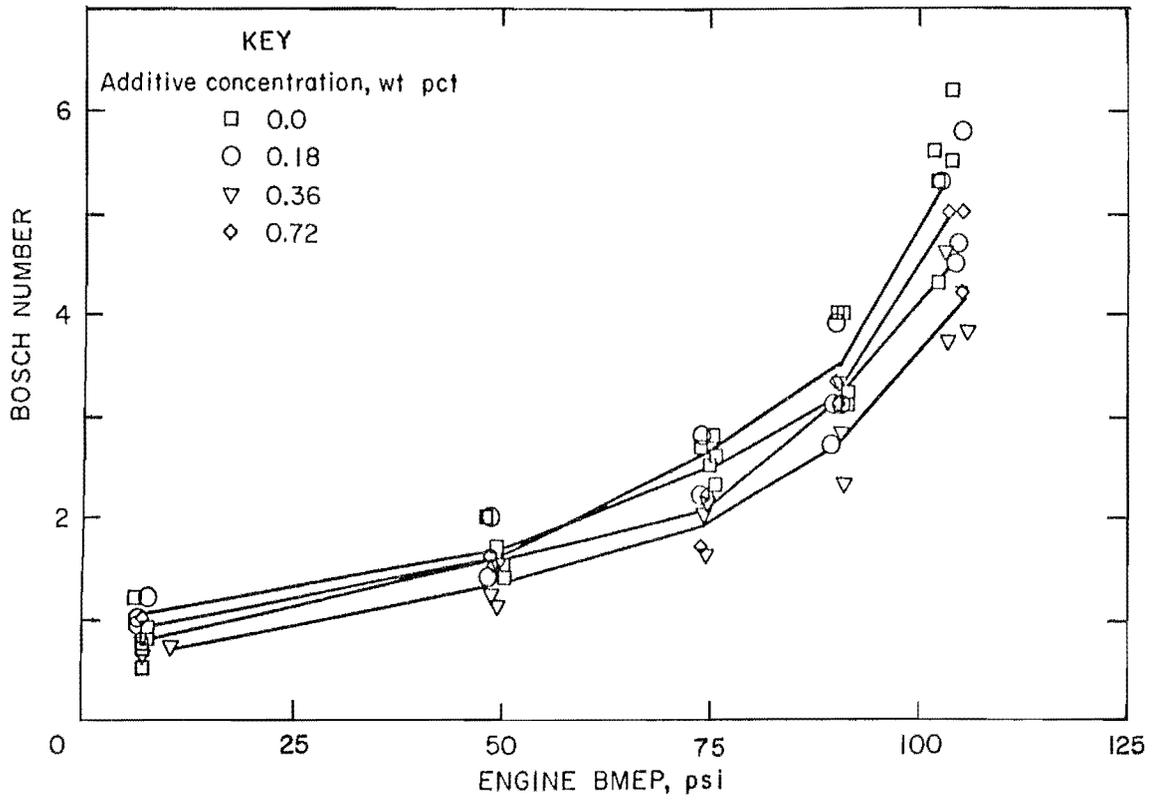


FIGURE 7.—Engine load and fuel additive effects on Bosch number.

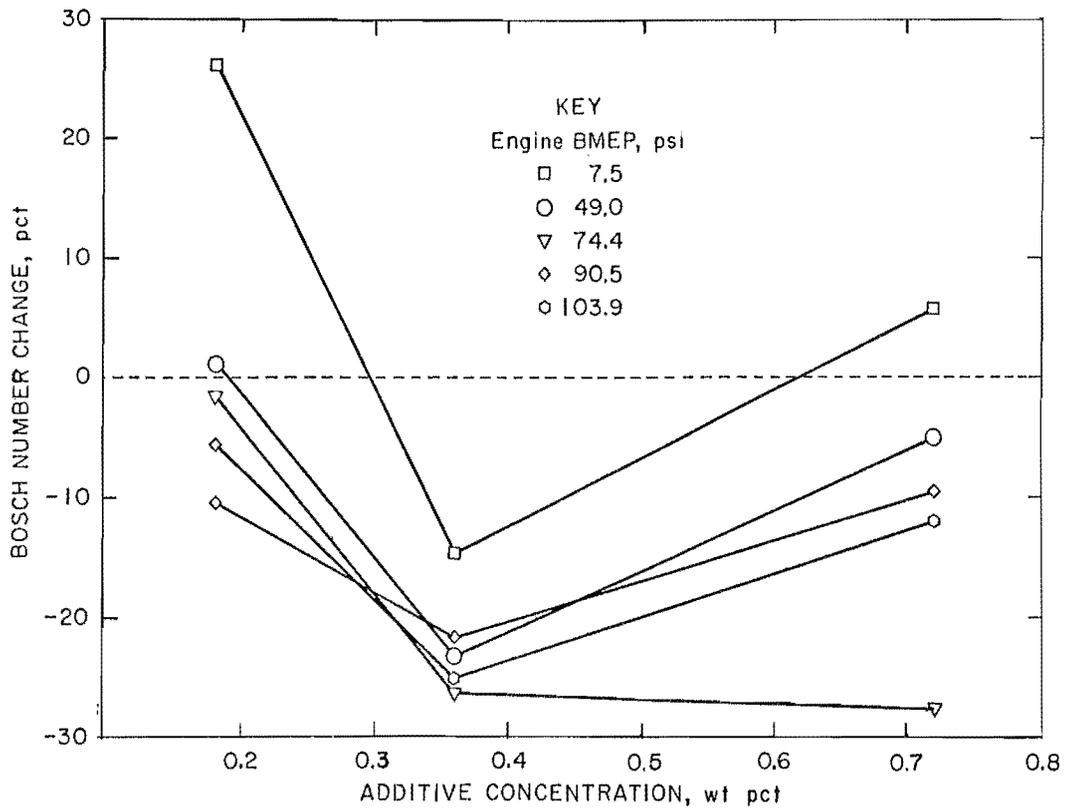


FIGURE 8.—Change in Bosch number for different additive concentrations.

An estimate of the bias in these results is based on the work of Alkidas (2), who determined the following relationship between mass concentration and Bosch number:

$$P = A\{(\ln[10/(10-Bn)])\}^{1.206}, \quad (2)$$

where P is the total soot mass concentration, A is a constant, and Bn is the Bosch number. A least-squares regression fit of equation 2 to our results produced values of the coefficient, A, of 283, 232, 356, and 359 for additive concentrations of 0, 0.18, 0.36, and 0.72 wt pct, respectively. For untreated fuel, Alkidas recommends an average value of 565 for the coefficient, A, which is twice the value of 283 we measured in this study. This comparison indicates that our measured Bosch numbers are too large because the effective volume sampled was increased by exhaust pressure pulsations.

A review of results by others indicates that the effects of barium-based additives on Bosch number are inconsistent and may be both engine and fuel dependent. For treated fuel in a single cylinder engine, Kittelson (20) measured a nearly constant Bosch number (approximately 0.5) independent of engine load (between 15 and 65 psi BMEP) and additive concentration. Hare and Springer (15) conducted tests on two engines and three barium-treated fuels. The results from one engine and fuel combination were similar to those by Kittelson (20) because the Bosch numbers were independent of engine load. The measured Bosch numbers from another engine increased with increasing engine load. Saito (27), who tested only at full load, found that Bosch number decreased with increasing concentration of barium in the fuel up to about 1.7 g/L of fuel, which is approximately equivalent to the minimum additive concentration of 0.18 wt pct used in the Bureau tests.

Opacity

The opacity meter measurements are plotted in figure 9 for all fuel treatment and engine load conditions. Regression analysis showed that the scatter for

untreated fuel at full load can be explained by the uncontrolled variability of oxygen concentration in the engine intake air. The lines drawn through the averaged replicates are the basis for the plots of opacity changes in figure 10, which shows substantial reductions in opacity, between 30 and 60 pct, for all additive concentration levels and engine loads compared with those for untreated fuel.

These results agree with those by others, such as Miller (24) and Golothan (14), who also found that barium-based fuel additives reduced opacity meter response. On the other hand, these opacity measures of additive effects on soot levels do not agree with results obtained gravimetrically in figures 4 and 5. Similar inconsistencies were reported by Hare and Springer (15) and by Truex (32). Explanations for these contradictory results are suggested in the following sections.

Particle Size

Analysis Methods

All of the particle size distribution results in this section are based on EAA measurements of the number of particles in the size range between 0.01 and 1.0 μm . Bimodal, log-normal size distributions were fit to the EAA data. The calculated size parameters for all of the test data are in table C-4. Following Khatri (19) and Kittelson (20), the small-particle mode (<0.03 μm) and the large particle mode (0.03 to 1 μm) are referred to as the nuclei and accumulation modes, respectively.

Volume Distributions

In figure 11 are examples of log-normal distribution fits to the EAA data at an engine load of 49 psi BMEP. These results were selected because they clearly illustrate the bimodal character of some of the data. The plots also show how increasing the additive concentration shifts the particulate volumes, which are proportional to the areas under the curves, from the accumulation mode into

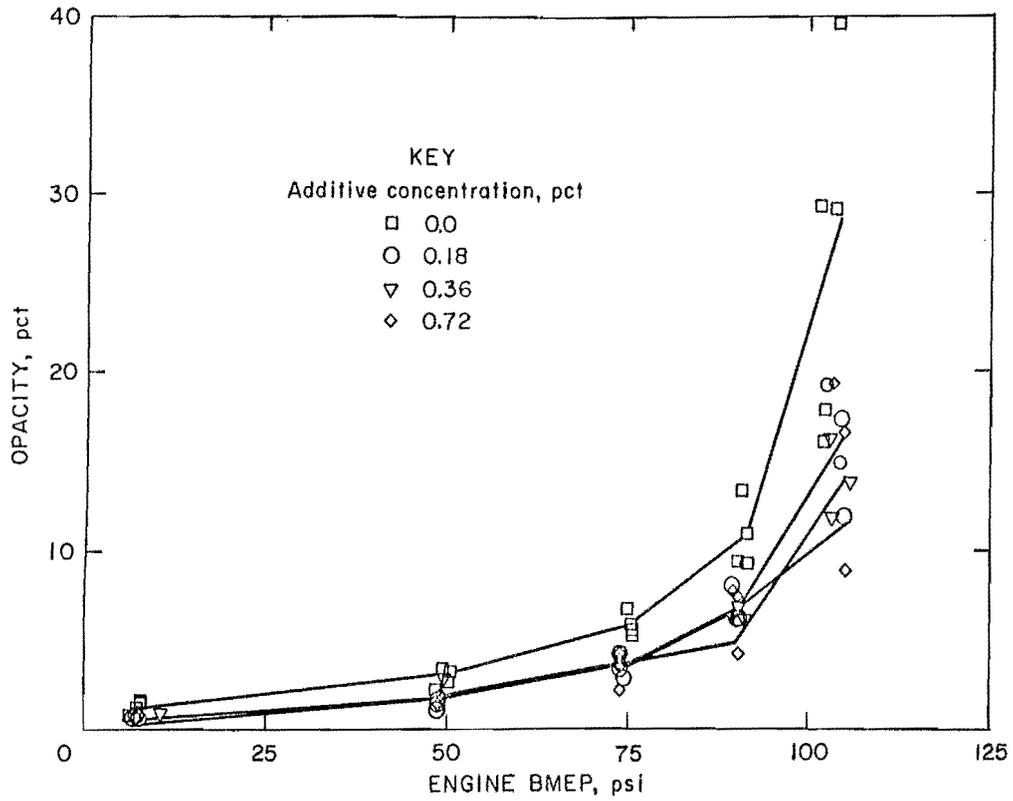


FIGURE 9.—Engine load and fuel additive effects on opacity.

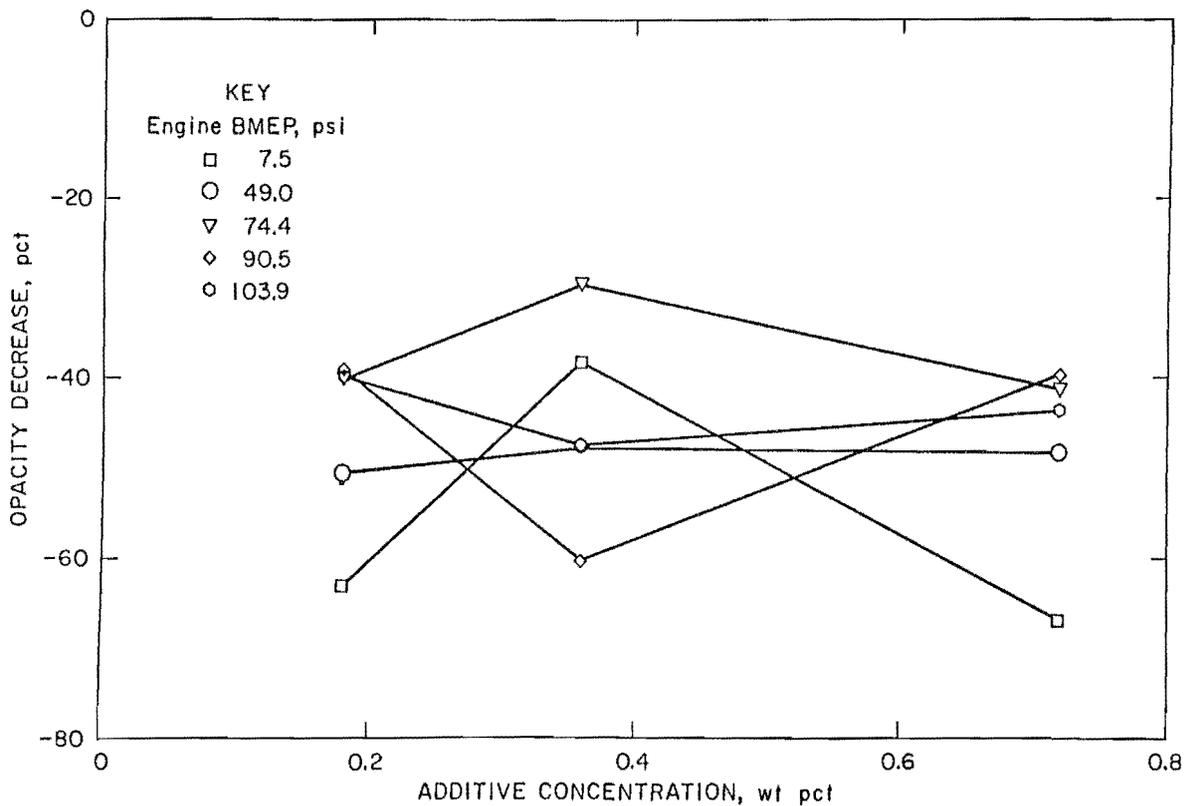


FIGURE 10.—Reduction of opacity for different additive concentrations.

