PARTICULATE MATTER EMISSION FROM ON-ROAD LIGHT DUTY VEHICLES

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INTRODUCTION

Mobile sources contribute to the atmospheric burden of particulate matter (PM) through three mechanisms: primary PM emissions; secondary PM formation, and fugitive emissions. Primary particles are those directly emitted by vehicles. They are emitted in the vehicle exhaust, and also are emitted via brake and tire wear. Secondary PM formation refers to the production of particles in the atmosphere from the reactions of gaseous emissions. Gases of concern are HC, NO_x, and SO_2. HC reacts in the atmosphere to form less volatile species that condense into the particle phase, thereby contributing to the carbonaceous particle burden. NO_x oxidizes to nitric acid which reacts with ammonia to form ammonium nitrate. The amount of ammonium nitrate in the particle phase is a function of temperature. SO_2 is oxidized to sulfuric acid, which exists primarily in the particle phase. Mobile sources are generally a major contributor to the HC and NOx burdens in urban areas, but are minor contributors to the SO_2 in most areas.

Fugitive emissions are PM that is re-suspended by the turbulence caused by vehicle traffic. The amount of dust generated is a function of the amount of fine particles (silt) on or near the roadway. This is a major source of PM-10 (particles with a diameter less than or equal to 10 μm), but will be less important for the new PM-2.5 (particles with a diameter less than or equal to 2.5 μm) standard since re-intrained dust is comprised mostly of large particles. The focus of this paper will be on the primary PM emissions from light-duty vehicles. The emphasis will be on exhaust emissions.

HISTORICAL PERSPECTIVE

Gasoline Vehicles: Exhaust particulate matter (PM) emissions have decreased dramatically in the last 25 years. Before 1975, gasoline vehicles were designed to operate on leaded fuel. The average lead content of fuel was 2 g/gal. Approximately 75% of the lead consumed was emitted as exhaust PM. This resulted in average PM emission rates of 200 to 250 mg/mi (Cadle et al, 1979). A few measurements of 1973 and 1974 vehicles operated on unleaded gasoline were reported as well. These vehicles had PM emission rates of approximately 25 mg/mi.

Leaded gasoline remained available, although at greatly reduced lead concentrations, until 1996. Average lead concentrations in gasoline were 1.4 g/gal in 1980, 0.5 g/gal in 1985 and less than 0.1 g/gal after 1985. As of 1996, no lead additives are allowed in gasoline in the United States. The actual rate at which lead PM emissions were phased out is a combination of three factors: fleet turnover to catalyst vehicles, misfueling of catalyst vehicles with leaded fuels, and the reduced concentrations of lead in gasoline. The phase out is dramatically reflected in the 1994 air quality data which show lead concentrations reduced by 97% since 1975, despite major increases in vehicle miles traveled (EPA, 1995).

Oxidation catalysts were introduced on vehicles in 1975. Since lead poisons these catalysts, the vehicles were required to operate on unleaded fuels. By 1977 all new cars were equipped with catalysts. Introduction on light-duty trucks was somewhat slower, with 7% of these vehicles being non-catalyst in 1978, and non-catalyst vehicles completely phased out by 1982.

The introduction of the oxidation catalyst raised a new concern, namely that vehicles would emit sulfuric acid particles. Sulfur is naturally present in crude oil, and is present in gasoline as a variety of organo-sulfur compounds. The national average sulfur content of gasoline in 1975 was approximately 0.03% by weight.

During combustion almost all of the sulfur is converted to sulfur dioxide, which remains in the gaseous state in the exhaust. These sulfur dioxide emissions are a very small portion of the total anthropogenic sulfur emissions inventory in most locations, and thus were of little concern. Oxidation catalysts, however, were capable of promoting the oxidation of a small fraction of the sulfur to sulfur trioxide (SO_3), which reacts immediately with water to form sulfuric acid. Typical sulfate emission rates from vehicles with oxidation catalysts with and without air pumps were 16 and 5 mg/mi, respectively, at an average speed of 19.6 mph (the average Federal Test Procedure speed) and 20 and 5 mg/mi, respectively, at higher speeds. These figures include the mass of the water associated with the sulfuric acid. Changes in catalyst tech-nology and improved A/F control have greatly reduced sulfuric acid emissions, although few measurements have been reported for late-model vehicles. Furthermore, reductions in fuel sulfur content will reduce the sulfate emission rate. Thus, in California, which has adopted the use of low sulfur fuels (<0.005% S), the sulfate emissions will be considerably lower than the values stated above.

