Methods of Characterizing the Distribution of Exhaust Emissions from Light-Duty, Gasoline-Powered Motor Vehicles in the U.S. Fleet

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ABSTRACT

Mobile sources significantly contribute to ambient concentrations of airborne particulate matter (PM). Source apportionment studies for PM_{10} and $PM_{2.5}$ indicate that mobile sources can be responsible for over half of the ambient PM measured in an urban area. Recent source

apportionment studies attempted to differentiate between contributions from gasoline and diesel motor vehicle combustion. Several source apportionment studies conducted in the U.S. suggested that gasoline combustion from mobile sources contributed more to ambient PM than diesel combustion. However, existing emission inventories for the U.S. indicated diesels contribute more than gasoline vehicles to ambient PM concentrations.

A comprehensive testing program was initiated in the Kansas City metropolitan area to measure PM emissions in the light-duty, gasoline-powered on-road mobile source fleet to provide data for PM inventory and emissions modeling. The vehicle recruitment design produced a sample that could represent the regional fleet, and by extension, the national fleet. All vehicles were recruited from a stratified sample based on vehicle class (car, truck) and model-year group. The pool of available vehicles was drawn primarily from a sample of vehicle owners designed to represent selected demographic and geographic characteristics of the Kansas City population. Emissions testing utilized a portable, light-duty chassis dynamometer with vehicles tested using the LA-92 driving cycle, on-board emissions measurement systems, and remote sensing devices. Particulate mass emissions were the focus of the study, with continuous and integrated samples collected. In addition, sample analyses included criteria gases (CO, CO2, NO/NO2, HC), air toxics (speciated VOCs) and PM constituents (elemental/organic carbon, metals, semi-volatile organic compounds). Results indicated that PM emissions from the in-use fleet varied by up to three orders of magnitude, with emissions generally increasing for older model year vehicles. The study also identified a strong influence of ambient temperature on vehicle PM mass emissions, with rates increasing with decreasing temperatures.

IMPLICATIONS

Motor vehicles significantly contribute to local and national emissions of a number of air pollutants that can lead to adverse health effects. This study integrated random vehicle recruitment techniques, portable dynamometer emissions testing, on-board emissions measurements, and particulate matter and gaseous pollutant sample collection and analysis to represent the distribution of emissions in the U.S. fleet. This paper provides a detailed description of the methods implemented in one of the largest motor vehicle field emissions testing projects ever conducted. Results from this study will be used in broad environmental applications including emissions model evaluation and improvement, air quality assessments,

and health effect evaluations through improved understanding of light-duty, motor vehicle emissions. Understanding the factors contributing to elevated pollutant concentrations will improve policies for protecting public health through improved voluntary, regulatory and planning actions.

INTRODUCTION

Airborne particulate matter (PM) has been linked to numerous adverse effects including health impairment, ecological damage, decline in visibility, and climate change. Mobile sources significantly contribute to ambient concentrations of PM. Recent source apportionment studies have attempted to differentiate motor vehicle contributions, including identifying impacts from gasoline and diesel combustion. These studies have given conflicting results. Some studies indicated that gasoline combustion from mobile sources contributed more to ambient PM than diesel combustion.[1,2] However, other studies suggested that diesel combustion contributed more than gasoline combustion to ambient PM.[3,4,5] Existing U.S. emission inventories also suggest diesels contribute more than gasoline vehicles to ambient PM concentrations.

PM emitted from gasoline-powered motor vehicles generally consists of a mixture of organic carbon, elemental carbon, and small quantities of trace metals and sulfates; however, exhaust emissions of PM from gasoline-powered motor vehicles and diesel powered vehicles have changed significantly over the years.[6,7,8,9] These changes have resulted from reformulation of fuels, the wide application of exhaust gas treatment in gasoline-powered motor vehicles, and changes in engine design and operation. Because of these evolving tailpipe emissions, along with the wide variability of emissions between vehicles of the same class, well-defined average emissions profiles for the major classes of motor vehicles have not been established.[10,11,12,13]

The distribution of emissions among light-duty motor vehicles is also not well understood. Older, poorly maintained gasoline vehicles could be significant sources of $PM_{2.5}$ mass,[1,12] in addition to being significant and disproportionate sources of gaseous pollutants.[14] Durbin noted that light-duty, gasoline vehicles with visible smoke emitted from the tailpipe constituted only 1.1 to 1.7% of the light-duty fleet in the South Coast Air Quality Management District in California, yet these vehicles contributed roughly 20% of the total PM emissions from the light-duty fleet.[15]

Due to the varying PM emissions estimates for gasoline-powered motor vehicles, whether from source apportionment studies or emission inventory projections, and the potentially significant contributions of this emission source to ambient PM concentrations, the Kansas City Light-Duty Vehicle Emission Study (KCLDVS) was initiated. The study provided a representative sample of light-duty, gasoline-powered motor vehicles to estimate the distribution of PM emissions in the U.S. light-duty fleet. The project also provided the opportunity to investigate emissions of other regulated and toxic pollutants.

METHODS

Discussions of the methods used in the study are divided into the major components of study: vehicle recruitment, emissions monitoring techniques, and sample analyses.

Vehicle Recruitment

A major obstacle in previous emissions testing studies has been identifying how the vehicles tested represent the corresponding fleets, and how the vehicles' emissions represent the distribution of national mobile-source emissions. This study was specifically designed to allow emissions information from the vehicles tested to be projected back onto the overall Kansas City Metropolitan Area vehicle fleet. At the time of the study, Kansas City was the largest metropolitan area in the United States without an Inspection/Maintenance (I/M) program. Accordingly, this study was the first large-scale study of its kind which utilized a weighted stratified random sampling scheme to represent the distribution of vehicle PM emissions in a large population.

The recruitment process entailed deriving a targeted, stratified sample of vehicles from a cohort of households drawn from the recently completed 2004 Kansas City Travel Behavior Survey[16] conducted for the Mid-America Regional Council (MARC), the planning organization for the Kansas City metropolitan area. This survey was conducted in Spring 2004 using a Random Digit Dialing (RDD) sampling method . For this survey, 5,500 KCMSA regional households were randomly sampled and contacted. Of these, 4,001 agreed to participate in the Travel Behavior Survey and 3,049 completed all aspects of the survey. Non-respondents in the 2004 MARC Survey were those 1,500 households that were contacted and firmly refused to participate. Most refusals took place during the introduction to the study, prior to the interviewer

obtaining any demographic information about the household. Information pertaining to the characteristics of those 1,500 households that chose not to participate is very limited. The only information that could be reviewed was the geographic distribution of refusers, since all sampled telephone numbers were initially flagged with the anticipated county of residence. This review showed the proportion of refusals matched the proportion of participants by county of residence.

In addition to demographic data on households, the MARC Survey provided information on vehicles owned by households including make, model, model year, body type, and fuel type for each vehicle as well as home address and preferred method for contacting the participant. Use of the survey also helped ensure non-registered vehicles were available for recruitment throughout the study.

In order to ensure the MARC Survey data satisfactorily represented the Kansas City MSA population, a comparison of this data was made with Census 2000 data at the household and person levels using a number of demographic and geographic characteristics including household size, number of vehicles in the household, income, type of residency, ethnicity, respondent age, and geographic distribution. The comparisons showed that the MARC Survey data represented the Census 2000 data for the Kansas City MSA population at the household and person levels using relevant demographic and geographic characteristics.