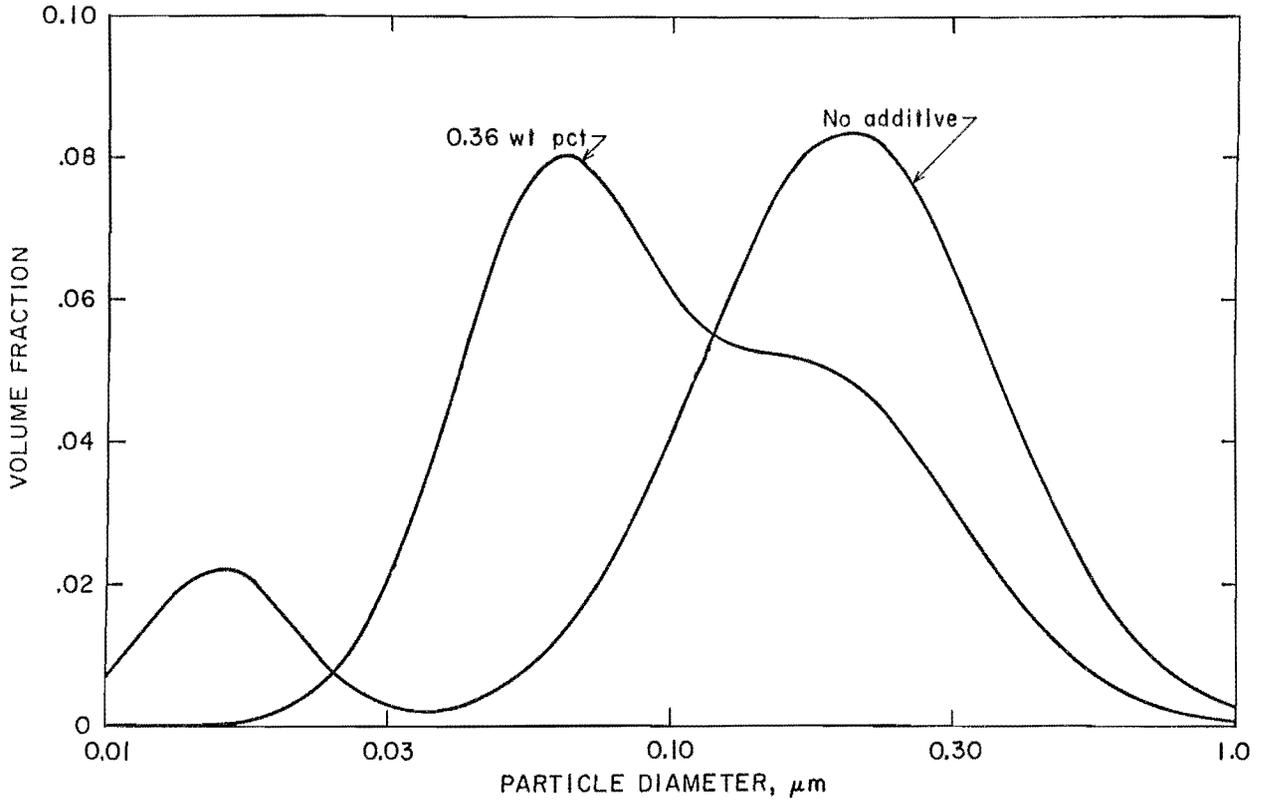


FIGURE 11.—Effect of fuel additive on soot volume distributions.

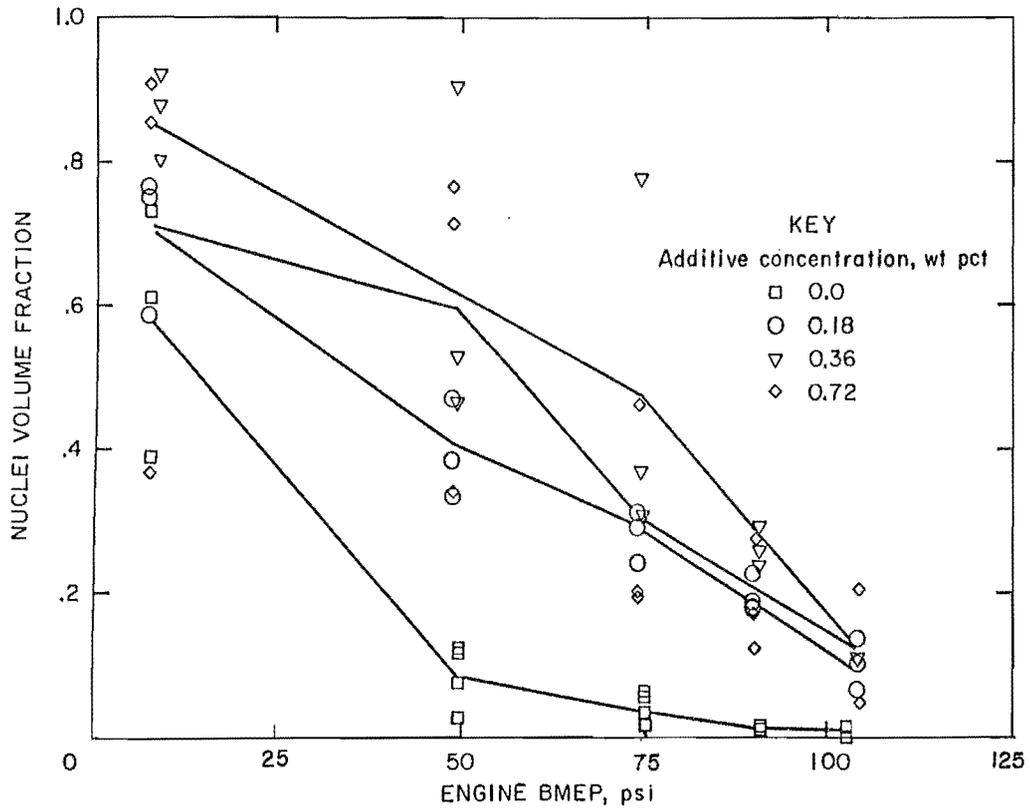


FIGURE 12.—Additive and engine load effects on nuclei volume fraction.

the nuclei mode. In this example (49 psi BMEP, 1,200 r/min), the volume fractions in the nuclei mode are 0.12 and 0.90 at additive concentrations of 0.0 and 0.36 wt pct, respectively.

The particulate volume fractions in the nuclei mode calculated for all the tests are plotted in figure 12. These results show that an additive concentration of 0.36 wt pct produces the maximum shift from the accumulation mode into the nuclei mode. Increasing engine load reduces the nuclei volume fraction for untreated fuel conditions. These results agree generally with the findings by Kittelson (20), for treated and untreated fuels, and by Baumgard (7) for untreated fuel.

Figures 13 and 14 show how the fuel treatments and engine operating conditions affect volume mean diameters within the nuclei and accumulation modes, respectively. The effect of the additive is reversed in the two modes. In the nuclei mode (fig. 13), the additive increases the mean particle diameter from about 0.025 μm to over 0.05 μm except at 7.5 psi BMEP. In the accumulation mode (fig. 14), the additive decreases the volume mean diameters at all load conditions. Except for the relatively large volume mean size at 7.5 psi BMEP, engine operating load has little effect on the volume mean sizes in the nuclei mode. In the accumulation mode, on the other hand, the untreated fuel and the 0.18 pct conditions exhibit similar trends with a maximum volume mean at 74.4 psi BMEP. For the 0.36 and 0.72 wt pct additive concentrations, particle size generally increases with increasing engine load.

Apparent Density

A linear regression fit of EAA volume concentration data to mass concentration of soot, measured gravimetrically, is plotted in figure 15. The values of the regression parameters are summarized in table 2 along with values determined by Zierock (35), who tested two engines at a large number of operating conditions between 1,200 and 4,500 r/min and 15 to 103 psi BMEP. These results confirm that the EAA provides volume measurements that are closely correlated with particulate mass. The differences between sets of regression parameters are probably due to variations in particle properties, mainly particle density, which depends on degree of agglomeration, primary particle density, and fraction of adsorbed hydrocarbons.

Plots of the ratio of particulate mass concentrations to volume concentrations for all test conditions are shown in figure 16. Because this ratio has the units of mass per unit volume, it can be interpreted as a measure of particle density. Kittelson (20) refers to this ratio as an "apparent" particle density and emphasizes the influence that measurement methods may have on the calculated values of the ratios. For example, sampling losses into the EAA are likely but not known. As a result, the calculated volume concentrations in figure 16 are too small in proportion to the lost particle volume in the sampling lines. Therefore, the calculated apparent density is overestimated, and the actual soot bulk density is probably less than unity.

TABLE 2. - Fit parameters for linear regression of EAA volume concentration on mass concentration

Data source	Intercept, ¹ mm ³ /mg	Slope, ² mm ³ /mg	Correlation coefficient
Figure 15.....	-5.3	0.97	0.97
Engine 1 ³	6.4	1.08	.87
Engine 2 ³	-5.5	1.07	.81

¹On EAA volume concentration axis.

²Reciprocal of particle density.

³From reference 35.

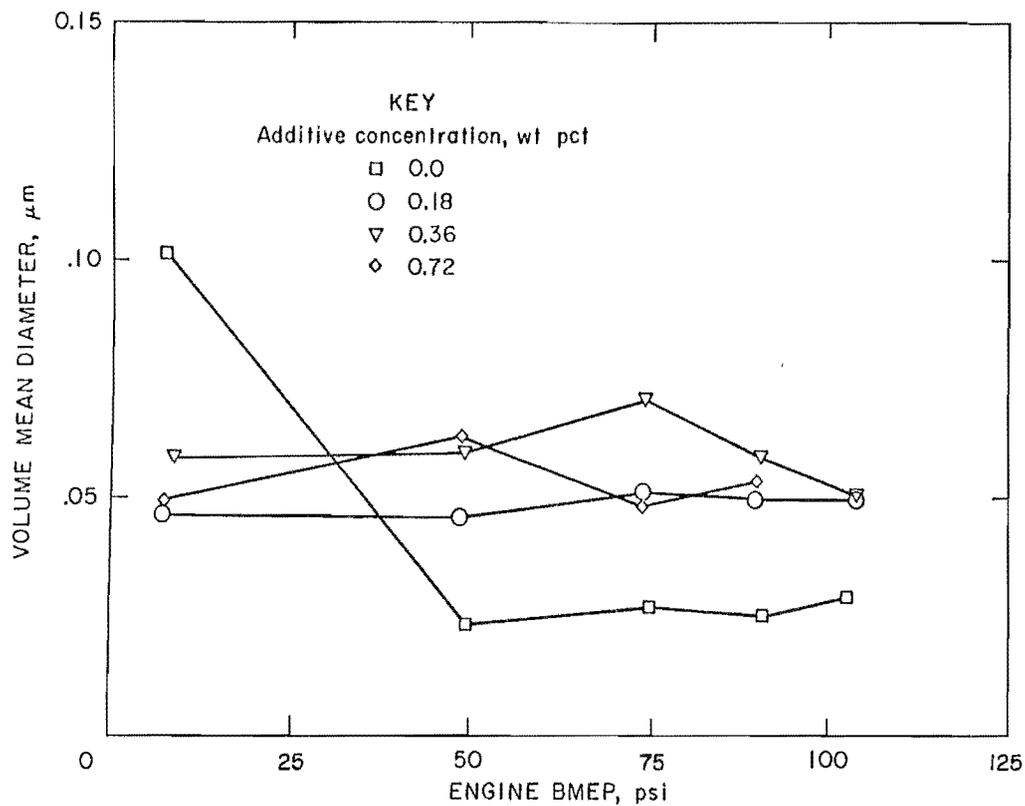


FIGURE 13.—Volume mean diameters in nuclei mode.

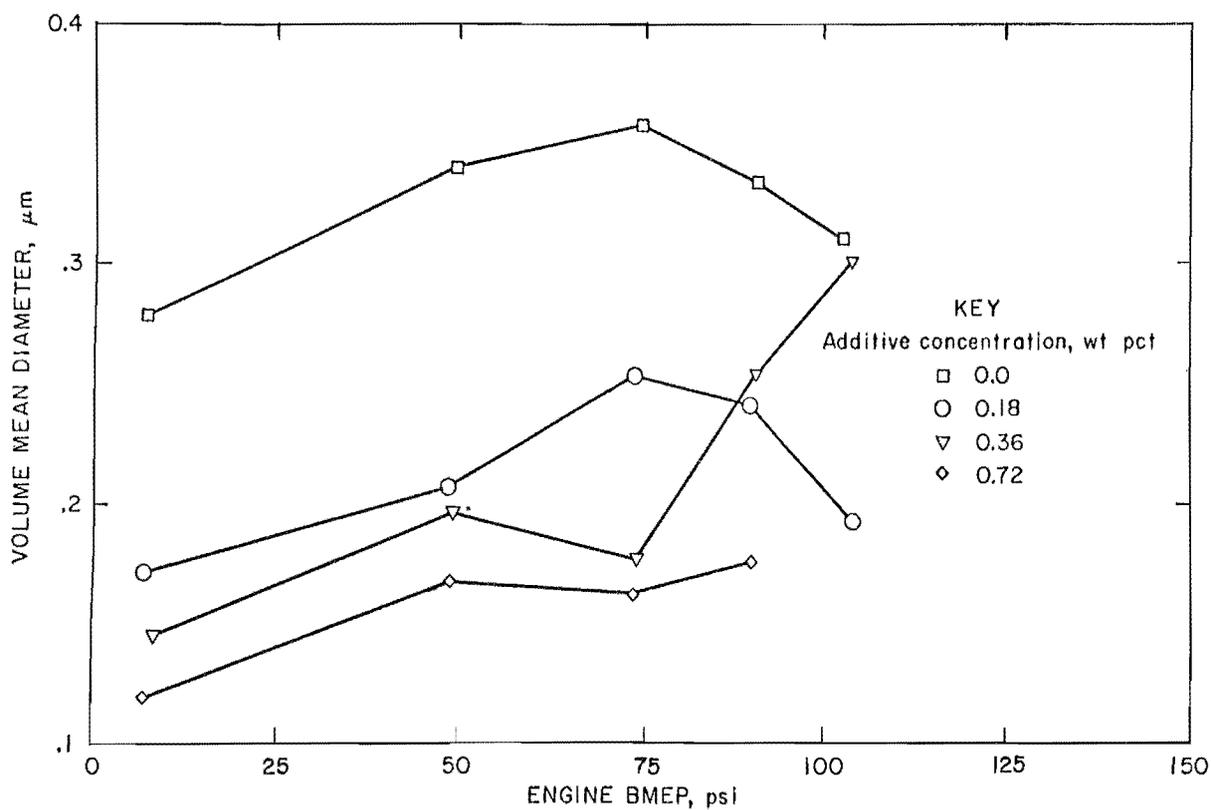


FIGURE 14.—Volume mean diameters in accumulation mode.