Improvements in vehicle emission control tech-nology have continued to reduce exhaust PM emission rates. For example, Mulawa and Dasch (1995) reported average FTP PM emission rates from 12 1987-1990 light-duty gasoline vehicles of 7 mg/mi. A more recent study, discussed later in this report, shows that PM emission rates have been reduced further.

Diesel Vehicles:. Light-duty diesel vehicles remain a very small fraction of the on-road light-duty vehicle fleet (<1%). However, their relatively high PM emission rates may make their contribution to the light-duty vehicle exhaust particulate inventory significant. Cadle et al. (1979), reported that 1978 and 1979 production light-duty diesels had PM emission rates of 700-850 mg/mi. Similarly, Energy and Environmental Analysis (1985) reported that pre-1981 light-duty diesels averaged 700 mg/mi. Post-1987 diesels were reported to have emission rates of 200 - 260 mg/mi. All Federal Tier 0 vehicles had to meet a PM standard of 200 mg/mi, while Federal Tier 1 vehicles must meet a PM standard of 80 mg/mi. While they apply to all light-duty vehicles, these standards were set to control diesel emissions. Thus, considerable progress has been made in controlling diesel PM emissions as well as gasoline PM emissions.

Tires and Brakes: Particle emissions from tires are dominated by very large particles that are produced under high-wear driving modes (cornering, braking, accelerating). It was estimated in 1978 that the average tire wear rate was 140 mg/mi per tire. Most of the particles formed, however, were found to be very large. Tire wear rates have improved, but there have not been any additional comprehensive emission characterization studies. Brake wear emissions have also been studied. Most of the studies, however, focused on asbestos brake pads. Since asbestos has been largely eliminated from the brake pads used on new vehicles, the current fine particle emission rate from brakes is not well known.

EMISSION MODELS

PART5 is the U.S. EPA's official model for projecting PM emissions from on-road motor vehicles. Most of the elements of this model are included in the California EMFAC model as well. PART5 uses the same vehicle classes and activity data as the MOBILE5 model, and estimates PM emissions from both light-duty and heavy-duty vehicles. However, the algorithms for PM are much simpler than those used for other emissions. For example, there is no separation of cold start from other operating modes, no effect of ambient temperature, and no consideration of the vehicle's accumulated mileage. This is largely because there is little data on which these effects can be based. Despite these limitations, and those discussed above regarding tire and brake emissions, it is worth while examining the model predictions.

The PART5 estimated in-use, light-duty vehicle fleet average PM-10 emissions using the default fleet age distribution for 1995 are 14 mg/mi for exhaust, 13 mg/mi for brake wear, 8 mg/mi for tire wear, and 36 mg/mi for fugitive emissions, for a total emission rate of 57 mg/mi. PART5 projects no change in brake or tire wear in the future. Exhaust and fugitive PM are predicted to

decrease to 12 and 35 mg/mi, respectively by the year 2010 (Rykowski, et al, 1996).

Exhaust PM emissions from different classes of vehicles can be compared as well. For example, the EMFAC7G model predicts Southern California PM-10 emissions of 1.65, 0.67, 0.84, and 15.8 tons/day from light-duty gasoline, medium-duty gasoline, light-duty diesel and medium- and heavy-duty diesels, respectively, for the year 1995. These change to 1.45, 0.19, 0.12, and 7.48 tons/day for the year 2005, reflecting a major improvement from the diesel vehicles. Finally, the exhaust emissions can be combined with the brake and tire wear emissions to give the comparison shown in Figure 1. This figure shows that tire and brake wear emissions dominate the PM-10 from light-duty vehicles in the year 2005 and that together these emissions are similar to those from the diesel trucks. It should be emphasized, however, that there are major uncertainties in all of the model projections. Given the model uncertainties, it is best to examine recent studies of PM emissions from light-duty vehicles.