A challenge during the first round of testing was fewer than expected older vehicles available for recruitment. By the end of Round 1 (summer), the available pool for recruiting the oldest vehicles (Strata 1, 2, 5, 6 in Table 1) had been virtually exhausted. This posed a challenge for Round 2 (winter) testing. In order to enhance vehicle recruitment in these older strata, the Kansas and Missouri KCMSA Vehicle Registration databases were used in order to gain access to a larger pool of vehicles available for recruitment. Those databases were used to draw representative stratified random samples for recruiting as many vehicles as necessary to achieve the desired sampling targets. The use of the vehicle registration databases did not conflict with the use of the MARC Survey; these databases triggered the adoption of an efficient dual frame sample design. The adoption of a dual frame design provided several benefits including (1) the registration database frame was complete, with virtually 100% coverage of the vehicle fleet population; (2) the efficiency of identifying rare or low prevalence vehicles (e.g., older trucks)

from the registration database was considerable relative to the alternative of large scale screening of households; and (3) use of the registration database served to reduce any potential influence created by the 1,500 refusals from the original 2004 MARC Survey used as the cohort for this study.

In order to understand the influence of non-respondents to the overall sample, a non-response study was conducted in Round 1 of the study. Of the 261 households that ultimately had their vehicles tested in Round 1, 23 had initially refused to participate during the recruitment call, but converted after another focused attempt with increased incentives. An additional 29 households cancelled their initial scheduled testing, but agreed again to have the vehicle tested later during Round 1. Using these refusal conversions, a comparison between Round 1 participants and those that refused testing in terms of the county of residence, income, and vehicles owned revealed little difference in the proportions of refusers and first-time participants by county of residence. The refusers were more likely to report a lower income than the first-time participants (22% compared to 16%, respectively). The refusers were more likely to own a truck and an older vehicle.

After completion of the field study, the characteristics of the participants from both rounds of the study were compared with both the MARC Survey and the Census 2000 data for the study area. Overall, the weighted data of the study participants compared favorably with the census data, indicating that the survey data set represented the regional population. However, a difference was identified in household residence type (a higher recruitment rate was seen for single family residence as opposed to "all other types", such as apartment rentals). This was likely due to listed telephone numbers (those with complete address information for the household) being associated with households of longer tenure, which is correlated with living in a single-family dwelling and home ownership. Renters, who are considered to be more transient and living in housing types not characterized as single-family dwellings, may change telephone numbers more often and are typically more likely to have a number that is incomplete or not included in the listed telephone number database. An effort more focused on renters would have required the use of more unlisted than listed numbers, which was not possible within the project's budget. Thus, the desire to achieve a good mix of residence type was balanced with the

project budget and as a result, residence type came within 10% of the census parameters, but not within 5% as with the other variables.

As shown in Table 1, recruitment strata were based on vehicle type (car vs. truck) and model-year group. The model-year groups chosen represented changes in technology based on model years (pre-1981 carburetors; 1981-1990 spark-ignition injectors; 1991-1995 phase-in Tier-1 emission standards; 1996-present (2005) Tier-1 and NLEV emission standards). The sample stratification and allocation targets identified for this study were based on the variability of PM emissions measured from prior studies[i] and DMV registration data. Several factors were used to determine the optimal allocation of test vehicles across the eight sampling strata, including PM emission rates from previous studies, annual PM emission estimates using the MARC Survey vehicle distributions and MOBILE6 emission rates, and proportional emissions in each strata weighted by average PM emission rates to account for occasional high emitting vehicles within each stratum. Table 1 presents the sample allocation used in the KCLDV project, and the distribution of vehicle test targets vs. actual vehicles tested in both rounds of the study. Tables 2 and 3 show the demographic and geographic distributions, respectively, for study participants.

Vehicle Emissions Testing

This study incorporated multiple emission testing techniques for each vehicle to capture a wide range of emissions information. Real-time gaseous and PM emissions measurements were collected using a chassis dynamometer operating over an established driving cycle. Integrated gaseous and PM emissions measurements were also collected over this same driving cycle for pollutants without real-time techniques or as quality assurance comparisons with the real-time devices. On-board, portable emissions measurement systems (PEMS) collected "real-world" mass-based gaseous vehicle emissions from the vehicle's exhaust while driving on pre-established routes in Kansas City prior to each dynamometer test. A subset of test vehicles were also outfitted with PEMS, collecting tailpipe emissions data while the vehicle owners drove their regular routes for a period of several days. Finally, a subset of test vehicles also had select gaseous emissions measurements collected by driving past Remote Sensing Devices (RSD) as part of the pre-established driving route. The location of these RSD deployments also provided on-road fleet emissions data for the Kansas City area. Testing occurred during two rounds: summer (Round 1) and winter (Round 2). Approximately ten percent of the vehicles recruited

participated during both rounds of the study. Emissions testing also included replicate sampling for each vehicle. Each recruited vehicle underwent an inspection, conditioning run, emissions testing, and quality assurance evaluation as described in the following sections.

Vehicle Inspection. Vehicles were inspected upon arrival at the test site to insure no safety concerns existed for testing on the dynamometer. Considerations for rejection of a vehicle included unsafe tires, inadequate brakes, excessive exhaust or fluid leaks, or oversized vehicles. A vehicle information data form detailed the pertinent specifications and condition of each recruited vehicle. Recorded information included test date, test number, vehicle license plate, make, model, model year, VIN, engine displacement, number of cylinders, emission controls, engine family and serial number, fuel type, fuel level and date of last refueling, and oil level and date of last oil change. Test inertia and horsepower (hp) settings were determined from computerized EPA Inspection/Maintenance (I/M) lookup tables. A visual and odor inspection of the exhaust was conducted, and oil and fuel samples were collected. Information was collected for each vehicle recruited, regardless of whether the vehicle was tested.

Vehicle Conditioning. Vehicles passing the safety inspection underwent exhaust system conditioning prior to testing. Conditioning reduced the potential for biasing results based on driving behavior prior to testing (e.g. removing carbon deposits in exhaust system due to urban, low speed driving). Conditioning consisted of driving the vehicle for approximately thirty-minutes along an established route in the vicinity of the testing facility. The conditioning route was chosen to represent the dynamometer testing cycle, and included multiple acceleration periods, extended freeway driving at speeds greater than 55 mph, and minimal idling periods. Mass-based PEMS emissions measurements were collected during all conditioning drives. The vehicles remained at the testing facility overnight after conditioning, and not started again until the next day's emission test.

Emissions Testing. Vehicle driving simulation occurred on a Clayton Model CTE-50-0 chassis dynamometer. The dynamometer can simulate a continuous spectrum of loads from 3 to 50 Hp at 50 mph and inertias from 1750 to 3000 pounds in 250 pound increments and 3000 to 5500 pounds in 500 pound increments. Cooling fluid for the dynamometer's water brake power

absorption unit consists of a 50/50 mixture of water and glycol. The fluid re-circulates and cools by a self-contained pumping and cooling system.

The portable chassis dynamometer was located inside a garage with large bay doors at each end of the building. These doors remained open at all times of the study. In addition, all building vents remained open during the study. Ambient and garage temperatures were checked daily to insure that indoor and ambient temperatures were equivalent during all testing and vehicle soak times.