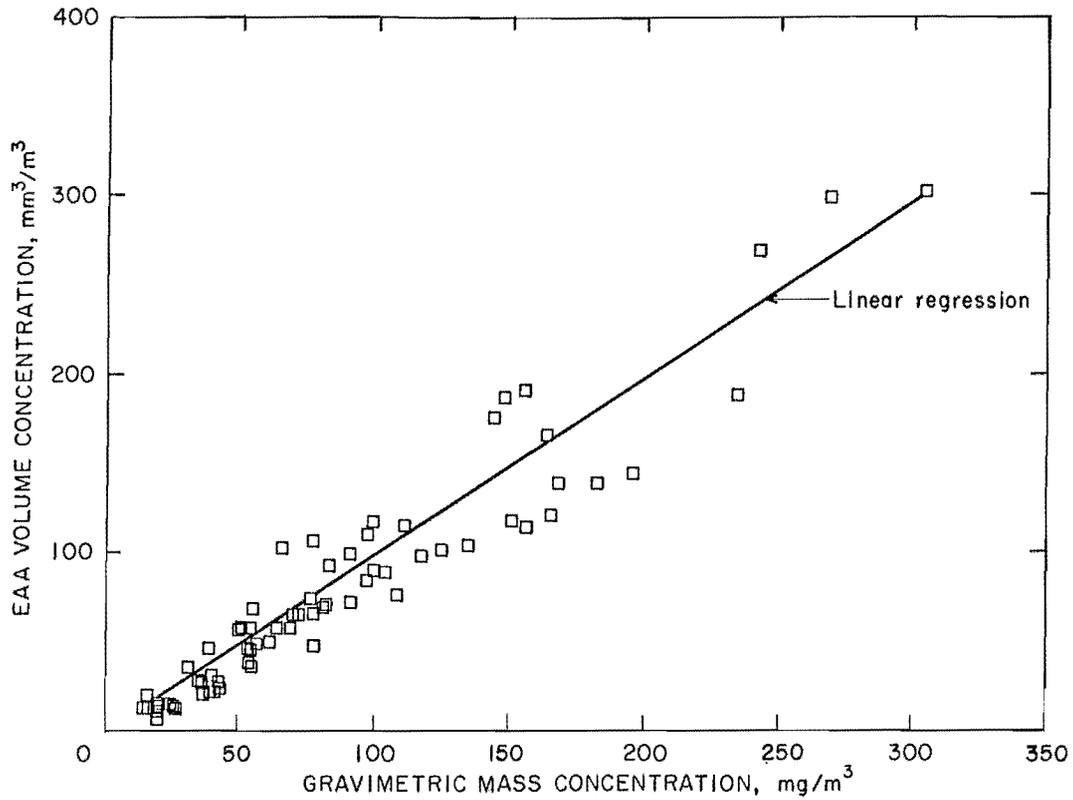


FIGURE 15.—Regression fit of EAA, bimodal volume concentrations to gravimetric mass concentrations.

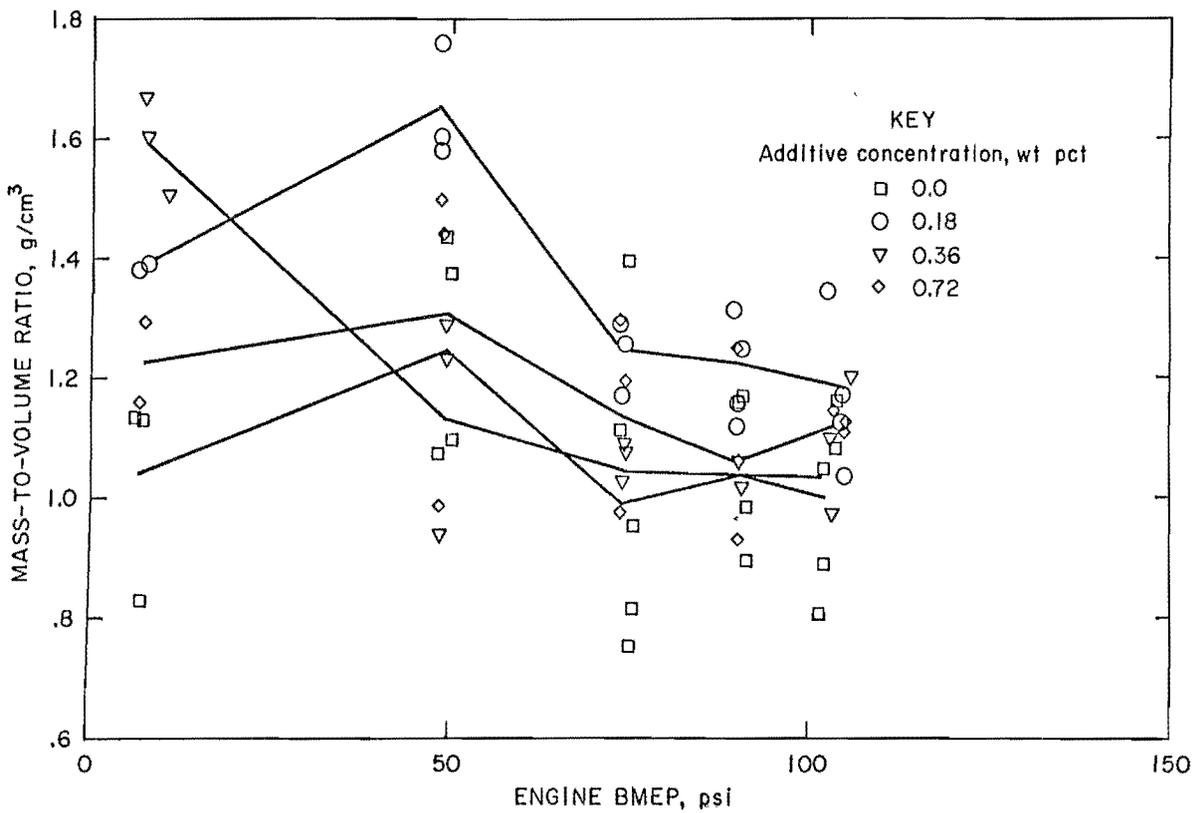


FIGURE 16.—Ratio of gravimetric mass to EAA volume concentration.

The average calculated particle density for untreated fuel data in figure 16 is 1.09 g/m^3 . This result agrees exactly with the value determined by Groblicki (14) for five diesel automobiles. The calculated average for all the additive data is about 1.25 g/m^3 . Kittelson (20) also observed larger apparent densities (up to 2 g/m^3) for treated fuels, a result for which he suggested two explanations: (1) for barium-treated fuels, the less dense carbon fraction in the particles is replaced with higher density barium compounds, and/or (2) the treated fuels produce smaller volume mean diameters in the accumulation mode, suggesting that the particles may consist of more compact agglomerates having higher densities.

Soot Composition

In this section, the volatile hydrocarbon (HC) carbon, and barium compound mass concentrations in the engine exhaust are estimated. The results are used to help account for the barium and are related to instrument response and health effects. Volatile HC and barium data are presented in table C-5.

Volatile Particulate

The same filters used for gravimetric analysis were heated in a 300°C (572°F) oven for about 1 h and reweighed to determine the volatile HC loss. The individual filter data were combined, and the average volatile fractions were determined for each combination of additive concentration and engine load. The mass concentration of particulate HC is the product of the volatile fraction and total soot concentration. The results (fig. 17) show that the average concentration of volatile HC adsorbed on the filters ranged between about 3 and 13 mg/m^3 . The maximum HC levels were produced at 49 psi BMEP or about 50 pct of full load.

The results in figure 17 are not intended to imply that the values represent the gaseous HC concentration in the exhaust. Cuthbertson (10) showed that the quantity of volatile substances adsorbed

on filters depends on numerous variables such as exhaust temperature, dilution volume, dilution rate, sampling time, and filter temperature. Reichel (26) determined that the adsorbed volatiles are a maximum for dilution ratios in the 25:1 to 30:1 range used in the Bureau study. Larger or smaller dilution ratios will produce smaller quantities of volatile substances on the filters.

Bergin (8) suggested a relationship between particulate HC and gaseous HC for untreated fuels, but a correlation for barium-treated fuels is not available. Apostolescu (5) measured no significant effect of fuel additives on the distribution of HC; Miller (24) obtained mixed results: Total unburned HC were unaffected by additives in two cases and were reduced by 30 pct in two other cases.

Barium Recovery

The barium fraction in the exhaust was determined using atomic absorption (AA) analysis on filter deposits. In general, these were not the same filters used for gravimetric and volatility analysis because considerably more sample weight was required for AA analysis. In some cases, not enough sample was obtained so data are not available for every test condition. The averages are plotted in figure 18, which shows that the barium fraction on the filters tends to decrease with increasing additive concentration in the fuel.

The mass rates of barium into and out of the engine were calculated based on the measured fuel consumption rate, the additive concentration in the fuel, and the AA data. Figure 19 shows that, on the average, the barium in the exhaust accounts for only about 40 pct of the barium into the engine. There was no attempt to account for the other 60 pct of the barium, but Miller (24) and Brandes (9) found that much of it is deposited on engine and exhaust system surfaces and in the lubricating oil.

Turley (33), Golothan (13), Miller (24), and Apostolescu (5) determined that most of the barium in the exhaust is insoluble barium sulfate plus small percentages of soluble barium carbonate,

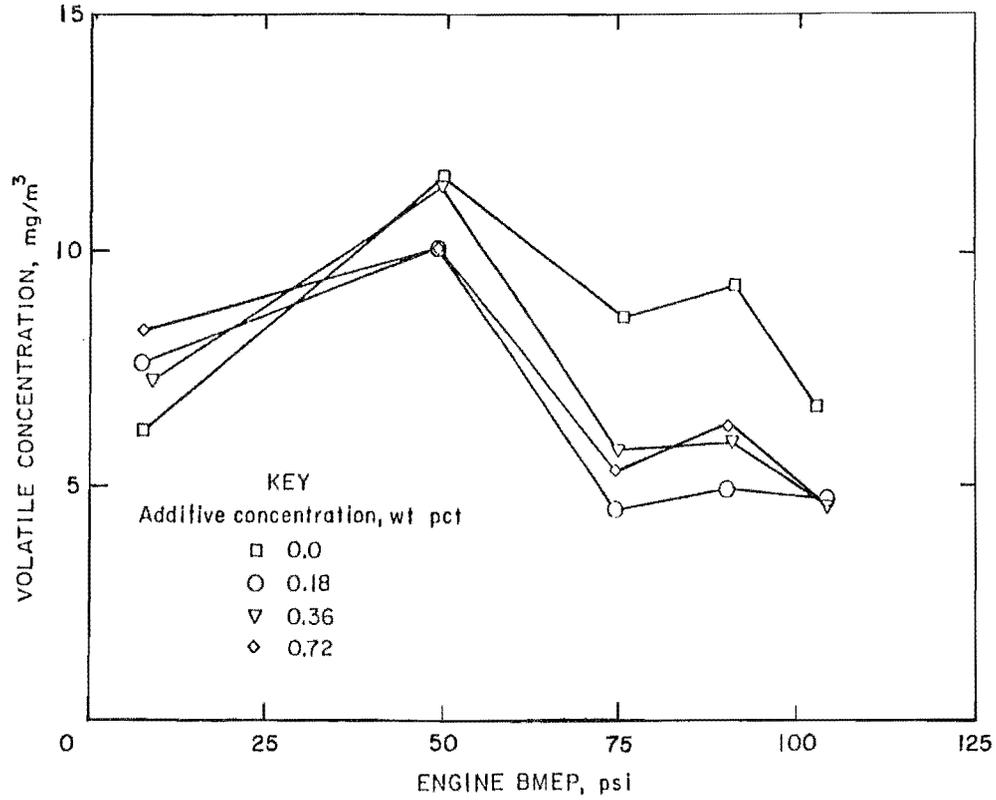


FIGURE 17.—Volatile mass concentration.