RECENT LIGHT-DUTY VEHICLE EXHAUST PM STUDIES

Environmental Research Consortium Study. The Environmental Research Consortium (ERC) project was conducted by Chrysler, Ford Motor Co., and General Motors Corporation. The purpose was to determine exhaust PM emission rates from late-model, high-production-volume gasoline vehicles at both low and high mileage. Vehicles were tested on chassis dynamometers using the Federal Test Procedure (FTP) Urban Dynamometer Driving Schedule (UDDS). Filter samples of the particulate were collected for each of the three FTP phases. The vehicles were operated on California Phase II fuel. Preliminary work indicated that it was necessary to collect PM on the same set of filters over four FTP tests in order to obtain enough mass to weight reliably. Results of the study are summarized in Table 1.

When examining the results in Table 1 it should be noted that the emission rates for HC, CO, and NO_x are given in grams per mile while those for PM are in milligrams per mile. All of the vehicles were operating properly and met the applicable emission standards, as evidenced by the average regulated emission rates given in the table. PM emission rates were very low for both the cars and trucks, and increased only slightly with mileage. The average PM emission rate for these vehicles, approximately 1 mg/mi, can be compared to the average rate of 14 mg/mi predicted by PART5. One difference is that these vehicles were operated on a low sulfur fuel, while the PART5 prediction is based on a national average fuel. This, however, is not expected to have a large impact. These results suggest that the current model greatly overestimates PM exhaust emission rates for properly-functioning, light-duty gasoline vehicles in future years.

Off-cycle and In-use PM Emission Rates: Two additional factors need to be considered when estimating the light-duty vehicle PM emission rate. The first is that not all driving occurs under FTP conditions. Thus, studies need to be conducted that determine the effect of other driving cycles, ambient temperature, altitude, and fuel on PM emission rates. Currently, no data are available for PM emission rates on high-speed, high-load driving cycles such as the US06. Studies are underway to address this deficiency. The next study discussed has some information on the effects of fuel and temperature, and the Coordinating Research Council study discussed

Table 1. Summary of Results from the ERC Study

Vehicle	Number	Mileage	HC, g/mi	CO, g/mi	NOx, g/mi	PM, mg/mi
Car	6	7,000	0.13	1.28	0.18	0.60
Car	3	93,000	0.19	1.75	0.43	0.81
Truck	7	9,500	0.18	1.91	0.27	1.22
Truck	3	95,000	0.31	2.86	0.38	1.60

later is generating data at low temperature at altitude. The second factor that must be considered is the effect of vehicle condition on PM emission rates. Studies conducted over the last ten years have demonstrated that in-use vehicle emissions from the on-road fleet are dominated by the emissions from a small percentage of vehicles that are poorly maintained. The situation is expected to be the same for PM emission, since casual observation confirms that there are still a few gasoline vehicles in the in-use fleet that emit visible smoke during operation. Thus, to determine the in-use contribution of gasoline vehicles to the PM inventory, it is necessary to measure emissions from a representative sample of the in-use vehicle fleet.

Effect of Ambient Temperature and E-10 Fuel. During the winter of 1994-95, the State of Alaska Department of Environmental Conservation conducted a study in Fairbanks to evaluate the impact of switching from regular grade gasoline to an E-10 fuel (gasoline with 10% ethanol). The U.S. EPA and the GM R&D Center were participants in that study. As part of that program, 10 vehicles were tested on a chassis dynamometer at temperatures of 20°, 0°, and -20° F. Three of the vehicles were retested in the EPA facilities at Research Triangle Park, NC, under the same conditions and at the standard FTP temperature of 75° F. Vehicles were tested using the FTP and PM-10 emission rates were determined. Results have been published (Mulawa et al, 1997). Figure 2 shows the results for the three vehicles tested at EPA.