Vehicles were operated over the LA92 Unified Driving Cycle. The LA92 cycle provided information regarding cold-start emissions, hot-stabilized operation emissions, and hot-start emissions that included low-speed/low-load and relatively high-speed/high-load operations. The LA-92 cycle used consisted of a cold start Phase 1, a hot-stabilized Phase 2, a ten-minute engine off warm soak, and a warm start Phase 3. In order to obtain a true cold-start test, vehicles were pushed onto the dynamometer after an overnight soak at ambient temperatures and not started until the beginning of the LA92 test. Figure 1 presents a speed trace for the LA92 driving cycle, and an example of the conditioning route speed trace for comparative purposes.

A positive displacement pump-constant volume sampling (PDP-CVS) system quantitatively diluted exhaust gas from the vehicle operating on the dynamometer for emissions measurements. The PDP-CVS system used an 8-inch diameter stainless steel dilution tunnel and a SutorBilt Model GAELAPA (6-LP) PDP operating at 500 cubic feet per minute (cfm). Dilution air passed through a charcoal bed (for hydrocarbon stabilization) followed by a high-efficiency (HEPA) filter for particle removal prior to mixing with vehicle exhaust. The tunnel operating temperature was maintained at approximately 125°F for all testing analyses. Propane injections to the CVS/dilution tunnel verified CVS flow. Triplicate injections were evaluated on three separate dates during the summer and winter testing rounds. Figure 2 shows a diagram of the dynamometer sampling system.

The CVS and tunnel dilution air heater were turned on at least 45 minutes prior to engine start and run to purge the exhaust transfer line and dilution tunnel. Pumps at the analytical bench were run to purge all sample lines. The CVS, tunnel heater, and sample pumps continuously ran throughout the day, only shutting down at the conclusion of testing. Emissions testing began when the temperature in the dilution tunnel became stable (no increase in temperature over a

three-minute period). For real-time gaseous emission measurements, multipoint calibrations were conducted at the beginning and end of each testing round.

Within two minutes of the start of the initial test of the day, background total hydrocarbon (THC), carbon monoxide (CO), nitrogen oxides (NO_x), and carbon dioxide (CO_2) concentrations in the dilution tunnel were recorded and referenced as background levels for the tests that immediately followed that day. If background concentrations for succeeding tests that day exceeded the reference background by more than fifteen percent, testing stopped until corrective measures could be implemented. During this study, testing was never interrupted due to elevated background levels.

PM filter collection to identify mass emission concentrations occurred separately for each phase of the LA92 driving cycle to minimize potential loss of the volatile fraction of PM during hot-stabilized operating conditions. PM integrated over each phase were collected from the dilution tunnel through a sampling train consisting of a probe, a particle size limiting cyclone, filter cassette, and flow control system (see Figure 3). In addition to the integrated filter measurement, a continuous exhaust aerosol mass measurement system (Quartz Crystal Microbalance (QCM), Booker Systems Ltd., Ann Arbor, MI) provided real-time measurements of exhaust PM mass concentrations. The sampling probe matched the flow velocity of the dilution tunnel to that of a 2.5 μm size-selective cyclone. The cyclone and filter cassette were constructed of stainless steel. The sampling train collected particles having a mass median diameter (MMD) of less than 2.5 μm. Flows were controlled with a mass flow controller at 16.7 lpm for PM samples collected on 47-mm Teflo™ (2.0 um pore size) Teflon membrane filters (Pall-Gelman, Ann Arbor, MI).

In addition to PM collection, real-time THC measurements used a Heated Flame Ionization Detector (HFID). Background THC levels were monitored with a second HFID. NO_x measurements used a chemiluminescent instrument. CO and CO_2 were analyzed with infrared (IR) instruments. A third IR instrument analyzed low (< 1000 ppm) CO concentrations. In addition to the real-time measurements, integrated Tedlar bag samples were collected for later comparison with integrated real-time measurements.

Independent second-by-second measurements of gaseous pollutants (THC, CO, CO_2, NO_x and O_2) were also collected in tandem using a SEMTECH-G PEMS and exhaust mass flow measurement device system (Sensors, Inc., Ann Arbor, MI). This provided a mass-based

comparison of gaseous pollutants over the LA92 cycle as measured by the dynamometer bench and CVS.

PDP and ambient temperatures, relative humidity and atmospheric pressure were measured at the test site. A digital optical encoder driven from the dynamometer's roller as part of the driver's aid system measured vehicle speed.

A Pentium class computer logged real time output signals from the regulated emissions instrumentation, meteorological devices, and speed sensors. The real-time system was controlled by a commercial software package to configure sampling rates, engineering conversion factors, and data storage modes for each sampling and control channel.

On-road mass-based measurements of gaseous pollutants (THC, CO, CO_2, NO_x and O_2) were collected using the PEMS on the conditioning route. This provided a comparison of the vehicle's on-road emissions with those measured as the vehicle underwent a simulated dynamometer test.

After the conditioning drive and LA92 dynamometer testing, vehicles were returned to their owners. Approximately 60 vehicle owners agreed to extended testing with PEMS units on-board. The vehicle owner was encouraged to drive and operate the vehicle normally, allowing activity, emissions, and fuel economy information to be gathered under "real-world" on-road driving conditions. The on-board units continued to operate until the battery supply depleted, typically obtaining data for 6 to 8 hours of operation.

Quality Assurance. Dynamometer calibration checks were performed daily through a combination of coast-downs and speed calibrations. PDP rpm was also checked daily. The dynamometer's torque sensor was calibrated after field set up using dead weight techniques. Tunnel blanks were collected to identify the potential for background pollutant concentration to affect tailpipe emission measurements.

Five percent of all vehicles tested in Rounds 1 and 2 were randomly selected for replicate emissions testing. After the initial test, the selected vehicles repeated a ten-minute engine off soak, a warm start Phase 3, and a stabilized Phase 4. If PM concentrations measured during the replicate test by the QCM were not within $\pm$15 percent of the initial Phase 3 and 4 results, a third test was conducted and reported. In addition to the randomly selected vehicles, any vehicle with

a QCM measured emission rate greater than 200 mg/mile during the initial phase 1 through 4 testing was required to complete a replicate phase 3 and 4 test.

Two round-robin tests were conducted between the portable dynamometer in Kansas City and the EPA National Vehicle and Fuel Emissions Laboratory (NVFEL) dynamometers in Ann Arbor, Michigan. Three EPA vehicles tested at NVFEL were shipped to Kansas City for testing on the portable dynamometer. Fuel canisters were also transported with the vehicles. After replicate testing in Kansas City, the vehicle returned to NVFEL for a third suite of tests. Round-robin testing revealed differences of less than five percent for all real-time emissions measurements collected in Kansas City and at NVFEL.

Regulated emission analysis instrumentation was zeroed and spanned before each test. Calibration gases consisting of a NO in Nitrogen mixture (90.2 ppm NO) and CO, CO_2, and propane in air mixture (900 ppm CO, 300 ppm propane, and 2.54 % CO2) were obtained. Cylinder concentrations were verified through comparison to NIST standards. Zero air and the FID fuel (60% H2/40% He) were obtained to specification. Zero air for real-time analyzers had a certification of < 0.5 ppm CO, < 1 ppm CO_2, and < 0.1 ppm HC. After arrival in the field, all real-time emissions analyzers underwent multipoint calibrations to confirm linearity.