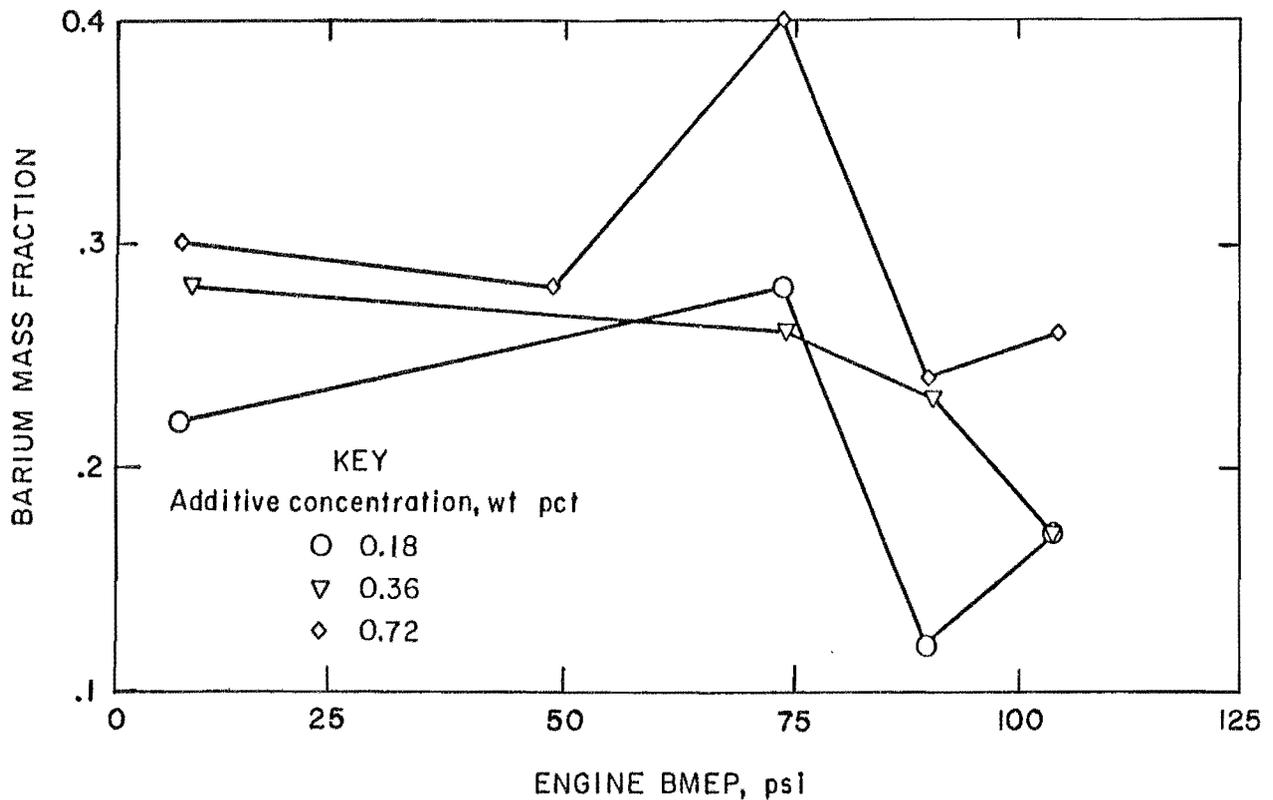


FIGURE 18.—Additive and engine load effects on mass fraction of elemental barium in exhaust.

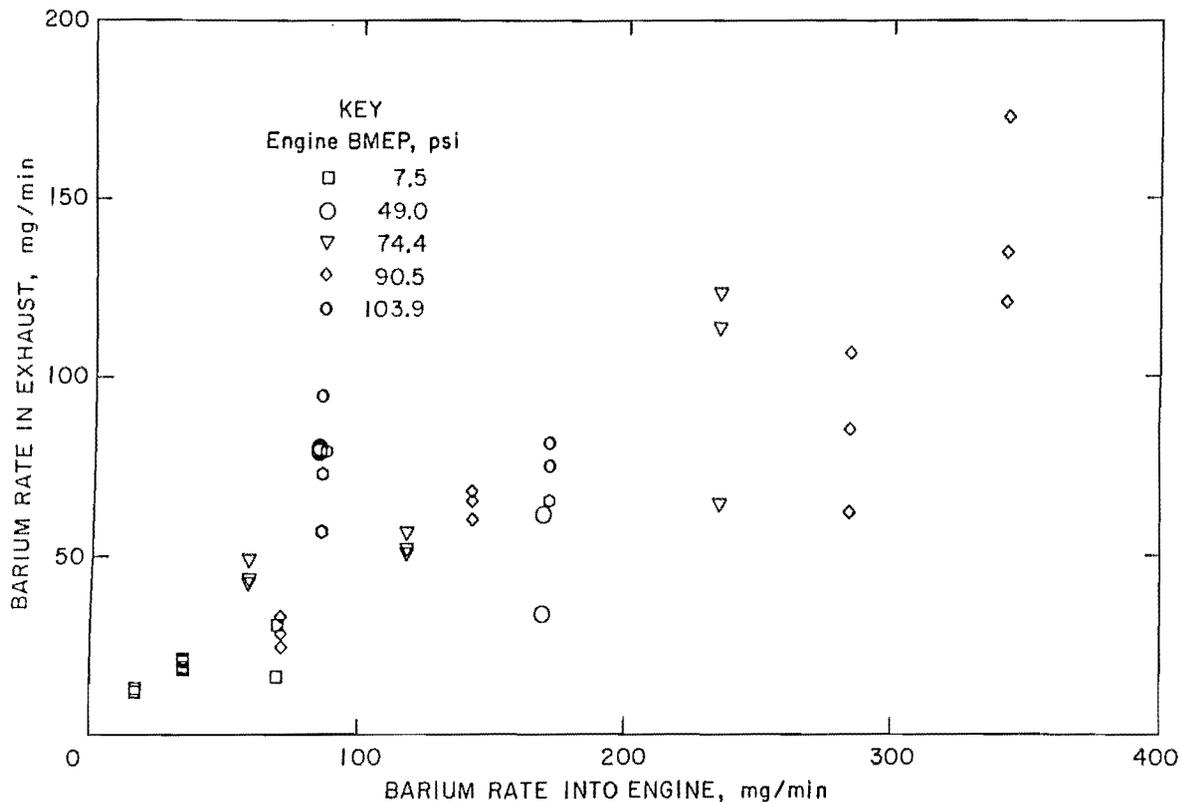


FIGURE 19.—Effect of engine load and barium in fuel on barium in exhaust.

which is toxic. Assuming that all of the barium is in the form of barium sulfate, estimates of the mass concentration of barium compounds in the exhaust are plotted in figure 20. These plots show that barium sulfate concentration ranges between 10 and 90 mg/m^3 and increases with additive concentration and engine load. Note that the actual values are overestimated slightly in proportion to the amount of barium carbonate present.

Carbon Particulate

The mass concentration estimates of carbonaceous soot in figure 21 were obtained by subtracting the sum of the barium sulfate concentrations (fig. 20) and the volatile concentrations (fig. 17) from the total soot concentrations (fig. 4). The carbon concentration differences between untreated and treated fuel (fig. 22) confirm that treated fuels reduce exhaust carbon by 20 to 50 pct for most engine loads. These results agree with

the conclusions by Truex (32), who found that additives reduced carbonaceous soot by 30 pct, and by Tessier (31), who stated that the effect of the additives is to promote more effective combustion of carbon.

Figure 23 shows how the opacity meter response to diesel soot decreases with increasing additive concentration. Plots (not shown) of opacity against nonvolatile mass are similar to those in figure 23 except that they are shifted slightly to the left. Figure 24 shows that opacity meter response is mainly dependent on carbon concentration only. The intercept and slope of the linear regression fit to the data in figure 25 are -0.377 and 0.127 . The correlation coefficient for this fit is 0.99 . These results are consistent with those by MacDonald (23), Scherrer (28), Gerke (12), and Japar (18), all of whom have shown that opacity meter response is linear with the carbon component in exhaust soot.

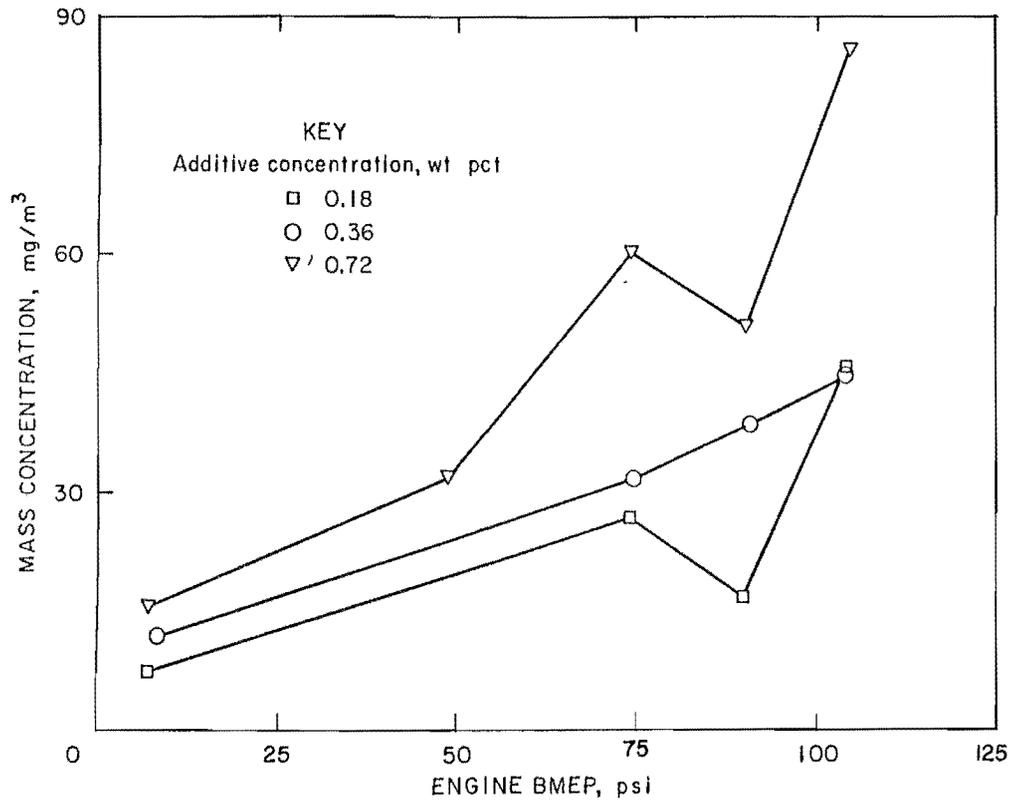


FIGURE 20.—Estimated concentration of barium sulfate in exhaust.

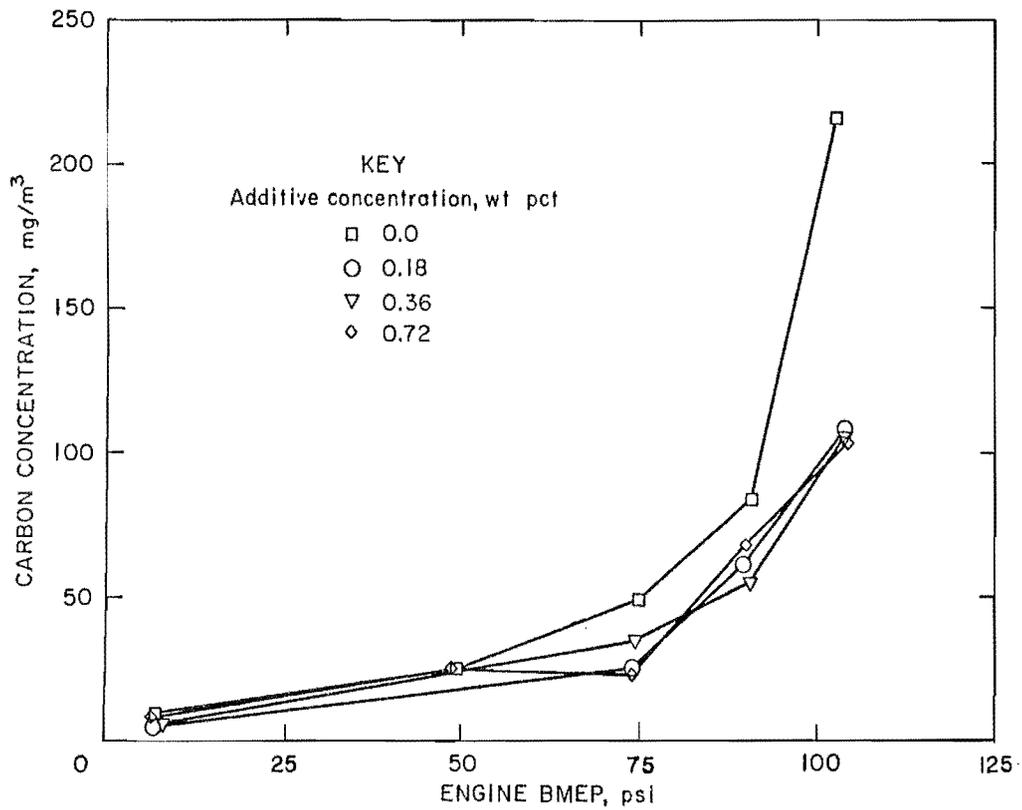


FIGURE 21.—Calculated carbon concentration in exhaust.