The three vehicles in Figure 2 were a 1994 Ford Aspire, a 1989 Chevrolet Celebrity, and a 1987 Plymouth Voyager. PM emission rates at 75 °F on gasoline were similar to those discussed above, with the Aspire, Celebrity, and Voyager having PM emission rates of 1.2, 1.6, and 7.5 mg/mi, respectively. PM emission rates clearly increase with decreasing temperature. Thus, at -20° F, the PM emission rates were 24, 45, and 64 mg/mi., respectively. The one exception was that the PM emission rate for the Aspire did not increase between 75 and 20° F. This may be because this vehicle was designed to meet the low temperature CO emission standard, whereas none of the other vehicles were designed to this standard. The increase in PM emission rate with decreasing temperature was primarily a cold-start effect. For these three vehicles, the average emission rates for the three phases of the FTP at -20° F were 203, 0.5, and 7.4 mg/mi., respectively. Figure 2 also shows that the use of E-10 fuel reduced the PM emission rate in most instances. It is likely that this is due to an increase in A/F ratio during the cold start portion of the FTP. Overall, the results demonstrate that modeling PM emissions from light-duty vehicles should separate cold-start emissions from hot stabilized emissions, and that the impact of oxygenated fuels should be considered as well.

CAWRS Study:. The Clark and Washoe Remote Sensing Study (CAWRSS) was conducted in 1994 in the Clark and Washoe Counties (Reno and Las Vegas) of Nevada. Participants were the GM R&D Center, the Desert Research Institute, and the U.S. EPA. The primary purpose of the study was to determine the emission rates of HC and CO from the local in-use light-duty vehicle fleet. Vehicles were screened by infrared remote sensing and high-emitting vehicles were pulled over and asked to volunteer for a roadside I/M test. The remote sensing selection criteria were approximately 4% CO or 0.3% HC. The U.S. EPA operated a portable chassis dynamometer at the site that was equipped with a dilution tunnel. This setup afforded an opportunity to conduct a screening program to determine if in-use vehicles had significant PM emission rates. A dual PM-10 sampling system was added to the dilution tunnel for this purpose. The system permitted the simultaneous collection of a Teflon filter for mass determination and a quartz filter backed up by a PUF-XAD trap. The quartz filter was used to determine the organic and elemental carbon content of the PM as well as the non-volatile PAH (polyaromatic hydrocarbon) compounds. The PUF-XAD trap was analyzed for the volatile PAH compounds. The purpose of examining the PAH compounds was primarily to develop a light-duty gasoline PM fingerprint that could be used in source-apportionment studies. Results of the study have been published (Sagebiel, et al., 1997).

The 23 recruited vehicles had an average age of 10.5 years and an estimated average odometer reading of 99,800 miles. The average regulated

emission rates were 3.5, 45.0, and 1.72 g/mi for HC, CO, and NO_x, respectively. The average PM-10 emission rate was 183 mg/mi. This average rate is biased high by the specific recruitment of 5 vehicles that were observed to have visible smoke emissions. The average PM-10 emission rate for these five vehicles was 558 mg/mi. The average PM-10 emission rate of the remaining 18 vehicles was 51 mg/mi. Analysis of the filter samples indicated that most of the PM was carbonaceous material. The organic carbon content of the carbonaceous material was 77%, on average, with a wide range (35 - 95%). Sulfate emissions were also measured and were found to be very low, averaging 0.12 mg/mi, with one high emitter omitted. Twenty-four PAH compounds were measured and it was determined that there were significant differences in the PAH profiles between the smoking and other vehicles. Overall, it was concluded that there is a set of in-use, on-road vehicles with relatively high PM emission rates, and that further studies would be needed to characterize these vehicles further.

Orange County Study: An opportunity to obtain additional information on in-use vehicle PM emission rates occurred in 1995 with a South Coast Air Quality Management District sponsored study (Program for the Use of Remote Sensing Devices to Detect High-Emitting Vehicles). Participants were the Environmental Research Consortium, the U.S. EPA, and the Desert Research Institute. The study was similar to the CAWRS Study in that remote sensing was used to identify high emitting vehicles which were subsequently pulled over and tested on a chassis dynamometer using the IM240 driving cycle. An additional aspect to this program was that vehicles were repaired after the initial test and then returned to the roadside site for a retest to determine repair effectiveness.