Over 40 vehicles tested during Round 1 of the program were re-tested during Round 2 to determine comparability between testing events, most notably effects of ambient temperature on vehicle emissions. Table 4 shows strata goals for these retests.

Chemical Analysis

Real-time and integrated gaseous and PM sample collection provided detailed information on motor vehicle emission factors, improved profiles for source apportionment, and data for emission trends assessments. Sample air for gaseous real-time and integrated speciation analysis came from the dilution tunnel. Sample air for PM analysis came from the dynamometer dilution tunnel via two isokinetic probes inserted prior to a 90-degree bend in the dilution tunnel. Heated conductive lines carried air from the probes to the continuous instruments. Insulated copper tubing carried sample air to the time-integrated samplers. Filter samples were collected during each phase of the LA92 driving cycle tests. Sample air was drawn from the CVS via ½" insulated copper tubing to a small heated stainless steel chamber. The sample air exited via a $PM_{2.5}$ cyclone in the chamber to a heated diffusing chamber approximately 50 cm tall, equipped

with a temperature and humidity probe. From this chamber, the sample air exited through two filter cartridges. Up to eight cartridges could be installed in the base of the diffusing chamber, allowing four successive pairs of filters to sample without changing cartridges. Flow rates for each filter were set to 56 liters per minute (lpm) using a single, oil-less pump. Prior to the start of phase 1, all samplers were leak checked and flow calibrated using an NIST-traceable flow meter (Gillibrator). Figure 3 illustrates the PM instrumentation sampling train, including the integrated measurements as well as the real-time measurements previously described.

Log sheets recorded the run number, start and stop time, elapsed time, initial and final flow rate, and any exceptional occurrences for each integrated sample. Bar coded stickers with unique media IDs tracked all media and corresponding log sheets. All media were packed in ice storage and shipped overnight to the analytical laboratory.

Continuous Measurements. Real-time instruments measured gaseous, $PM_{2.5}$ mass and elemental carbon (EC) concentrations. Each method provided information on the amount and composition of emissions from motor vehicles, as well as identified the need for replicate testing and proper dilution tunnel conditioning before each test. Continuous regulated gases measured included CO, CO2, NOx and THC as previously described. PM2.5 mass measurements were collected on a quartz crystal microbalance (QCM) and optical nepholometer (TSI Dustrak). The QCM monitors the accumulation of particles on a surface in real-time. A clean-air dilution system is used in conjunction with this instrument to reduce the dynamic range of the source aerosol concentration. The Dusttrak estimates the concentration of particulate mass by measuring the intensity of light scattered perpendicular to a laser beam directed through the air flow stream. A Photoacuoustic Elemental Carbon analyzer (DRI) was used to continuously measure EC concentrations. This instrument continuously measures the concentration of light-absorbing carbonaceous material (black carbon) in the airstream by the photoacoustic principle, in which the absorption of modulated light by particles results in thermal-acoustic pulses that can be detected by a highly-sensitive transducer and phase-locked amplifier.

Integrated Measurements. Integrated gas and PM sampling allowed for detailed chemical characterization of exhaust components. Bag samples were collected for the regulated gases as a

quality assurance check of the continuous analyzers. Vehicle exhaust samples were also collected in evacuated summa canisters for speciated analysis of volatile organic compounds (VOCs). Multiple filters collected PM2.5 samples for mass, elemental/organic compound (EC/OC), trace elements, and semi-volatile organic compound (SVOC) analysis.

Gravimetric Analysis. Pre-weighed Gelman polymethylpentane ringed, 2.0 um pore size, 47 mm diameter PTFE Teflon-membrane Teflo filters (#RPJ047) collected particles for measurement of gravimetric mass and elements (described in next section). The filters were equilibrated at a temperature of 20 ±5℃ and a relative humidity of 30±5% for a minimum of 24 hours prior to weighing. Weight measurements occurred on a microbalance with ±0.0001 mg sensitivity. Exposure to a polonium source for 30 seconds prior to the filter being placed on the balance pan neutralized any potential charges on each filter. The balance operator was also grounded during filter measurement. Pre- and post-weights, check weights, and re-weights were conducted and recorded for quality control management.

Elements Analysis. The Teflon-membrane filter samples also provided a substrate for elemental chemical analysis. Analysis was performed for the following elements: Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Y, Zr, Mo, Rh, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Pt, Au, Hg, Tl, Pb, and U on a subset of the vehicles tested using Inductively Coupled Plasma - Mass Spectrometry (ICP-MS).

A quality control standard and a replicate from a previous batch were analyzed with each set of 10 samples. When a quality control value differed from specifications by more than ±5% or when a replicate concentration differed from the original value (when values exceed 10 times the detection limits) by more than ±10%, the samples were re-analyzed. If further tests of standards showed the system calibration had changed by more than ±2%, the instrument was re-calibrated.

Elemental/Organic Carbon Analysis. Pallflex 47 mm diameter pre-fired quartz-fiber filters (#2500 QAT-UP) were used for water-soluble chloride, nitrate and sulfate and for organic and elemental carbon measurements. The thermal/optical reflectance (TOR) method measured organic (OC) and elemental (EC) carbon. The method is based on the principle that different

types of carbon-containing particles convert to gases under different temperature and oxidation conditions.

Fine Particulate/Semi-Volatile Organic Compounds. Organic compound samples were analyzed by gas chromatography/mass spectrometry (GC/MS). Calibration curves for the GC/MS quantification were made for the most abundant and characteristic ion peaks of the hopanes, steranes, PAH and other organic compounds of interest using the deuterated species most closely matched in volatility and retention characteristics as internal standards.

Samples were collected by a separate sampler for determination of particulate and semi-volatile organic compounds on Pallflex TX40HI20-WW 102 mm diameter Teflon-impregnated glass fiber (TIGF) filters followed by glass cartridges containing Aldrich Chemical Company, Inc. 20-60 mesh Amberliete XAD-4 (polystyrene-divinylbenzene) adsorbent resins at a flow rate of 112 lpm. The material collected on these media are removed by solvent extraction and analyzed at DRI by gas chromatography and mass spectrometry. A single filter and adsorbent pair were collected for each unified cycle, combining phases 1, 2 and 3. Sampling was suspended during the 10-minute soak period by turning off the pump. Sample air was drawn from the dynamometer CVS via ½" insulated copper tubing to a small heated stainless steel chamber. As previously described, the sample air exited via a $PM_{2.5}$ cyclone to a heated diffusing chamber. From this chamber, the sample air exited via the filter followed by the XAD cartridge.

Volatile Organic Compounds. volatile organic compounds (VOCs) were collected in summa canisters and analyzed by GC/MS. VOCs of interest included benzene, formaldehyde, acetaldehyde, 1,3-butadiene, acrolein, toluene, ethylbenzene, xylene, and styrene.

For VOCs, sample air was drawn from heated cyclone chamber via a ¼" diameter Teflon hose and passed through a Teflon filter and a cobalt oxide coated denuder coated to remove NOx before being pumped into a Summa polished steel canister. Air flow was controlled by a needle valve to obtain the necessary flow rate to fill the canisters to approximately 15"Hg positive pressure over the duration of the complete unified cycle. Sampling was interrupted during the 10-minute soak by switching to a bypass channel. The sampler draws a total flow of 2 lpm with only approximately 300 cubic centimeters per minute pumped into the canisters. Sampling was

suspended during the 10-minute soak by switching to an unused channel by a relay linked to transistor-to-transistor logic (TTL) line signals from the dynamometer control.