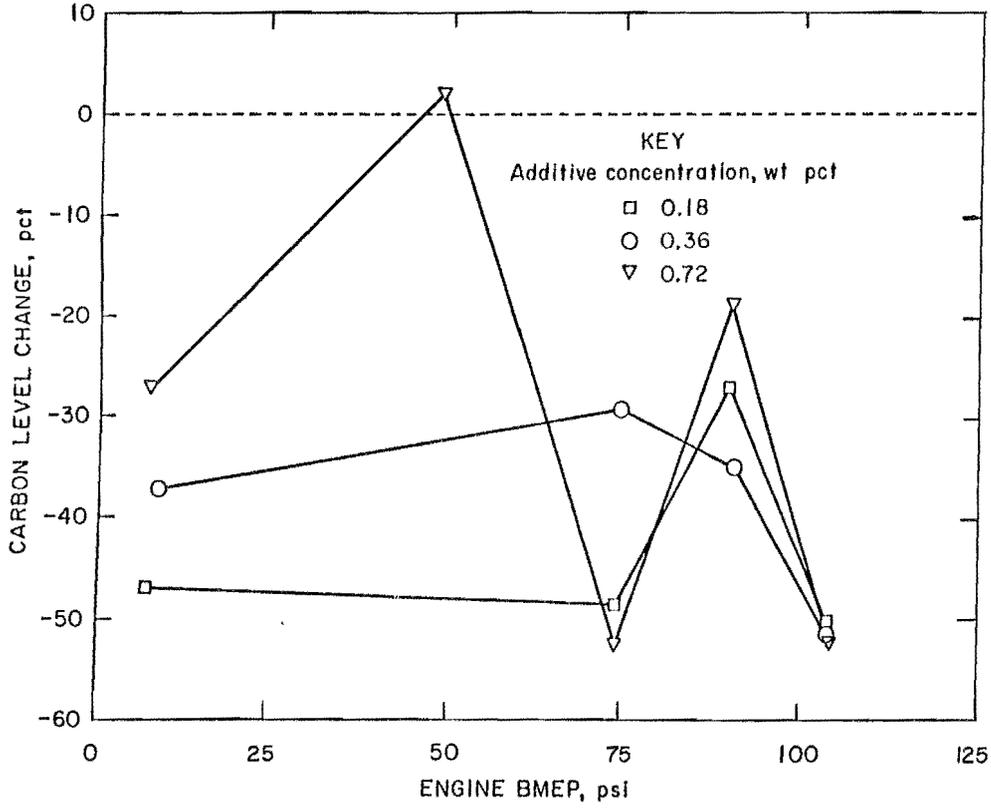


FIGURE 22.—Additive and engine load effects on exhaust carbon concentration.

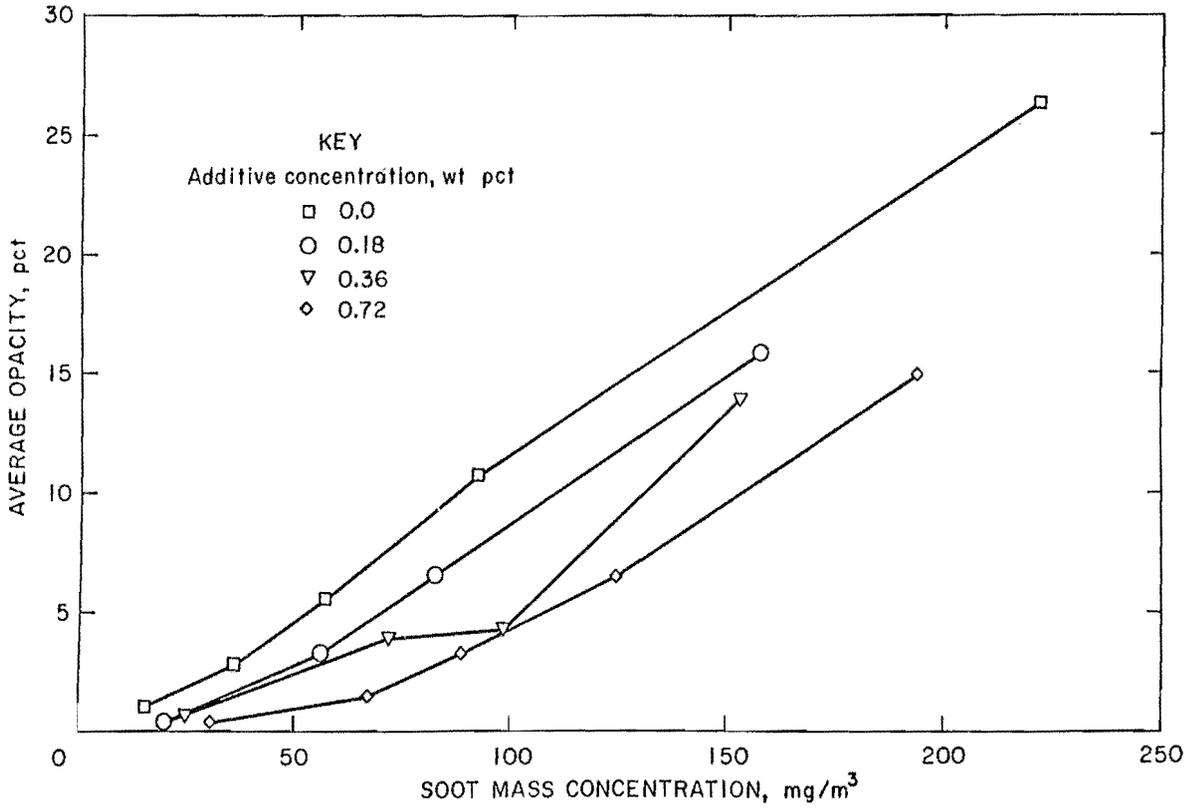


FIGURE 23.—Variation of opacity with soot mass concentration.

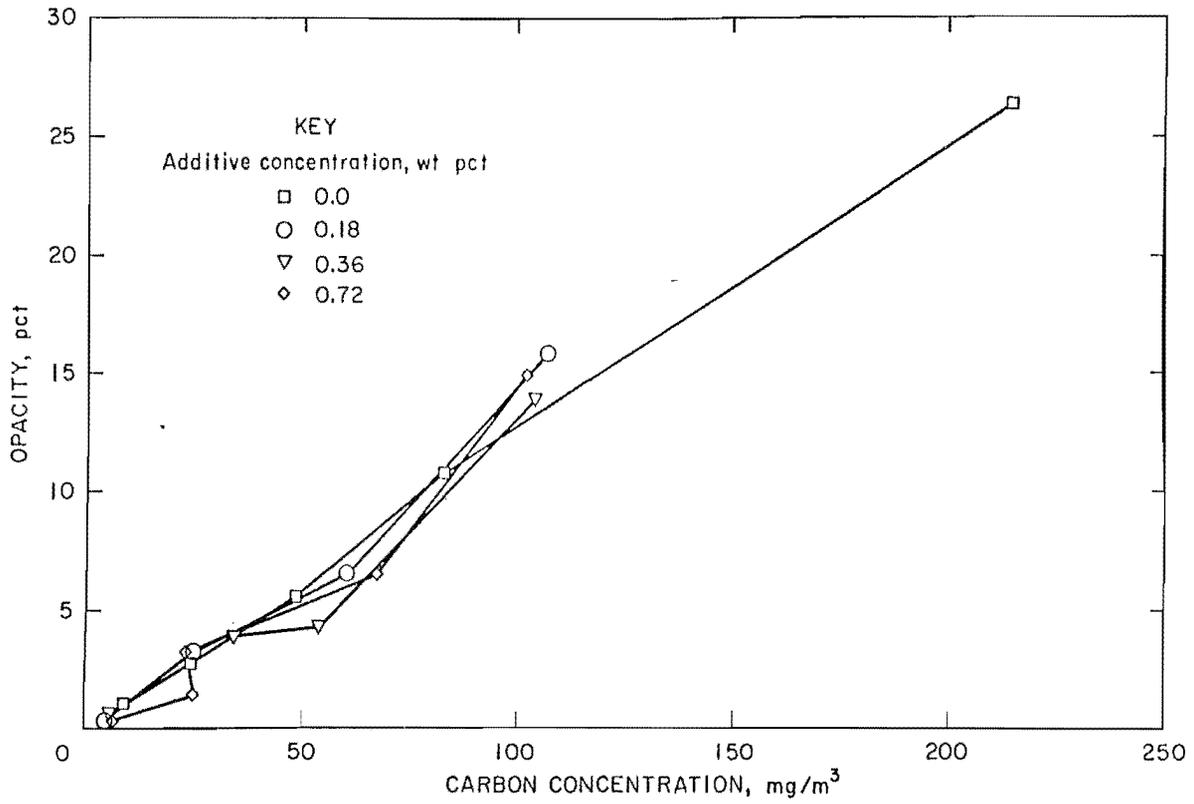


FIGURE 24.—Correlation of opacity with carbon concentration.

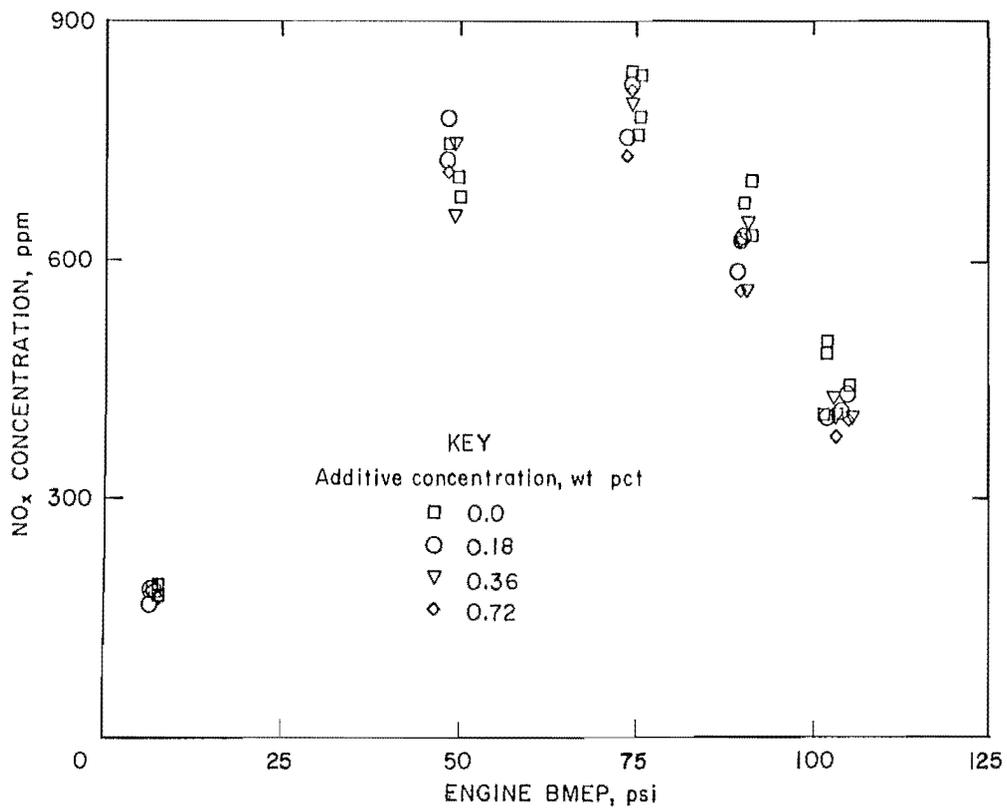


FIGURE 25.—Additive and load effects on NO_x concentration.

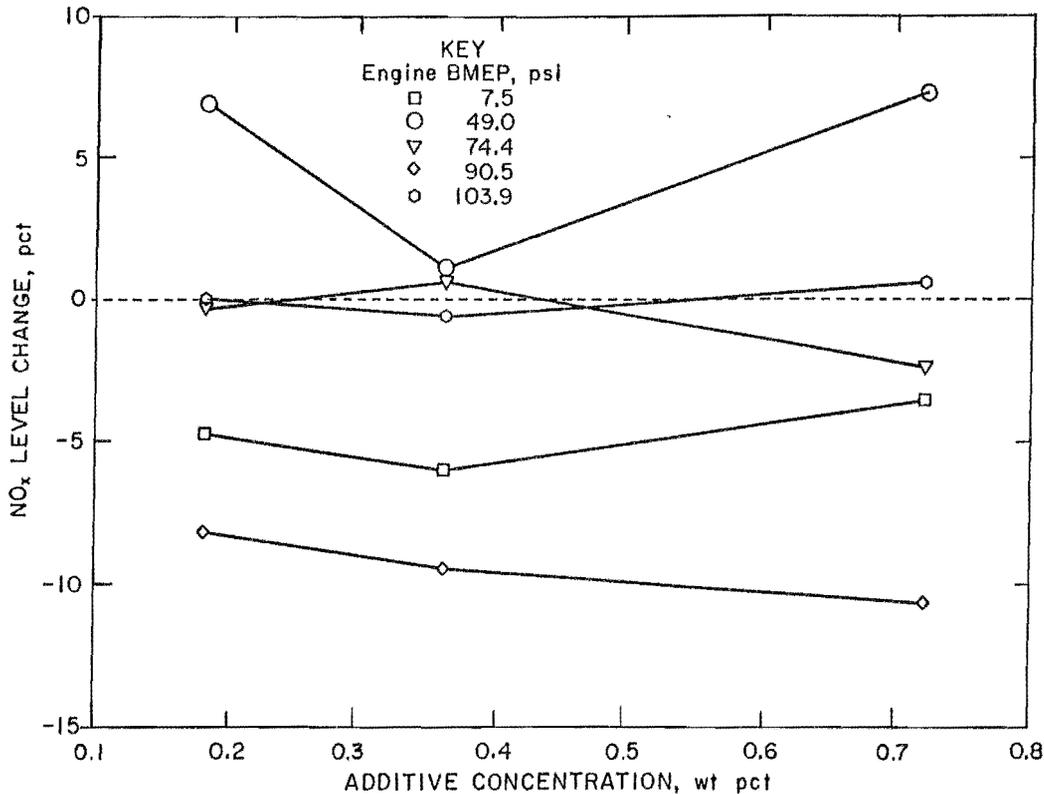


FIGURE 26.—Dependence of NO_x changes on additive and load.

ADDITIVE EFFECTS ON GASEOUS EMISSIONS

The only nonparticulate emissions measured in the exhaust were nitrogen oxides (NO_x) and oxygen. In figure 25, the measured NO_x concentration ranges from about 170 to 900 ppm. These results agree with those obtained by Baumgard (7) on a similar engine. The change (eq. 1) in NO_x levels (fig. 26) show that, in almost all cases, small reductions (up to 10 pct) in NO_x concentration were measured for the treated fuels. These

results are consistent with those obtained by others (5, 13) who observed either no change in NO_x levels or slight reductions that were generally considered insignificant.

Oxygen concentrations in the exhaust are plotted against engine BMEP in figure 27. The average changes in oxygen concentration are plotted in figure 28, which shows that, at many operating conditions, exhaust oxygen was reduced slightly for treated fuels.

HEALTH IMPLICATIONS

A quantitative assessment of the health effects of barium fuel additives on engine exhaust toxicity is beyond the scope of this study. The purpose of this section is to point out that the physical and chemical changes in diesel particulate caused by fuel additives may have beneficial and harmful implications.