PM-10 IM240 emission rates were determined on 103 high-emitting vehicles. Filter samples were analyzed for organic and elemental carbon and trace elements. Figure 3 shows a histogram of the pre-repair PM-10 emission rates. Each bar gives the number of vehicles within the emission rate bin. PM-10 emission rate bins start with a range of 0 to 0.049 g/mi and increase to the last bin which is all vehicles with a rate greater than 0.5 g/mi. The mean PM-10 emission rate was 138 mg/mi. Vehicles that had been identified as having visible smoke emissions averaged 395 mg/mi, while those without visible smoke average 96 mg/mi. The fleet of vehicles tested was older than those in Nevada, averaging 12.3 years old, and had an average estimated odometer reading of 125,564 miles.

Vehicles were retested after repair. While HC and CO were greatly reduced, the average PM emissions were approximately the same. This result, however, is confounded by preconditioning effects. During the repair, the vehicles experienced extended idle time. They were not reconditioned for test upon their return, and it is likely that PM emission rates were elevated. In this regard, when 10 vehicles were given back-to-back IM240 tests after recruitment, nine had significantly lower PM-10 emission rates on the second test. The average decrease was 32%. It was concluded that further work is needed to study preconditioning effects.

Analysis of the filter samples showed, again, that most of the PM was carbonaceous material. The carbonaceous material was 71% organic carbon, on average. The organic content ranged from 19 to 97%. Elements were determined by X-ray fluorescence analysis. Fourteen elements were routinely detected above the detection limit. These were, sulfur, lead, zinc, phosphorous, calcium, magnesium, copper, iron, aluminum, silicon, chlorine, and bromine. Altogether, they comprised 3.5% of the PM-10 mass. Their mass contribution ranged from 0.4% to 12.3%, being highest for the lower emitting vehicles. Oil consumption was clearly a major source of many of these elements.

Overall, it was concluded on the basis of this study that there is a population of older, high-mileage vehicles in the current in-use fleet whose PM-10 emission rates are higher than those of new vehicles. Since the vehicles recruited in this study and the CAWRS Study were not randomly recruited, the data can not be used to estimate the in-use fleet emission rate. Therefore, it was recommended that a study be conducted that measures PM emission rates from a random selection of vehicles. Further, it was recommended that the study conduct the

tests using the FTP so that the effect of cold start versus other operating modes could be investigated. The Coordinating Research Council has undertaken the recommended study.

Coordinating Research Council Study: The Coordinating Research Council (CRC) Project E-24, Measurement of Exhaust Particulate Matter Emission from In-Use Light-Duty Motor Vehicles, is being performed by contractors at three locations: Denver, Colorado; Riverside, California; and San Antonio, Texas. At all three locations in-use, light-duty gasoline and diesel vehicles are recruited for FTP emission testing. Vehicles are tested as received on a chassis dynamometer and PM-10 emission rates are measured along with the regulated pollutants. In addition, filter samples and PUF/XAD cartridges are collected for subsequent analysis for organic and elemental carbon, elemental composition, sulfate and nitrate, PAHs, and hopanes and steranes.

The Denver portion of the study is sponsored by the CRC, General Motors Corp., the National Renewable Energy Laboratory (NREL), and Total Petroleum Co. The contractors are the Colorado Department of Public Health and Environment (CDPHE), General Motors Corp., U.S. EPA, and Colorado State University. This study measured emissions from 174 light-duty gasoline vehicles and 22 light-duty diesel vehicles. The study was conducted in two segments, one during the summer of 1996 and one during the winter of 1996/1997. During the winter portion of the study, vehicles were tested both indoors on the CDPHE chassis dynamometer and outdoors on the EPA portable dynamometer. The Riverside portion of the study is being conducted by the CE-CERT which is part of the University of California at Riverside. Sponsors are the CRC, the South Coast Air Quality Management District, and NREL. They will test 100 light-duty gasoline vehicles and 19 light-duty diesel vehicles. The San Antonio study is being conducted by the Southwest Research Institute under CRC, NREL, and the Texas Natural Resources Conservation Commission sponsorship. They will test 60 light-duty gasoline vehicles and 8 light-duty diesel vehicles. All programs are expected to be completed by the end of 1997.