Aldehydes were collected on 2,4-dinitrophenylhydrazine (DNPH) cartridges using a 6-channel sampler with integrated pump and mass flow controller. Sample air was drawn from heated cyclone chamber via a ¼" diameter Teflon hose at 500 cc/min. A single cartridge was exposed for the duration of the 3 phases of the unified cycle. Sampling was suspended during the 10-minute soak by switching to an unused channel by a relay linked to TTL line signals from the dynamometer control.

DISCUSSION

The KCLDVS provided the opportunity to investigate motor vehicle emission rates from a variety of vehicle types, technologies, and model years operating over variable environmental temperature conditions. This section provides results of general trends in PM mass emission rates. Subsequent papers will explore additional study results including emission rates of gaseous compounds, speciated PM compound emissions, and the influence of specific environmental and vehicle parameters on emissions.

Figure 4 presents the distributions of PM emission rates for each vehicle evaluated in the study, binned by model year strata, for the summer Round 1 (a) and winter Round 2 (b) results. The box and whisker plots shown in this figure detail the twenty-five to seventy-five percentile emission rates by the edges of the box, the fifty percentile line within the box, and the whiskers representing the fifth and ninety-fifth percentiles. Note that the summer, pre-1981 trucks only had two vehicles tested; thus, a box and whisker plot could not be drawn. This figure indicates that emissions within each strata often varied by as much as two orders of magnitude. The figure also shows a general trend of reduced PM emissions for newer model year vehicles, with the exception of the pre-1981 truck strata. Comparing the summer and winter round distributions, this figure indicates that the vehicles tested during the colder winter temperature tests also generally experienced higher emission rates. Figure 5 displays the fleet distribution of PM emissions across all model years as a histogram and cumulative percentage plot for the summer Round 1 (a) and winter Round 2 (b) tests. This figure also suggests that the winter tests yielded higher PM emission rates than the summer tests, with a bimodal distribution evident for the winter vehicle fleet.

Figure 6 compares PM2.5 emission rates for each vehicle tested during the study based on the vehicle's model year. This figure also differentiates data points for vehicles whose owners initially agreed to participate (open diamond data points) and owners who initially refused to participate in the study (solid square data points) as described in the "Vehicle Recruitment" section. The figure confirms results demonstrated in the previous figures, with PM emission rates from all vehicles tested in the KCLDVS ranging approximately three orders of magnitude. In addition, older model year vehicles emitted higher levels of PM than the newer vehicles. When comparing emission rates from vehicles whose owners initially agreed to participate in the study to the emission rates from vehicles whose owners initially refused to participate, no statistically significant difference existed in the variability or magnitude of PM2.5 emissions. These results suggest that no sampling bias existed between vehicles from owners who initially participated and owners who initially refused to participate for this study; thus, suggesting that the vehicles tested in this study represented the Kansas City metropolitan area fleet.

Several factors contributed to the wide range of emission rates experienced by the vehicles tested. One factor of primary interest for this study was the influence of ambient temperature. Figure 7 compares $PM_{2.5}$ emission rates for the forty-three vehicles tested in both the summer and winter rounds of the study. For colder ambient temperatures, these vehicles generally emitted much higher and more variable levels of $PM_{2.5}$ mass. Quantification of the relationship of ambient temperature and $PM_{2.5}$ and other pollutant emission rates will be further evaluated in other papers.

SUMMARY

This paper summarized methods used in a field study to evaluate the distribution of PM and other regulated and toxic pollutants emitted from light-duty, gasoline powered vehicles in the U.S. fleet. This study utilized novel approaches in vehicle recruitment, dynamometer and on-board emissions characterization, and PM sample collection and analysis. Results from this study described the distribution of PM motor vehicle emissions in the studied fleet for both summer and winter temperature conditions. PM emissions varied by two orders of magnitude within each vehicle model year classifications, and by up to three orders of magnitude across the entire fleet and temperature testing conditions. The results also demonstrated that a vehicle's

model year affected PM emissions. PM emissions also generally increased with decreasing temperatures. Further analyses will be conducted and reported to quantify these effects on PM and other pollutant emissions. These results will be used in broad environmental applications including emissions model evaluation and improvement, air quality assessments, and health effect evaluations.

ACKNOWLEDGMENTS

The authors wish to thank the numerous individuals and organizations who assisted in the planning and implementation of this project. Kitty Walsh and Dave Plegge of the EPA were instrumental in all contract development and management aspects of this project. We also wish to thank Steve Cadle, Kevin Black, Doug Lawson, and Brent Bailey, who provided critical input on the design and implementation of this project through their representation of the Coordinating Research Council's Real World Group. The authors also wish to express thanks to numerous individuals from the Mid-America Regional Council, the National Association of Clean Air Agencyies, EPA's Region 7, and EPA's Office of Air Quality Planning and Standards for their assistance in site logistics, vehicle recruitment, and project funding.

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Table 1. Target and Actual Sample Sizes by Stratum

Stratum (*h*)	Vehicle Class	Model-Year Group	Target Sample size*	Actual Sample Summer	Actual Sample Winter
1	Truck	Pre 1981	30	2	9
2	Truck	1981-1990	50	21	29
3	Truck	1991-1995	50	18	31
4	Truck	1996 and newer	75	39	50
5	Car	Pre 1981	30	6	14
6	Car	1981-1990	100	49	36
7	Car	1991-1995	65	39	37
8	Car	1996 and newer	80	87	29
Total			**480**	**261**	**235**

* Half of each sample to be collected during the summer round, and the remaining half during the winter round.

Table 2. Demographic Comparison of RDD 2004 Survey of Households and Census 2000 Distributions

Demographic Characteristic	RDD Survey (n=4,001)	Census 2000
Household size		
1	26.8%	27.4%
2	33.3%	33.0%
3	16.0%	16.2%
4+	23.9%	23.4%
total	100.0%	100.0%
HH Vehicles		
0	5.8%	7.4%
1	32.9%	33.9%
2	42.7%	41.7%
3+	18.6%	17.0%
total	100.0%	100.0%
HH Income		
< 15k	9.9%	12.2%
15k - < 25k	10.2%	11.3%
25k - < 50k	30.2%	30.1%
50k - < 100k	35.9%	33.6%
100k +	13.8%	12.8%
(refusal)	(5.9%)	--
total	100.0%	100.0%
Residency Type		
single family	76.8%	69.0%
all other	23.2%	31.0%
total	100.0%	100.0%
Race		
White	81.3%	81.6%
Black/African American	10.7%	14.1%
Other	8.0%	4.3%
total	100.0%	100.0%
Respondent Age		
< 20	29.6%	29.1%
20 - 24	4.3%	6.1%
25 - 54	43.3%	45.3%
55 - 64	9.9%	8.2%
65 +	12.8%	11.3%
refusal	(1.2%)	--
total	100%	100.0%

603 **Table 3.** Comparison of RDD 2004 Survey and Census 2000 Geographic Distributions

County, State:	**Census 2000**	**RDD Survey (N = 4,001)**
Cass County, MO	4.6%	4.9%
Clay County, MO	11.1%	12.3%
Jackson County, MO	40.6%	39.9%
Platte County, MO	4.5%	4.6%
Johnson County, KS	26.6%	26.1%
Leavenworth County, KS	3.5%	3.3%
Wyandotte County, KS	9.1%	8.9%
total	100%	100%