The increase (figs. 4-5) in total soot mass concentration in diesel exhaust is a serious objection to the use of barium-based fuel additives. Even though most

of the added particulate is in the form of nontoxic barium sulfate, it adds to the airborne dust level in mines and increases problems of compliance with dust standards. Some of the added particulate are in the form of soluble barium compounds (e.g., barium carbonate), which are toxic. Golothan (13) concluded that the injection of soluble barium compounds into the general environment should not pose a health problem because of the large dilution factors expected and also

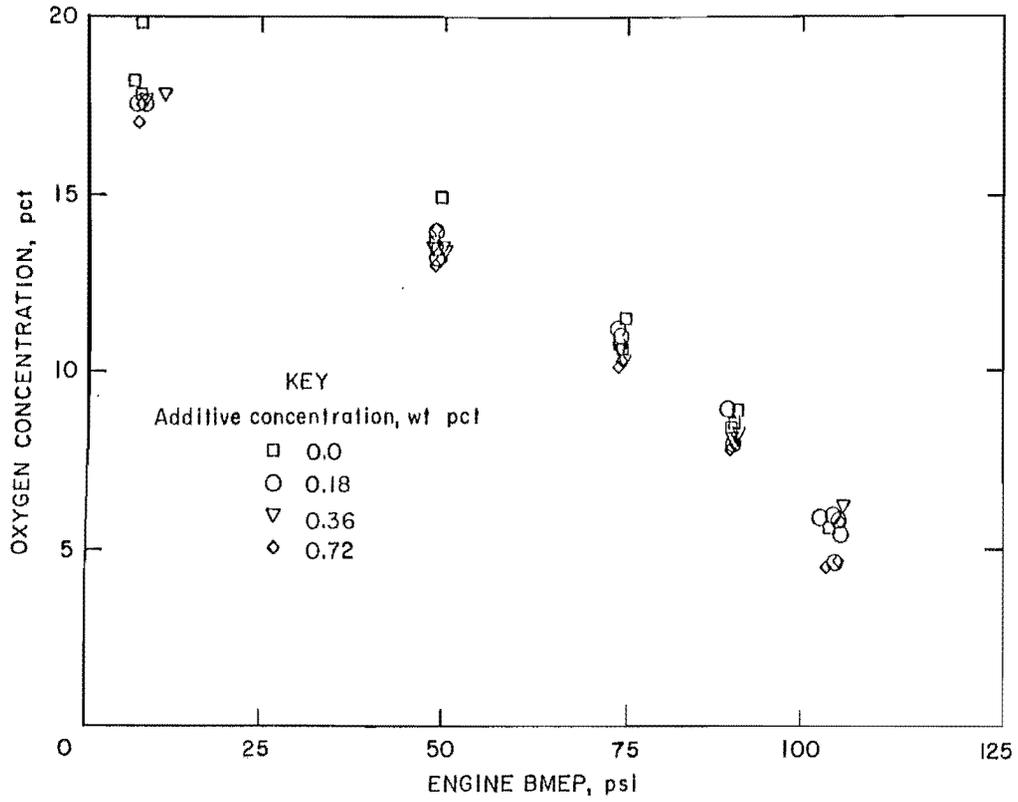


FIGURE 27.—Additive and engine load effects on oxygen level in exhaust.

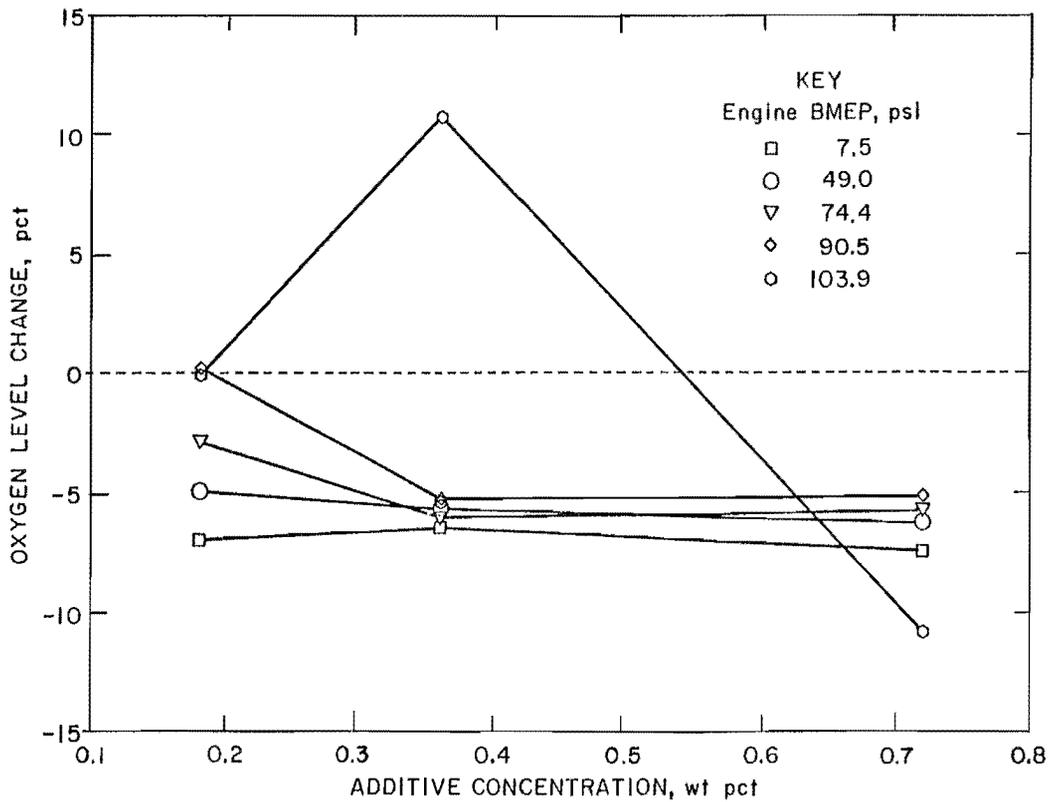


FIGURE 28.—Dependence of oxygen level in exhaust on additive concentration and engine load.

because no problems of chronic exposure to low levels, less than the TLV of soluble barium, have been identified or are expected. However, the effect of the limited ventilation and dilution factors found in mines was not considered.

Our results show that the maximum concentration of barium in the raw exhaust from the engine, at full load, is about 25 mg/m^3 when the recommended additive concentration of 0.36 wt pct is used in the fuel. The assumptions of a maximum upper limit (13) of 25 pct as soluble barium and a "worst" dilution case of 20:1 (6) results in 0.31 mg/m^3 toxic barium in the mine atmosphere for the tested engine at full load. This is less than the full-shift, time-weighted TLV of 0.5 mg/m^3 (3), but there is little margin for error. If more than one piece of equipment is operating in a drift with limited ventilation the TLV could be exceeded even allowing for less than full-load operation.

The results reported here and those by Kittelson (20) show that barium-based fuel additives decrease soot particle size at all engine operating loads. For a fixed mass, surface area increases as particle size decreases. Consequently, barium-based additives in diesel fuel not only increase particulate mass but also increase the surface area for adsorption of potentially harmful substances, which may eventually deposit in the lung.

Particle size also determines the degree of lung penetration and deposition. Pulmonary deposition is minimal (21, 34) for $0.5\text{-}\mu\text{m}$ particles and increases as particle size decreases. The fact that mean soot sizes in the accumulation mode (fig. 14) at all engine loads are similar for treated fuels compared with untreated indicates a potential for increased lung deposition.

The reduction in carbon of up to 50 pct in figure 22 is an important result, but the health significance is not clear because carbon is not generally considered to be a health hazard (29). However, this reduction in carbon may help account for the result in figure 29, which shows that treated fuels reduce volatile hydrocarbons in the soot by up to 50 pct at moderate-to-full loads. Carbon may be a better adsorbant for HC than are barium compounds. Therefore, soot particles composed of both carbon and barium compounds simply do not adsorb volatile substances as effectively as carbon alone. Unfortunately, there is not enough information available to determine any benefits from this observed volatile reduction.

It is important to note that these observations must be qualified by the fact that they are based on data obtained at steady-state engine operating conditions and may not be representative of emissions from engines operated at real-world duty cycles.

SUMMARY OF RESULTS

1. At full load, exhaust particulate levels are inversely related to the oxygen concentration of engine intake air for both treated and untreated fuels. For example, a 6.5 pct reduction of oxygen into the engine (from 0.0155 to 0.0145 lb/ft^3 because of reduced barometric pressure and/or increased temperature and humidity) doubled the soot mass concentration in the exhaust. For untreated fuel, similar but weaker trends were observed at engine loads of 75 and 90 pct of full load. For barium-treated fuels at less than full load, no dependence of soot concentration on oxygen level was observed.

2. Except at light load (7.5 psi BMEP), additive-treated fuel reduced volatile hydrocarbons adsorbed on filter deposits by up to about 50 pct. Although the percent reductions were large in some cases, the absolute reductions were small, a few milligrams per cubic meter, because the actual volatile mass concentration was small.

3. Compared with untreated fuel, using the manufacturer's recommended concentration of additive increased the gravimetrically measured mass concentration of total particulate by 30 to 80 pct for all steady-state engine operating conditions

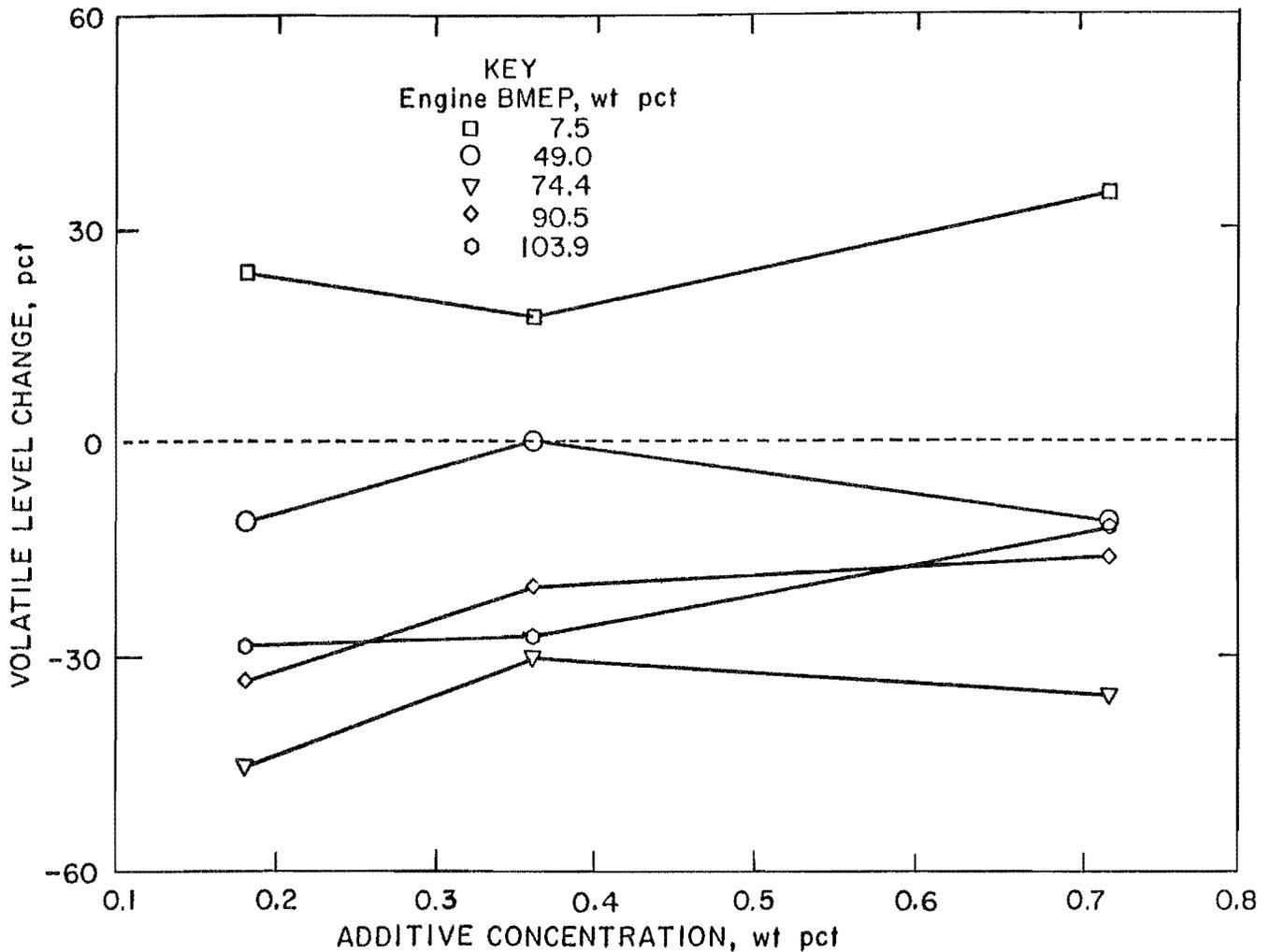


FIGURE 29.—Additive and engine load effects on volatiles.

except full load, where a 30-pct soot reduction was observed.

4. Soot concentration measurements from two different optical smoke meters did not agree with the gravimetric standards. The response of the opacity meter and the Bosch meter were affected by soot particle size and a lack of sensitivity to barium compounds. Both meters underestimated soot concentration when additives were used. Opacity meter measurements correlated linearly with mass concentration of the carbon fraction in the engine exhaust.

5. At constant engine load, average particle sizes were reduced by up to a

factor of two for additive-treated fuel compared with untreated fuel.

6. At most engine loads, the soot carbon fraction was reduced for treated fuels. For example, at full load, the carbon mass concentration was reduced from 210 mg/m^3 for untreated fuel to about 105 mg/m^3 for treated fuel.

7. NO_x emissions were reduced by up to 10 pct at the recommended concentration of additive in the fuel.

8. Atomic adsorption analysis showed that, on the average, the barium found in the exhaust accounted for only about 40 pct of the barium mixed with the fuel sand injected into the engine.