At this time, preliminary data is available from the Denver study only.

Table 2 gives the number of vehicles tested during both the summer and winter segments of the Denver program. Vehicle selection was weighted more heavily towards the older vehicles in anticipation that they would have higher PM emission rates than the newer vehicles, and thus would be more important in determining the total in-use vehicle exhaust emissions PM inventory. The vehicles were binned into model year categories based on emissions-control technology. Thus, 1981-85 vehicles are predominantly the first generation of vehicles with closed-loop catalyst systems. The smoking vehicles are vehicles that were specifically recruited because they had been observed to emit visible smoke. Their average age in the summer and winter were 13.8 and 17.4 years, respectively. The diesels covered a broad range of model years. Their average ages in the summer and winter were 7.3 and 15.2 years, respectively. The average age was older during the winter because most late-model diesels are in large trucks which could not be tested on the EPA chassis dynamometer.

Table 2. Vehicles Tested in the Denver Program

Vehicle Category	Summer	Winter
Pre-1981	25	17
1981-1985	26	16
1986-1990	22	14
1991-1996	20	10
Smokers	9	15
LD Diesels	10	12

Table 3 gives the average PM emission rates for the summer as well as the range in PM emission rates by vehicle category. Results should be considered preliminary at this time since data analysis for the program is not complete. As anticipated the average rate for the older vehicles is much higher than that for the new vehicles. The range in emission rates is very large, with some of the older vehicles having low PM emission rates and some of the pre-1991 vehicles having high rates. No post-1990 model

Table 3. PM-10 Emission Rates for the Denver Summer Segment

Vehicle Category	Average, mg/mi	Maximum, mg/mi	Minimum, mg/mi
Pre-1981	95.5	278	7.0
1981-1985	47.4	271	1.1
1986-1990	44.4	442	0.8
1991-1996	2.8	9.6	1.1
Smokers	225	551	39
LD Diesels	811	3988	113

year gasoline vehicle had a PM emission rate greater than 10 mg/mi.

Table 4 gives the average PM emission rates measured indoor and outdoors during the winter segment of the Denver study. PM emission rates indoors during the winter averaged lower than those observed during the summer for vehicles in the same categories. The actual vehicles tested were not the same. This may be due to oxygenated fuel use, which is mandated for the winter in Denver, but not for the summer. The outdoor emission rates were higher than indoors due to increased PM emissions during the cold-start portion of the FTP. Overall, the emission rates are in agreement those seen in the other studies.

CONCLUSIONS

Changes in fuel composition and vehicle technology have greatly reduced the PM exhaust emission rate from light-duty vehicles over the last 20 years. However, there remains a population of older, in-use vehicles with elevated PM exhaust emission rates. Fleet turnover and smoking vehicle regulations should reduce this population of high PM emitters in the future. Current mobile PM source emissions factor models need to be updated with new information on the performance of current in-use vehicles taking into account factors such as vehicle age, driving mode, and fuel use. Results of the studies discussed above will help in that regard.

Table 4. PM-10 Emission Rates for the Denver Winter Segment

Vehicle Category	Outdoors	Indoors
Pre-1981[2]	78.3	54.2
1981-1985	46.7	35.9
1986-1990	28.3	11.4
1991-1996[2]	24.9 (12.6)[1]	3.4
Smokers	434	380
LD Diesels	487	460

[1]one high emitter removed from the average
[2]not all matching vehicles between outdoors and indoors

The source profiles generated in this study will be used in the Northern Front Range Air Quality Study to make a top-down estimate of the contribution of light-duty vehicles to the ambient PM burden. In addition, the data will be used to modify the PM emission estimates in PART5 to generate an improved bottoms-up inventory of the light-duty PM emissions.