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606
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608 **Table 4. Number of Round 2 Re-Tested Vehicles by Stratum**

Stratum (*h*)	Vehicle Class	Model-Year Group	Sample size[1]
1	Truck	Pre 1981	1
2	Truck	1981-1990	4
3	Truck	1991-1995	2
4	Truck	1996 and newer	9
5	Car	Pre 1981	3
6	Car	1981-1990	4
7	Car	1991-1995	7
8	Car	1996 and newer	12
Total			**42**

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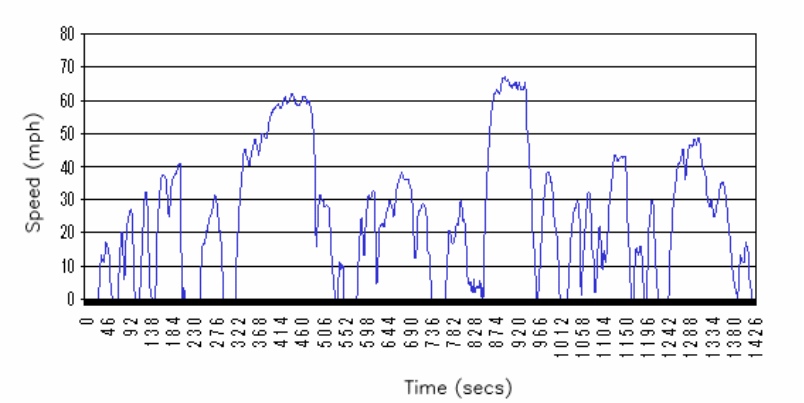
List of Figures

Figure 1. LA92 driving cycle used for driving simulation for the light-duty chassis dynamometer emissions testing (a) compared with an example speed and acceleration trace for the vehicle conditioning route.

(a) LA92 Driving Cycle



(b) Conditioning route speed trace

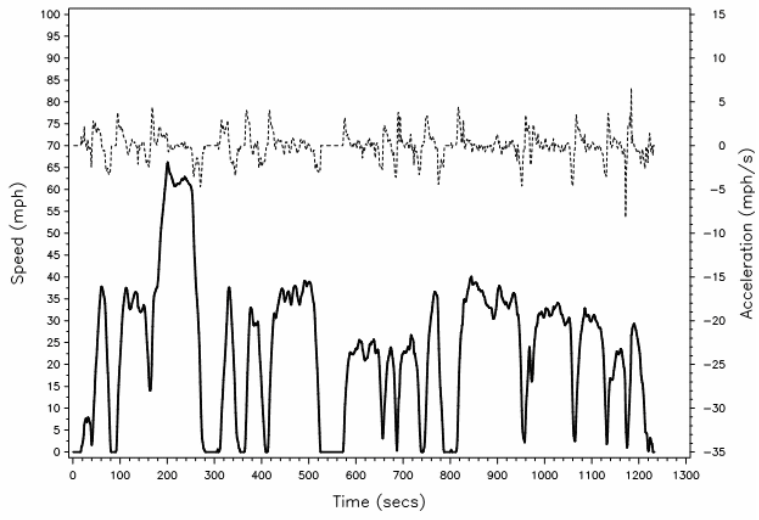
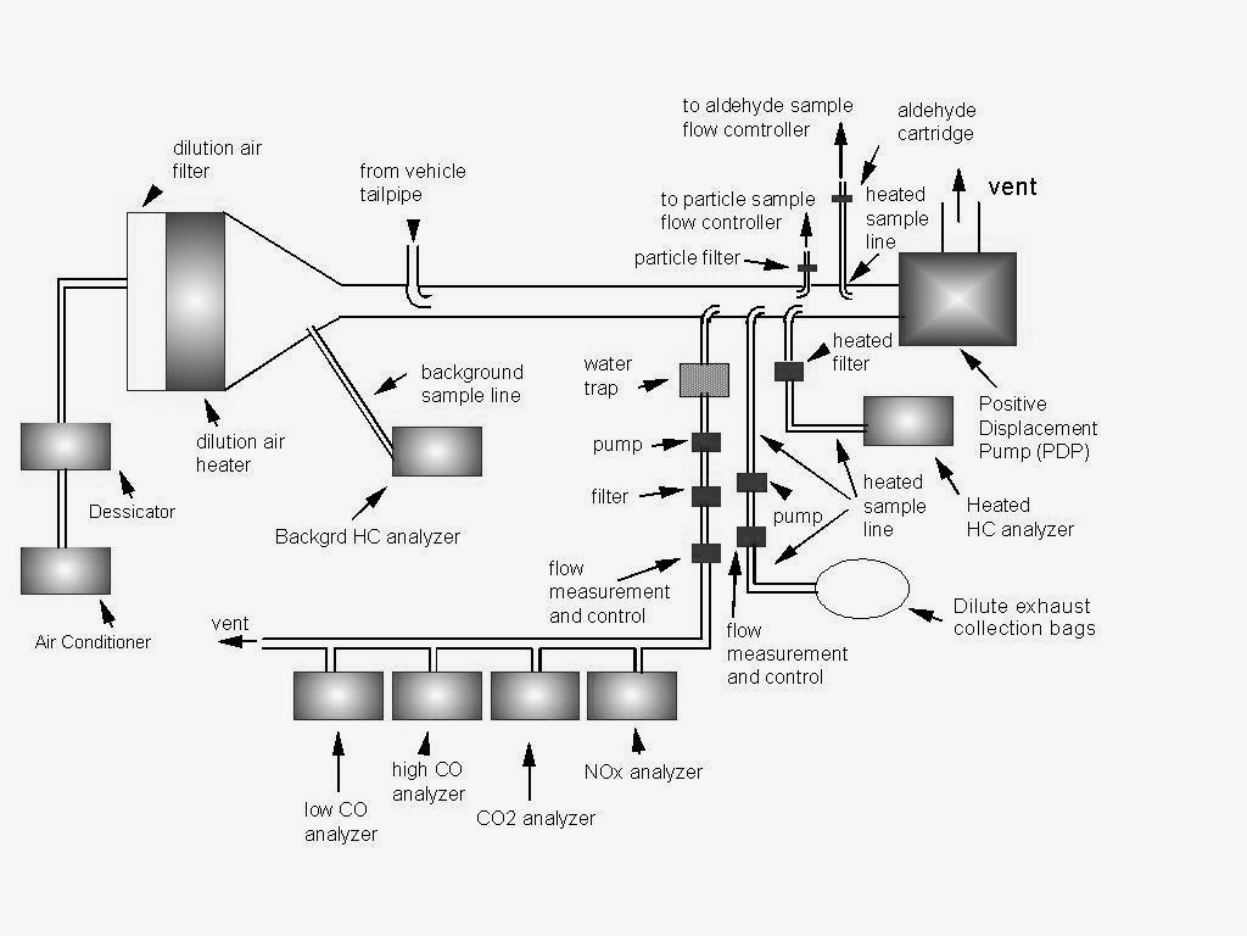


Figure 2. CVS sampling system for the light-duty chassis dynamometer.



