

Comparison of Regulated and PM_{2.5} EC/OC Emissions from Light-Duty Gasoline, Diesel and CNG Vehicles over Different Driving Cycles

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ABSTRACT

Understanding source contributions to ambient atmospheric particulate matter (PM) and toxic chemical pollution is critical to the development of effective mobile source emission control strategies. In this part of a larger PM source apportionment study, motor vehicle exhaust was characterized for eighteen light-duty vehicles driven over four driving cycles on a chassis dynamometer. Vehicle emission testing was conducted from July 2005 to May 2006 and included eleven gasoline vehicles, one gasoline-electric hybrid, two compressed natural gas (CNG) vehicles, and four diesel vehicles. Primary gaseous emissions of total hydrocarbons (THC), CO, NO_x, CO₂, as well as PM_{2.5} (PM with aerodynamic diameter less than 2.5 microns), PM_{2.5} elemental and organic carbon (EC/OC), polycyclic aromatic hydrocarbons (PAHs), and potential molecular markers, such as hopanes and steranes, were measured. Presented here are comparisons of regulated emissions, CO₂, and PM_{2.5} EC/OC from gasoline, CNG, and diesel vehicles. Detailed molecular level analysis of PAHs and marker compounds will be presented elsewhere.

INTRODUCTION

Emissions from on-road vehicles are a significant source of particulate matter (PM) and other toxic pollutants in ambient air, especially in urban areas (1,2). Mobile source PM, especially diesel soot, is associated with adverse health effects, visibility degradation (3), and potential contribution to global warming (4). Furthermore, diesel exhaust is considered a probable human carcinogen by several state, national, and international agencies (5).

Recognizing PM related health and environmental concerns, the United States Environmental Protection Agency (EPA) implemented more stringent PM limits in Tier 2 vehicle emission standards that are being phased in from 2004 through 2009. Furthermore, in 2006, EPA changed the 24-hour PM_{2.5} National Ambient Air Quality Standards (NAAQS) from 65 to 35 micrograms per cubic meter (μg/m³).

Serious challenges face state environmental agencies in developing effective PM control strategies, especially for non-attainment areas. Better understanding of atmospheric PM abundance, spatial distribution, and source apportionment is critical to successful PM management and control, hence current interest and study in these areas (1,2,6-11).

As most vehicle emission profiles have been developed from California vehicles operating in a warm climate (1), an objective of this study was to supplement the emission profile database with profiles from a select group of eighteen New York State light-duty gasoline, CNG, and diesel-powered vehicles operating in both warm and cold weather.

Study results will provide chemical information on PM_{2.5} that is important for source apportionment, especially in regard to estimating the mobile source contribution to ambient PM. Furthermore, data for automotive marker species should help in segregating gasoline and diesel PM contributions, which may, in turn, provide more specific mobile source PM control strategies for the greater New York City airshed.

Vehicle emission testing was performed by the New York State Department of Environmental Conservation (NYSDEC) Automotive Emissions Laboratory (AEL) located in Latham, NY. A total of 18 light-duty vehicles (11 gasolines, 1 gasoline-electric hybrid, 2 CNG, and 4 diesels) were tested from July 2005 to May 2006 on a chassis dynamometer over four different driving cycles.

Exhaust THC, CO, NO_x, and CO₂ were measured by AEL gas analyzers. Exhaust PM_{2.5} samples were collected on filters for gravimetric mass determination by AEL and for PAHs, hopanes, and steranes analyses by Rutgers University. Rutgers also prepared filters for elemental and organic carbon (EC/OC) analyses by a contract laboratory. Real-time PM mass concentration, total PM surface area, and PM-bound PAHs were measured by AEL using a tapered element oscillating microbalance (TEOM), a diffusion charger, and a photoelectric aerosol sensor, the latter two sensors being components of a Matter Engineering AG (Switzerland) NanoMet instrument. In addition, AEL employed PUF-XAD adsorbent cartridge to collect samples for gaseous PAH analysis.

EXPERIMENTAL

VEHICLE EMISSION TESTING

Due to technical limitations, the AEL Consine dynamometer was operated at vehicle target (track) load coefficients rather than at calculated vehicle dynamometer set coefficients, thus resulting in modest additional vehicle load relative to testing with dynamometer set coefficients. The additional loading, however, helped to insure collection of adequate PM_{2.5} sample mass from gasoline vehicles for PAHs, EC/OC, and organic marker compounds analysis and also may have better simulated real-world driving conditions.

For quality purposes, to insure the consistency of loading, a vehicle-dynamometer coastdown was performed after the second and last emission test of each testing day. Coastdown data quantify vehicle loading and could provide a means for reproducing test load conditions in any subsequent study.