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Figure 1. EMFAC7G Estimates of On-Road PM-10 Emissions for 2005

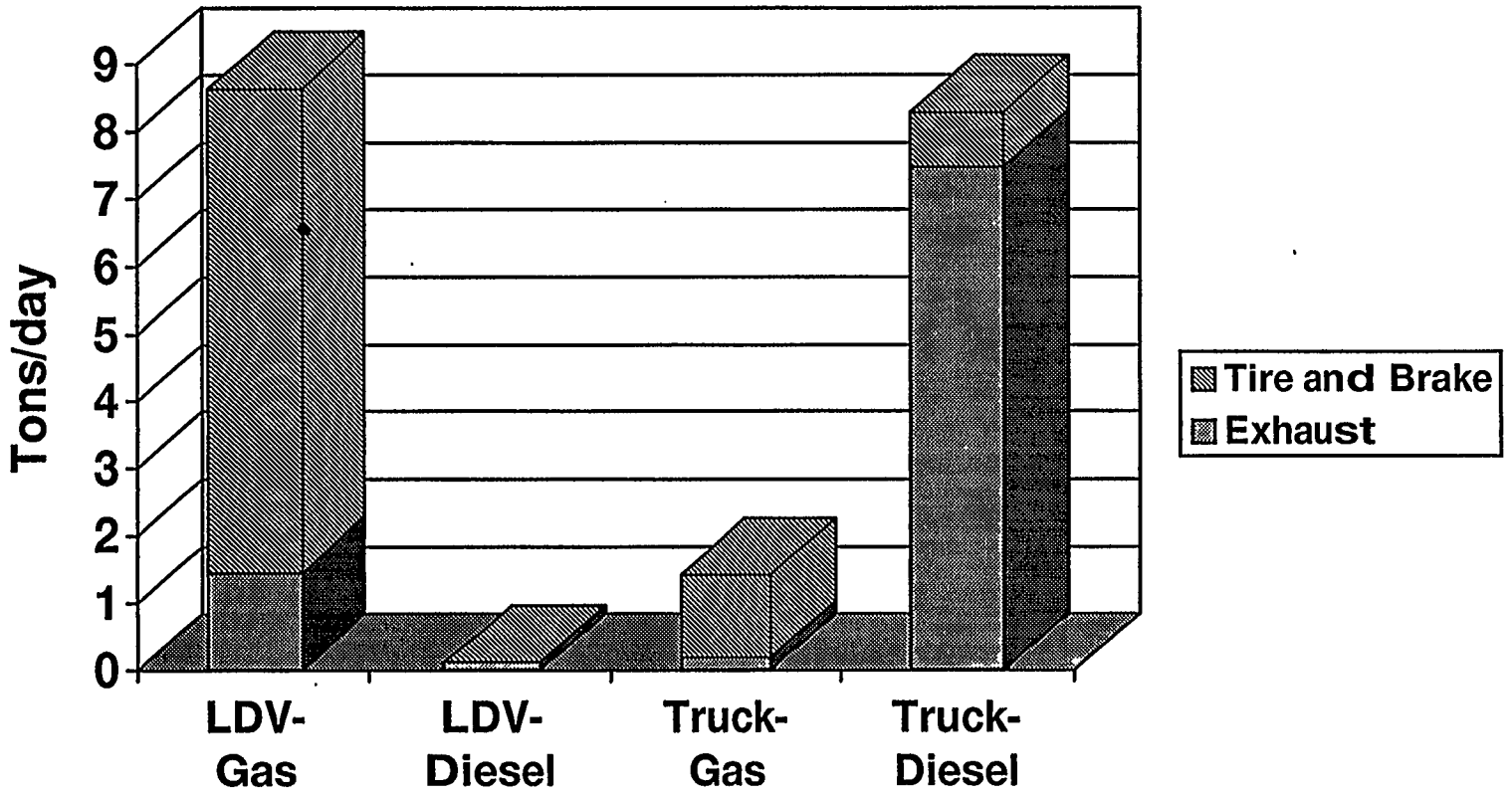

Figure 2. Effect of Ambient Temperature and E-10 Fuel on PM Emission Rates (mg/mi)