649 Figure 3. Sampling train for PM measurements using the light-duty chassis dynamometer.

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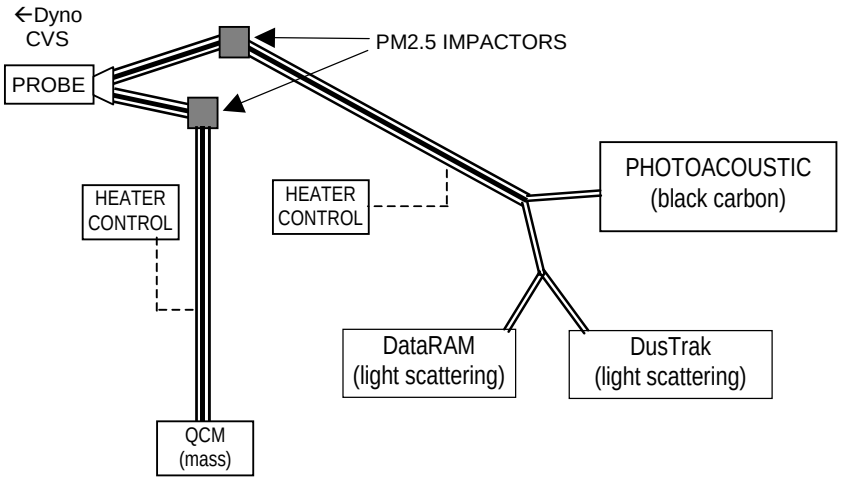

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654 Figure 4. PM emission rates sorted by model-year groups and test round for all vehicles tested
655 during the study.
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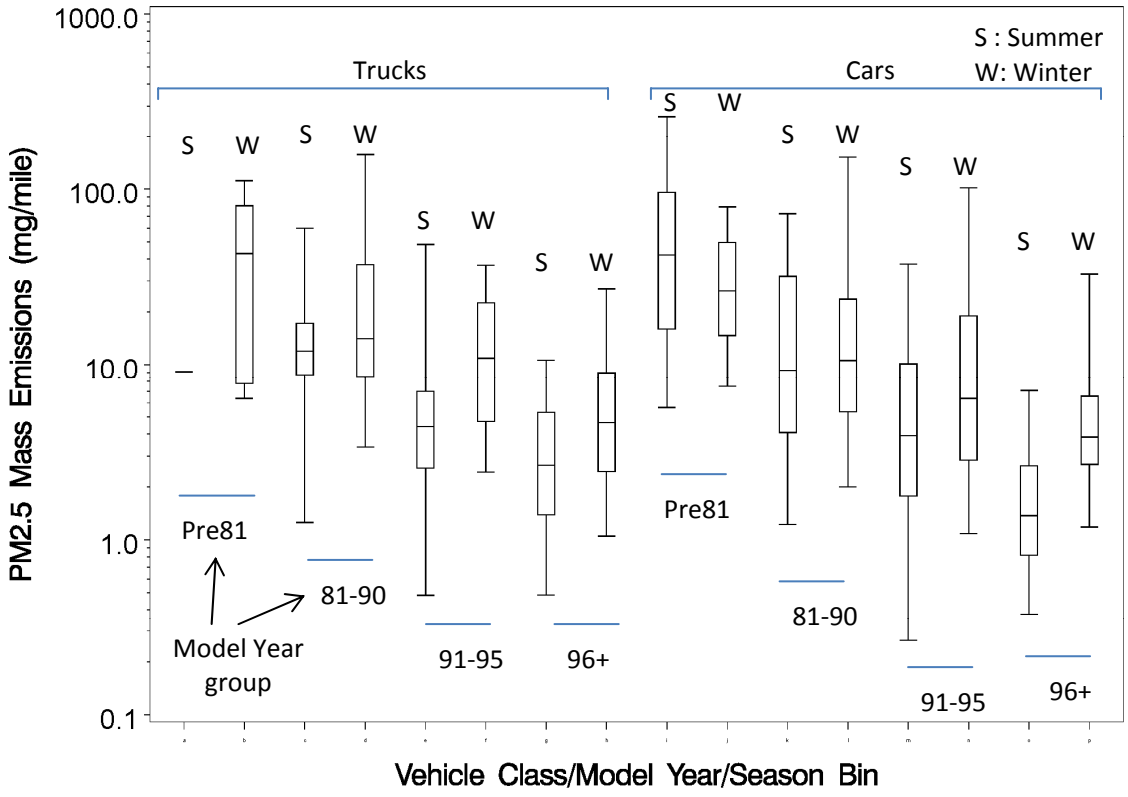

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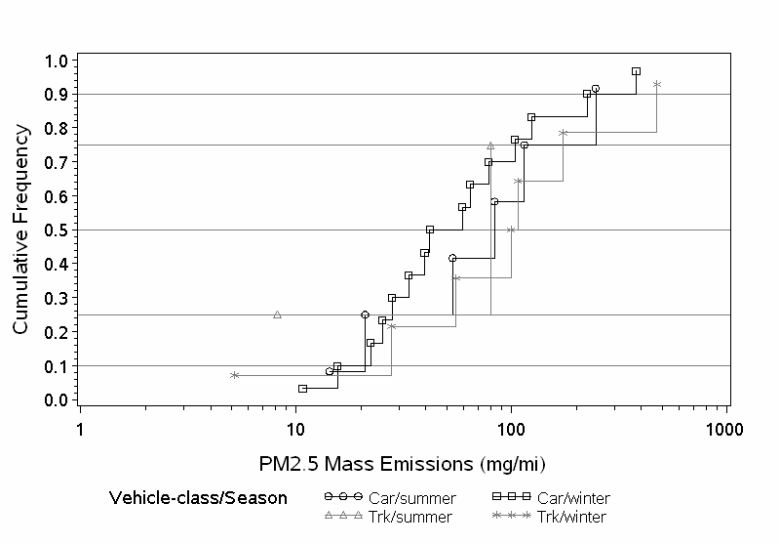
Figure 5. Cumulative distributions of PM emissions measured for the tested vehicle fleet sorted by vehicle type and test round for a) pre-1981 vehicles, b) 1981-1990, c) 1991-1995, and d) 1996-present.

a)



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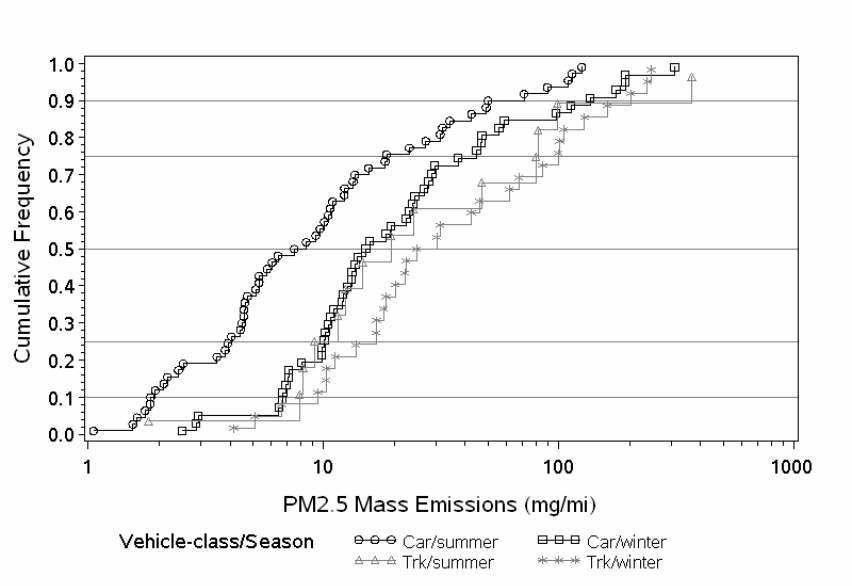