CONCLUSIONS

This research showed that a barium-based fuel additive reduced both the carbon and the hydrocarbon components in the exhaust. Although there is an exposure limit for carbon black, none has been established for respirable carbon in diesel soot. This research did not attempt to identify the specific hydrocarbons affected by additives, and there is no quantitative relationship available for estimating worker exposure to undesirable hydrocarbons based on the filter deposit data. Therefore, no quantifiable health benefits attributable to the use of barium-based additives in diesel fuel were identified.

Unfortunately, additives may also introduce health-related problems into the mining environment. The total particulate from the engine are increased for barium-treated fuel except at full load. As a result, equipment operated at typical duty cycles may actually increase particulate loading in mine air. A

substantial fraction of the barium will end up in the exhaust. Others have determined that up to 25 pct of the exhaust barium may be in a toxic form. Barium-based fuel additives reduce the size of particles in the exhaust. The health effects of reduced particle size are complicated and unclear at this time. Both theoretical and experimental results by others indicate that in the particle size range for diesel soot a decrease in particle size may increase pulmonary deposition.

It is important to note again that the results reported here are for one engine operated at steady-state conditions only. Tests on other engines and for operation at transient loading conditions might produce different results and conclusions. Consequently, any recommendation for or against the use of barium-based fuel additives in underground mining equipment is not appropriate.

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APPENDIX A.--DIESEL FUEL SPECIFICATIONS

Type: D-2 DCF, Lot G-075

TABLE A-1. - Fuel distillation data

		<u>Temp., °F</u>	<u>Distillation, D-86, pct</u>
API ¹ gravity at 60° F.....	35.2		
Sulfur.....wt pct..	0.35		
Particulate matter.....mg/L..	2.07		
Viscosity at 40° C.....cs..	2.52	375.....	(¹)
Flash point (PM).....°F..	162	415.....	5
Cloud point.....°F..	12	431.....	10
Cetane number.....	<u>46.2</u>	451.....	20
Composition (by FIA), vol pct:		469.....	30
Aromatics.....	32.1	487.....	40
Olefins.....	1.33	505.....	50
Paraffins and Naphthenes.....	<u>66.57</u>	523.....	60
<u>Total.....</u>	<u>100.00</u>	543.....	70
		567.....	80
		598.....	90
		628.....	95
		<u>653.....</u>	(²)

¹American Petroleum Institute.

¹Initial boil point.

²Endpoint.

APPENDIX B.--ADDITIVE SPECIFICATIONS

Type: Lubrizol 565

Recommended concentration: 0.36 wt pct, 0.25 vol pct

Specific gravity at 60° F.....	1.22
Viscosity at 100° C.....cs..	9.62
Barium content.....wt pct..	20-25
Sulfur content.....wt pct..	0.25-0.50
Nitrogen content.....wt pct..	0.4-0.6

APPENDIX C.--ENGINE AND EMISSION DATA

TABLE C-1. - Engine and environmental data

Additive conc, date, and mode ¹	BMEP, psi	Fuel rate, lb/h	Exhaust backpressure, in H ₂ O	Barometric backpressure, in Hg	Relative humidity, pct	Engine intake temp., °F
NO ADDITIVE						
11-20-84:						
Mode 1.....	7.8	5.7	10	29.6	20	67
Mode 2.....	50.3	13.9	15	29.7	15	65
Mode 3.....	75.7	19.1	18	29.7	14	67
Mode 4.....	91.6	23.2	19	29.7	15	68
Mode 5.....	102.2	26.1	19	29.7	17	71
11-21-84:						
Mode 1.....	7.8	5.7	10	29.6	16	77
Mode 2.....	50.0	13.9	15	29.7	15	80
Mode 3.....	75.6	19.2	20	29.7	15	82
Mode 4.....	91.5	23.0	20	29.7	17	82
Mode 5.....	102.3	26.3	20	29.7	19	80
11-27-84:						
Mode 1.....	7.3	5.7	10	28.7	21	82
Mode 3.....	75.3	19.4	17	28.7	20	87
Mode 5.....	101.7	27.1	20	28.8	22	87
11-29-84:						
Mode 1.....	7.4	5.8	10	28.9	19	78
Mode 2.....	49.4	13.9	14	28.9	18	83
Mode 3.....	74.9	19.2	18	28.9	17	85
Mode 4.....	90.9	23.2	20	28.9	16	88
Mode 5.....	103.9	27.9	21	28.9	18	89
11-29-84:						
Mode 1.....	6.3	5.8	9	29.5	15	76
Mode 2.....	48.4	14.0	14	29.1	14	81
Mode 3.....	73.9	19.3	17	29.1	13	84
Mode 4.....	90.2	23.3	18	29.1	12	88
Mode 5.....	103.8	28.4	20	29.1	13	87
0.18 wt pct						
12-12-84:						
Mode 1.....	6.5	5.8	19	28.9	13	75
Mode 2.....	48.4	14.0	10	28.9	14	80
Mode 3.....	73.7	19.4	15	28.9	13	83
Mode 4.....	89.5	23.5	20	28.9	14	83
Mode 5.....	102.6	28.3	20	28.9	16	85
12-13-84:						
Mode 4.....	89.7	23.2	20	29.4	13	83
Mode 5.....	104.4	28.2	10	29.4	16	79
12-18-84:						
Mode 1.....	6.7	5.7	10	29.3	11	71
Mode 2.....	48.6	13.9	14	29.4	11	75
Mode 3.....	74.0	19.1	18	29.4	11	78
Mode 4.....	90.1	23.0	19	29.4	11	80
Mode 5.....	105.1	28.0	20	29.3	12	77

See footnotes at end of table.

TABLE C-1. - Engine and environmental data--Continued

Additive conc, date, and mode ¹	BMEP, psi	Fuel rate, lb/h	Exhaust backpressure, in H ₂ O	Barometric backpressure, in Hg	Relative humidity, pct	Engine intake temp., °F
0.18 wt pct--Continued						
1-17-85:						
Mode 1.....	7.9	5.8	10	28.6	14	74
Mode 2.....	48.6	14.0	15	28.6	13	78
Mode 3.....	74.4	19.3	18	28.7	12	82
Mode 4.....	90.5	23.4	20	28.7	11	86
Mode 5.....	104.7	28.4	21	28.7	12	90
0.36 wt pct						
11-30-84:						
Mode 1.....	7.3	NA	10	28.9	19	77
Mode 2.....	49.4	NA	14	28.9	18	81
Mode 3.....	74.7	NA	18	28.9	17	84
Mode 4.....	90.8	NA	19	28.9	17	86
Mode 5.....	103.4	NA	21	28.9	20	83
12-05-84:						
Mode 1.....	10.6	5.6	10	29.1	16	75
Mode 2.....	48.7	13.6	15	29.1	15	82
Mode 3.....	74.2	19.0	19	29.1	15	85
Mode 4.....	90.4	23.2	20	29.1	15	85
Mode 5.....	103.1	27.5	21	29.1	17	82
1-21-85:						
Mode 1.....	7.7	5.7	11	29.0	11	72
Mode 2.....	49.5	13.8	15	29.0	11	75
Mode 3.....	74.4	19.0	19	29.0	10	78
Mode 4.....	91.1	23.1	20	29.0	11	80
Mode 5.....	105.8	27.4	21	29.0	11	79
0.72 wt pct						
12-06-84:						
Mode 1.....	NA	5.7	11	29.5	16	73
Mode 2.....	48.6	13.8	15	29.5	16	76
Mode 3.....	73.9	19.0	20	29.5	16	78
Mode 4.....	90.3	22.9	20	29.5	16	80
Mode 5.....	105.2	28.0	21	29.5	17	77
12-11-84:						
Mode 1.....	6.9	5.8	10	28.6	23	78
Mode 2.....	48.5	13.9	14	28.6	21	81
Mode 3.....	73.7	19.3	18	28.7	20	86
Mode 4.....	89.9	23.4	19	28.7	19	89
Mode 5.....	103.5	28.6	21	28.7	25	83
1-23-85:						
Mode 1.....	7.4	5.7	10	28.8	13	73
Mode 2.....	48.9	13.9	15	28.8	12	78
Mode 3.....	74.4	19.3	18	28.8	12	80
Mode 4.....	90.3	23.6	19	28.8	12	83
Mode 5.....	105.0	28.8	22	28.8	13	82

NA Not available.

¹Engine test loads as defined in table 1.

TABLE C-2. - Dilution¹ ratios

Additive conc, date, and mode	Primary dilution ratio	Secondary dilution ratio		Total dilution	Additive conc, date, and mode	Primary dilution ratio	Secondary dilution ratio		Total dilution
		Stage 1	Stage 2				Stage 1	Stage 2	
NO ADDITIVE					0.18 wt pct--Continued				
11-20-84:					1-17-85:				
Mode 1.	30.5	56.1	1.00	1710	Mode 1.	26.9	68.0	1.00	1827
Mode 2.	30.9	56.1	2.55	4412	Mode 2.	25.3	43.0	4.33	4717
Mode 3.	27.2	57.0	2.58	3998	Mode 3.	24.1	43.0	4.33	4492
Mode 4.	27.1	43.0	2.06	2396	Mode 4.	24.3	43.0	4.33	4514
Mode 5.	28.8	66.0	2.99	5680	Mode 5.	25.1	43.0	4.33	4665
11-21-84:					0.36 wt pct				
Mode 1.	30.4	43.0	1.00	1307	11-30-84:				
Mode 2.	28.0	69.0	3.14	6059	Mode 1.	29.0	43.0	4.33	5406
Mode 3.	26.6	47.9	2.28	2905	Mode 2.	27.0	48.4	4.77	6222
Mode 4.	26.9	43.0	2.06	2380	Mode 3.	25.3	48.4	4.77	5843
Mode 5.	28.0	74.0	3.40	7050	Mode 4.	25.6	48.4	4.77	5899
11-27-84:					Mode 5.	26.2	48.4	4.77	6053
Mode 1.	29.9	43.0	1.00	1285	12-05-84:				
Mode 3.	26.5	75.0	1.00	1986	Mode 1.	29.0	43.0	4.33	5402
Mode 5.	27.8	43.0	4.33	5171	Mode 2.	27.1	48.4	4.77	6245
11-29-84:					Mode 3.	26.3	48.4	4.77	6079
Mode 1.	29.1	43.0	1.00	1252	Mode 4.	25.6	48.4	4.77	5919
Mode 2.	26.6	43.0	4.33	4952	Mode 5.	26.2	48.4	4.77	6040
Mode 3.	25.2	43.0	4.33	4695	1-21-85:				
Mode 4.	25.3	77.0	1.00	1945	Mode 1.	26.6	43.0	4.33	4950
Mode 5.	26.1	43.0	4.33	4853	Mode 2.	25.3	48.4	4.77	5836
11-29-84:					Mode 3.	24.0	48.4	4.77	5548
Mode 1.	28.8	48.4	1.00	1394	Mode 4.	24.1	48.4	4.77	5573
Mode 2.	26.7	67.0	1.00	1789	Mode 5.	24.7	48.4	4.77	5698
Mode 3.	25.2	67.0	1.00	1689	0.72 wt pct				
Mode 4.	25.4	43.0	4.33	4726	12-06-84:				
Mode 5.	26.2	43.0	4.33	4870	Mode 1.	28.5	43.0	4.33	5311
0.18 wt pct					Mode 2.	26.8	57.0	5.37	8193
12-12-84:					Mode 3.	25.1	57.0	5.37	7667
Mode 1.	28.7	38.5	4.19	4635	Mode 4.	25.1	57.0	5.37	7685
Mode 2.	26.8	43.0	4.33	4991	Mode 5.	26.1	57.0	5.37	7976
Mode 3.	25.2	43.0	4.33	4695	12-11-84:				
Mode 4.	25.4	43.0	4.33	4719	Mode 1.	28.4	43.0	4.33	5279
Mode 5.	26.3	43.0	4.33	4899	Mode 2.	26.6	57.0	5.37	8129
12-13-84:					Mode 3.	25.0	57.0	5.37	7639
Mode 4.	25.5	43.0	4.33	4741	Mode 4.	25.4	57.0	5.37	7774
Mode 5.	25.8	43.0	4.33	4810	Mode 5.	25.9	57.0	5.37	7933
12-18-84:					1-23-85:				
Mode 1.	28.8	77.0	1.00	2216	Mode 1.	26.9	74.0	1.00	1987
Mode 2.	26.8	43.0	4.33	4985	Mode 2.	24.9	43.0	4.33	4631
Mode 3.	25.3	43.0	4.33	4700	Mode 3.	23.8	43.0	4.33	4427
Mode 4.	25.4	43.0	4.33	4728	Mode 4.	23.9	48.4	4.77	5527
Mode 5.	26.1	43.0	4.33	4864	Mode 5.	24.6	48.4	4.77	5689

¹Ratio of total diluted volume flow to sample flow (figs. 2-3).