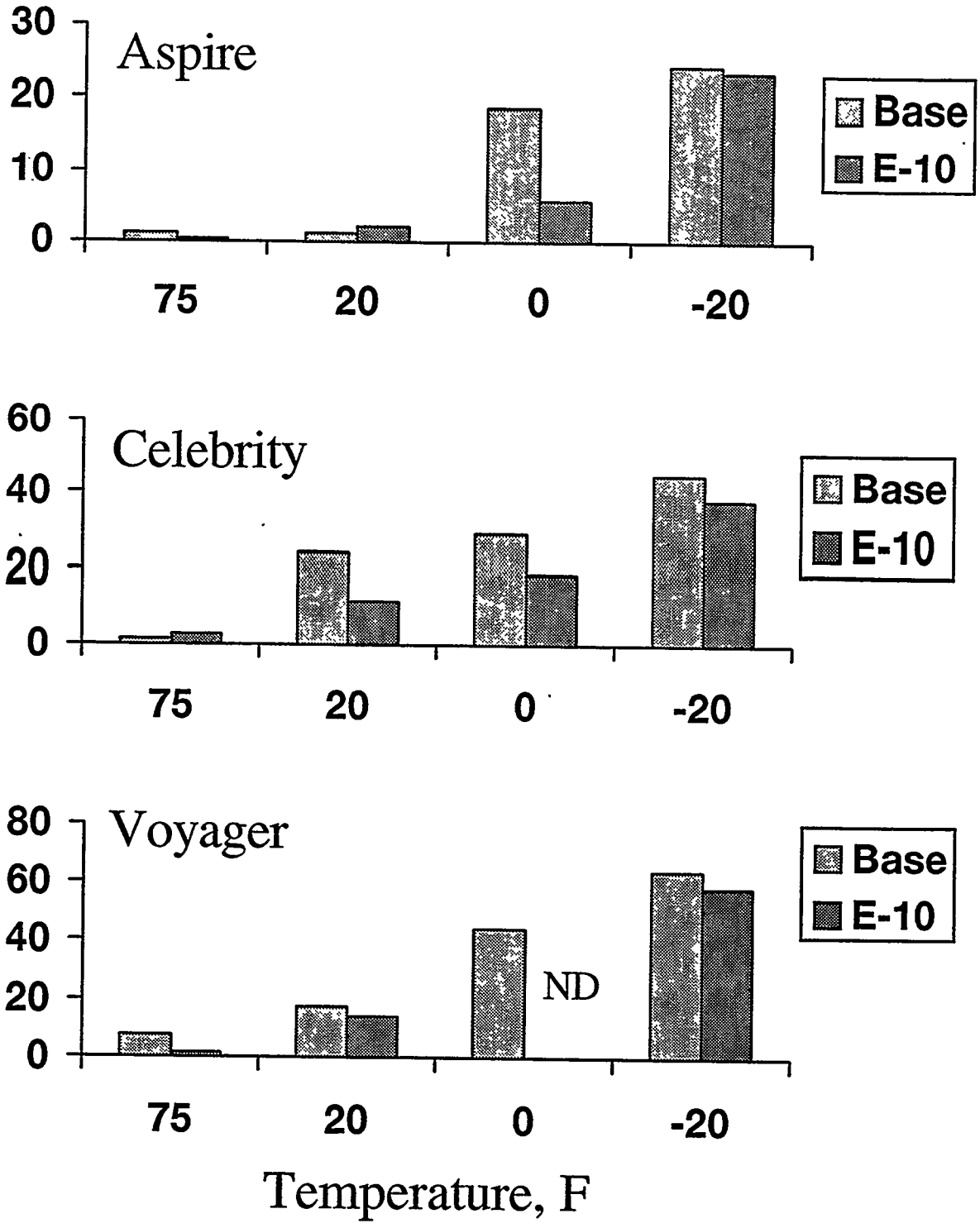

Figure 3. IM240 PM-10 Emission Rates From In-Use, High HC and/or CO Emitting Vehicles