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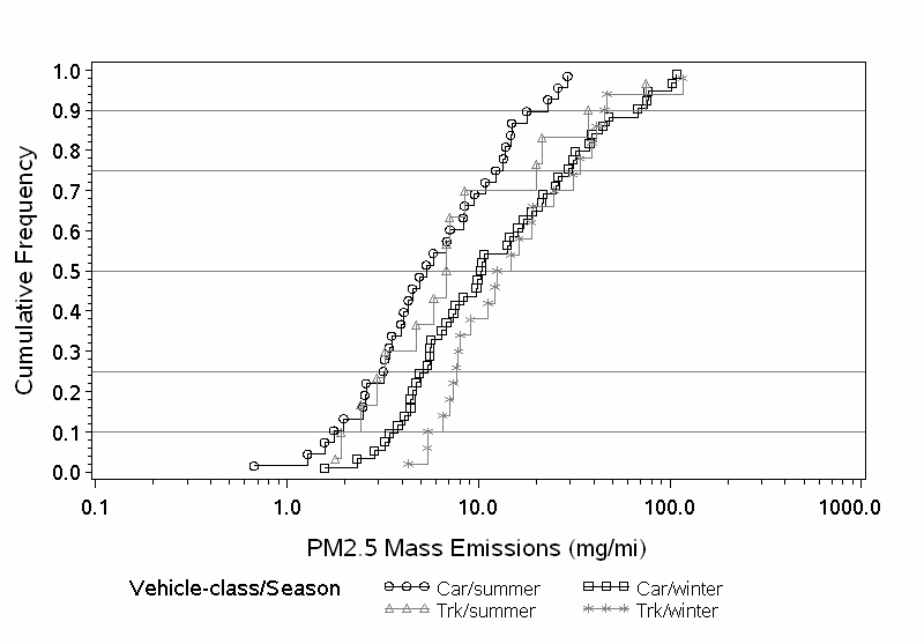
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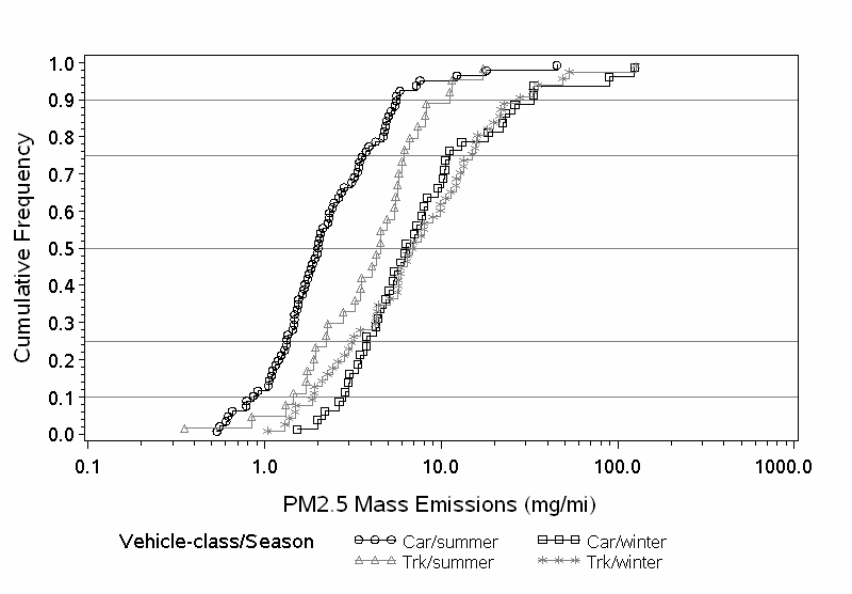
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
0.1
1.0
10.0
100.0
1000.0
PM2.5 Mass Emissions (mg/mi)
Vehicle-class/Season
Car/summer
Car/winter
Trk/summer
Trk/winter

d)


Cumulative Frequency
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0.8
0.7
0.6
0.5
0.4
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0.2
0.1
0.0
0.1
1.0
10.0
100.0
1000.0
PM2.5 Mass Emissions (mg/mi)
Vehicle-class/Season
Car/summer
Car/winter
Trk/summer
Trk/winter

Figure 6. PM2.5 emission rates for all motor vehicles tested during the study, sorted by model year. Open diamond data points indicate vehicle owners who agreed to participate in the study at first contact, while solid square data points identify vehicle owners who initially refused to participate, but agreed after further contact.

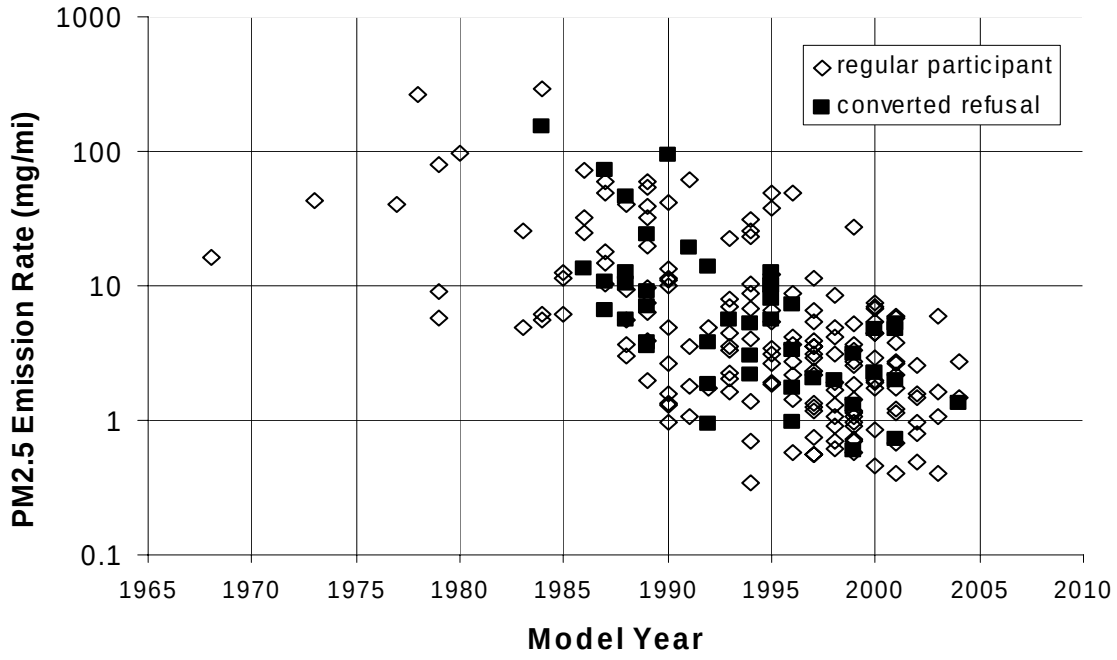

Figure 7. $PM_{2.5}$ emission rates shown as a function of ambient temperature for the 43 vehicles tested in both the summer and winter rounds. Lines connect each individual vehicle's emission rate for the summer and winter round test.