TABLE C-3. - Emissions

Additive conc, date, and mode	NO _x conc, ppm	Exhaust O ₂ conc, pct	Bosch number	Opac-ity, pct	Soot mass conc, mg/m ³	Additive conc, date, and mode	NO _x conc, ppm	Exhaust O ₂ conc, pct	Bosch number	Opac-ity, pct	Soot mass conc, mg/m ³
NO ADDITIVE						0.18 wt pct--Continued					
11-20-84:						1-17-85:					
Mode 1.	178	NA	0.9	1.5	14.7	Mode 1.	175	17.5	1.2	0.3	19.5
Mode 2.	675	NA	1.4	3.1	38.7	Mode 2.	775	13.2	2.0	1.0	42.2
Mode 3.	775	NA	2.6	5.5	50.2	Mode 3.	815	10.2	2.6	2.8	54.1
Mode 4.	625	NA	3.1	9.3	77.3	Mode 4.	620	7.9	3.1	6.2	77.4
Mode 5.	475	NA	5.3	16.0	144.4	Mode 5.	390	4.6	4.7	17.3	165.8
11-21-84:						0.36 wt pct					
Mode 1.	188	NA	.8	1.2	15.7	11-30-84:					
Mode 2.	700	NA	1.5	2.6	35.1	Mode 1.	172	17.8	0.6	NA	23.6
Mode 3.	828	NA	2.3	5.3	54.9	Mode 2.	650	13.5	1.5	0.1	53.6
Mode 4.	695	NA	3.2	10.9	97.6	Mode 3.	NA	10.3	2.1	3.3	76.2
Mode 5.	490	NA	4.3	17.9	148.2	Mode 4.	555	8.2	2.8	6.1	99.5
11-27-84:						Mode 5.	395	NA	3.7	11.7	155.7
Mode 1.	183	NA	.7	.7	15.8	12-05-84:					
Mode 3.	753	NA	2.8	5.8	65.7	Mode 1.	NA	17.8	.7	.8	25.2
Mode 5.	400	NA	5.6	29.2	269.0	Mode 2.	NA	13.5	1.2	1.7	53.9
11-29-84:						Mode 3.	NA	10.5	2.0	4.1	70.0
Mode 1.	NA	19.8	.5	1.2	16.3	Mode 4.	NA	8.2	3.3	6.7	104.0
Mode 2.	NA	14.9	1.7	3.4	39.7	Mode 5.	420	NA	4.6	16.1	168.3
Mode 3.	NA	11.5	2.5	6.8	63.9	1-21-85:					
Mode 4.	NA	8.9	4.0	13.4	111.3	Mode 1.	172	17.7	.8	.5	25.8
Mode 5.	400	5.6	5.5	39.4	304.7	Mode 2.	740	13.5	1.1	2.7	54.4
11-29-84:						Mode 3.	790	10.6	1.6	4.3	68.8
Mode 1.	NA	18.2	1.2	.8	14.7	Mode 4.	640	8.3	2.3	NA	91.6
Mode 2.	NA	13.7	2.0	2.2	30.9	Mode 5.	395	6.2	3.8	13.7	135.2
Mode 3.	NA	10.8	2.7	4.3	50.7	0.72 wt pct					
Mode 4.	NA	8.5	4.0	9.4	83.3	12-06-84:					
Mode 5.	NA	5.6	6.2	29.1	242.9	Mode 1.	NA	18.2	0.8	0.1	36.5
0.18 wt pct						Mode 2.	NA	14.0	1.6	1.1	80.7
12-12-84:						Mode 3.	NA	11.1	1.7	2.1	99.9
Mode 1.	183	17.5	0.9	1.7	19.6	Mode 4.	NA	8.9	3.3	4.2	125.3
Mode 2.	720	13.9	1.4	.4	40.8	Mode 5.	NA	5.8	5.0	8.8	182.8
Mode 3.	750	11.2	2.2	3.9	61.1	12-11-84:					
Mode 4.	580	9.0	2.7	8.1	97.2	Mode 1.	178	17.0	.8	.3	36.2
Mode 5.	395	5.9	5.3	19.2	196.1	Mode 2.	705	13.0	1.6	1.2	77.7
12-13-84:						Mode 3.	725	10.1	1.7	3.8	108.6
Mode 4.	618	9.0	3.1	5.9	82.3	Mode 4.	555	7.8	3.3	7.8	156.5
Mode 5.	405	6.0	4.5	14.9	150.8	Mode 5.	370	4.5	5.0	19.3	234.8
12-18-84:						1-23-85:					
Mode 1.	165	18.0	1.0	.5	20.8	Mode 1.	175	17.6	1.0	.7	19.1
Mode 2.	710	13.7	1.6	1.5	37.6	Mode 2.	770	13.2	1.5	2.0	42.3
Mode 3.	NA	11.0	2.8	3.2	53.2	Mode 3.	808	10.3	2.2	3.9	56.5
Mode 4.	NA	8.9	3.9	5.9	71.9	Mode 4.	624	8.0	3.1	7.5	91.2
Mode 5.	425	5.9	5.8	11.8	117.5	Mode 5.	390	4.7	4.2	16.5	164.1

NA Not available.

TABLE C-4. - Bimodal EAA results

Additive conc, date, and mode	GSD ¹	Nuclei mode				Accumulation mode			Both modes	
		Number fraction	Volume fraction	Mean diam, μm		GSD ¹	Mean diam, μm		Number conc, 100/cm ³	Volume conc, $\mu\text{m}^3/\text{cm}^3$
				Number	Volume		Number	Volume		
NO ADDITIVE										
11-20-84:										
Mode 1.	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Mode 2.	1.60	0.99	0.025	0.010	0.019	1.97	0.110	0.437	641	10.6
Mode 3.	1.91	.99	.064	.006	.019	2.12	.047	.256	400	14.1
Mode 4.	1.92	.99	.012	.005	.018	1.93	.099	.363	312	44.1
Mode 5.	1.97	.98	.010	.006	.022	1.97	.095	.373	188	30.8
11-21-84:										
Mode 1.	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Mode 2.	1.60	.99	.123	.010	.020	1.90	.066	.230	265	4.6
Mode 3.	2.09	.99	.052	.005	.026	1.93	.072	.264	380	23.6
Mode 4.	1.97	.97	.011	.006	.023	1.91	.088	.308	192	46.1
Mode 5.	1.76	.98	.012	.006	.015	1.89	.083	.281	148	26.4
11-27-84:										
Mode 1.	2.00	.96	.069	.029	.121	2.01	.068	.292	82	14.9
Mode 3.	2.21	.97	.016	.006	.037	2.33	.060	.518	184	51.6
Mode 5.	1.68	.52	.001	.012	.028	1.66	.114	.247	30	57.6
11-29-84:										
Mode 1.	2.01	.96	.728	.025	.108	1.78	.067	.183	100	11.1
Mode 2.	1.71	.99	.119	.012	.029	1.98	.078	.316	191	6.3
Mode 3.	1.85	.99	.034	.007	.023	2.00	.079	.330	180	12.2
Mode 4.	1.98	.89	.014	.009	.037	2.05	.072	.339	181	58.9
Mode 5.	1.97	.64	.001	.006	.022	1.89	.081	.273	50	62.1
11-29-84:										
Mode 1.	1.98	.92	.390	.020	.082	1.94	.056	.207	108	9.6
Mode 2.	1.60	.99	.076	.014	.027	1.90	.110	.379	510	20.2
Mode 3.	1.89	.99	.064	.009	.031	2.09	.081	.418	431	34.6
Mode 4.	1.90	.97	.009	.007	.023	1.89	.097	.324	69	19.6
Mode 5.	2.47	.54	.001	.005	.060	2.05	.080	.375	28	55.2
0.18 wt pct										
12-12-84:										
Mode 1.	1.69	0.99	0.764	0.020	0.046	1.86	0.052	0.166	73	1.5
Mode 2.	1.30	.91	.384	.046	.056	1.87	.072	.234	80	12.7
Mode 3.	1.53	.94	.313	.030	.051	1.83	.071	.214	104	9.8
Mode 4.	1.50	.92	.228	.030	.049	1.76	.081	.213	112	13.6
Mode 5.	1.43	.91	.099	.028	.041	1.47	.122	.191	114	20.1
1-17-85:										
Mode 1.	1.69	.99	.750	.022	.051	1.66	.072	.156	229	6.3
Mode 2.	1.37	.95	.469	.031	.042	1.77	.064	.169	97	5.1
Mode 3.	1.49	.94	.242	.029	.046	1.84	.077	.233	113	10.2
Mode 4.	1.47	.92	.180	.029	.045	1.79	.081	.226	116	14.5
Mode 5.	1.56	.88	.064	.025	.044	1.34	.136	.177	97	25.8

See footnotes at end of table.

TABLE C-4. - Bimodal EAA results--Continued

Additive conc, date, and mode	GSD ¹	Nuclei mode				Accumulation mode			Both modes	
		Number fraction	Volume fraction	Mean diam, μm		GSD ¹	Mean diam, μm		Number conc, 100/cm ³	Volume conc, $\mu\text{m}^3/\text{cm}^3$
				Number	Volume		Number	Volume		
0.36 wt pct										
11-30-84:										
Mode 1.	1.93	0.99	0.919	0.022	0.080	1.87	0.046	0.150	77	2.8
Mode 2.	1.42	.81	.463	.036	.053	1.82	.043	.126	80	6.1
Mode 3.	1.56	.91	.366	.037	.066	2.03	.061	.275	84	12.6
Mode 4.	1.57	.96	.257	.037	.068	2.02	.096	.425	85	19.8
Mode 5.	1.61	.88	.100	.033	.064	2.33	.063	.544	92	31.4
12-05-84:										
Mode 1.	1.49	.98	.798	.030	.049	1.83	.054	.160	73	2.7
Mode 2.	1.50	.98	.525	.037	.061	1.98	.085	.343	94	9.2
Mode 3.	1.47	.81	.305	.035	.055	1.89	.051	.171	89	10.6
Mode 4.	1.49	.94	.290	.035	.057	1.55	.115	.203	93	14.9
Mode 5.	.42	.86	.101	.031	.045	1.64	.098	.204	98	23.0
1-21-85:										
Mode 1.	1.44	.98	.875	.031	.046	1.77	.047	.123	83	2.6
Mode 2.	1.54	.98	.901	.037	.064	1.56	.066	.118	98	6.3
Mode 3.	1.79	.81	.774	.033	.091	1.61	.042	.083	116	10.3
Mode 4.	1.44	.70	.235	.034	.050	1.77	.049	.131	80	12.9
Mode 5.	1.40	.64	.107	.030	.042	1.84	.050	.153	124	18.2
0.72 wt pct										
12-06-84:										
Mode 1.	1.43	0.93	0.853	0.035	0.051	1.70	0.036	0.084	91	4.0
Mode 2.	1.52	.81	.712	.047	.080	1.80	.044	.125	61	8.5
Mode 3.	1.31	.63	.202	.040	.031	1.62	.061	.117	NA	11.7
Mode 4.	1.42	.68	.275	.043	.063	1.80	.056	.156	71	13.2
Mode 5.	1.54	.62	.204	.038	.068	1.88	.052	.172	81	17.4
12-11-84:										
Mode 1.	1.49	.98	.908	.038	.062	1.50	.063	.103	79	5.1
Mode 2.	1.51	.98	.762	.041	.068	1.64	.094	.196	60	5.9
Mode 3.	1.56	.86	.461	.039	.071	1.73	.064	.158	70	9.9
Mode 4.	1.43	.71	.176	.037	.054	1.78	.061	.164	75	14.6
Mode 5.	1.44	.76	.103	.034	.051	1.77	.078	.207	81	23.7
1-23-85:										
Mode 1.	1.41	.95	.367	.025	.036	1.83	.057	.169	108	7.3
Mode 2.	1.83	.93	.339	.029	.040	1.86	.057	.181	99	6.0
Mode 3.	1.44	.89	.195	.029	.043	1.91	.060	.212	112	11.0
Mode 4.	1.45	.80	.123	.029	.044	1.92	.058	.207	111	17.8
Mode 5.	1.39	.73	.047	.027	.038	2.00	.060	.252	111	29.0

NA Not available.

¹Geometric standard deviation.

TABLE C-5. - Barium and volatile data

Additive conc, date, and mode	Average nonvolatile fraction	Average barium fraction	Additive conc, date, and mode	Average nonvolatile fraction	Average barium fraction
NO ADDITIVE			0.18 wt pct--Continued		
11-20-84:			1-17-85:		
Mode 1.....	0.6	0	Mode 1.....	0.62	0.22
Mode 2.....	.68	0	Mode 2.....	.75	NA
Mode 3.....	.85	0	Mode 3.....	.92	.28
Mode 4.....	.9	0	Mode 4.....	.94	.12
Mode 5.....	.97	0	Mode 5.....	.97	.17
11-21-84:			0.36 wt pct		
Mode 1.....	.6	0	11-30-84:		
Mode 2.....	.68	0	Mode 1.....	0.71	0.28
Mode 3.....	.85	0	Mode 2.....	.79	NA
Mode 4.....	.9	0	Mode 3.....	.92	.26
Mode 5.....	.97	0	Mode 4.....	.94	.23
11-27-84:			Mode 5 ¹97	.17
Mode 1.....	.6	0	12-05-84:		
Mode 3.....	.85	0	Mode 1.....	.71	.28
Mode 5.....	.97	0	Mode 2.....	.79	NA
11-29-84:			Mode 3.....	.92	.26
Mode 1.....	.6	0	Mode 4.....	.94	.23
Mode 2.....	.68	0	Mode 5.....	.97	.17
Mode 3.....	.85	0	1-21-85:		
Mode 4.....	.9	0	Mode 1.....	.71	.28
Mode 5.....	.97	0	Mode 2.....	.79	NA
11-29-84:			Mode 3.....	.92	.26
Mode 1.....	.6	0	Mode 4.....	.94	.23
Mode 2.....	.68	0	Mode 5.....	.97	.17
Mode 3.....	.85	0	0.72 wt pct		
Mode 4.....	.9	0	12-06-84:		
Mode 5.....	.97	0	Mode 1.....	0.73	0.3
0.18 wt pct			Mode 2.....	.85	.28
12-12-84:			Mode 3.....	.94	.4
Mode 1.....	0.62	0.22	Mode 4.....	.95	.24
Mode 2.....	.75	NA	Mode 5.....	.97	.26
Mode 3.....	.92	.28	12-11-84:		
Mode 4.....	.94	.12	Mode 1.....	.73	.3
Mode 5.....	.97	.17	Mode 2.....	.85	.28
12-13-84:			Mode 3.....	.94	.4
Mode 4.....	.94	.12	Mode 4.....	.95	.24
Mode 5.....	.97	.17	Mode 5.....	.97	.26
12-18-84:			1-23-85:		
Mode 1.....	.62	.22	Mode 1.....	.73	.3
Mode 2.....	.75	NA	Mode 2.....	.85	.28
Mode 3.....	.92	.28	Mode 3.....	.94	.4
Mode 4.....	.94	.12	Mode 4.....	.95	.24
Mode 5.....	.97	.17	Mode 5.....	.97	.26

NA Not available.