Test vehicle selection considered availability, engine/fuel characteristics, and age/mileage. Of the 18 vehicles tested, 14 were recruited from the NYSDEC fleet, 2 vehicles were provided by other state agencies, and 2 diesel passenger cars were procured from private owners. Vehicles were tested as received with the summer or winter fuel. Engine lubricating oil samples were collected from each vehicle and sent to Rutgers University for organic marker compound analysis. Table 1 lists the vehicles tested.

A suite of four driving cycles — the California Unified Cycle (previously known as and referred to here as the LA92), the EPA Highway Fuel Economy Cycle (HWFE), the New York City Cycle (NYCC), and the EPA Supplemental Federal Test Procedure US06 Cycle (US06) — was used to simulate real world driving. Speed traces for the four driving cycles are shown in Figure 1. In this study, the HWFE cycle was driven twice in succession, as a single cycle without any pause between the two cycles. Reported HWFE emission

data, however, are for the second HWFE cycle (766 to 1539 seconds) only. Emission testing on a vehicle was typically performed over three consecutive days with morning and afternoon sessions consisting of a four cycle suite in each session. Each morning session began with a cold start LA92 (engine off and at ambient laboratory temperature, nominally 68 °F to 72°F, for 12 or more hours prior to testing) and was followed by the HWFE, NYCC, and US06 in that order, with each cycle separated by a fifteen-minute engine off period. The last three cycles in the test suite were termed “warm start” cycles (as opposed to the initial “cold start” LA92 cycle) because vehicle engine and catalyst temperatures were much closer to normal operating temperatures at cycle start than in the case of the cold start LA92. For the afternoon session LA92, however, the vehicle engine typically had been off for only 30 to 60 minutes; thus, the afternoon LA92 was considered a warm start test.

A constant volume sampling (CVS) system with blower was used to dilute vehicle exhaust with filtered ambient laboratory room air. The total volume of vehicle raw exhaust was transferred to the CVS through a stainless steel pipe. CVS flow was controlled by a critical flow venturi (CFV) located just upstream of the blower. Depending on the emission level of the test vehicle, one of two CFVs (nominally 415 or 615 cfm) was employed. CVS flow rates were calculated in scfm (68 °F, 1 atm) using a CFV calibration coefficient and CFV gas temperature and pressure data. Emission measurements were performed on dilute exhaust from the CVS system. Figure 2 provides a schematic of the CVS sampling system.

EMISSION MEASUREMENTS

During emission testing, constant flow samples of dilute exhaust were collected by CVS tunnel sample probes and directed to gas analyzers, PM instruments, and PM filters. Table 2 lists the instrumentation used for the emission measurements. A data acquisition system polled AEL continuous gas analyzers at 100 Hz and reduced data to one second average values. Separate TEOM instrument software reported second-by-second PM concentrations as 10-second running average values.

<i>Vehicle Make/Type</i>	<i>Model Year</i>	<i>No. of Cylinders</i>	<i>Engine Displacement (L)</i>	<i>Rated Power (hp)</i>	<i>Odometer (mi)</i>	<i>Test Wt. (lb)</i>
Gasoline						
Ford Taurus	2005	6	3	155	36,362	3625
Honda Civic Hybrid	2004	4	1.2	85	27,800	3000
Chevrolet Cavalier	2004	4	2.2	140	28,892	3375
Chevrolet Venture Minivan	2002	6	3.4	185	54,939	4000
Ford Windstar Minivan	2001	6	3.8	200	50,720	4500
Ford Crown Victoria	2000	8	4.6	200	55,792	4250
Jeep Cherokee	1998	6	4	185	93,839	3875
Oldsmobile Achieva	1997	4	2.4	150	172,270	3250
Jeep Grand Cherokee	1995	8	5.2	230	171,033	4000
Chevrolet Astro Cargo Van	1994	6	4.3	190	105,274	4500
Jeep Cherokee	1992	6	4	185	229,196	3875
Dodge Ram Van	1990	8	5.2	140	154,368	3750
CNG						
Chevrolet Cavalier	2004	4	2.2	140	28,892	3375
Ford Crown Victoria	1999	8	4.6	200	34,015	4500
Diesel						
Volkswagen Jetta	2003	4	1.9	90	74,391	3500
Volkswagen New Beetle	2001	4	1.9	90	64,546	3250
Ford F350	1999	8	7.3	350	115,115	4500
Dodge Ram	1999	6	5.9	300	101,745	4500

Table 1. Vehicle Information

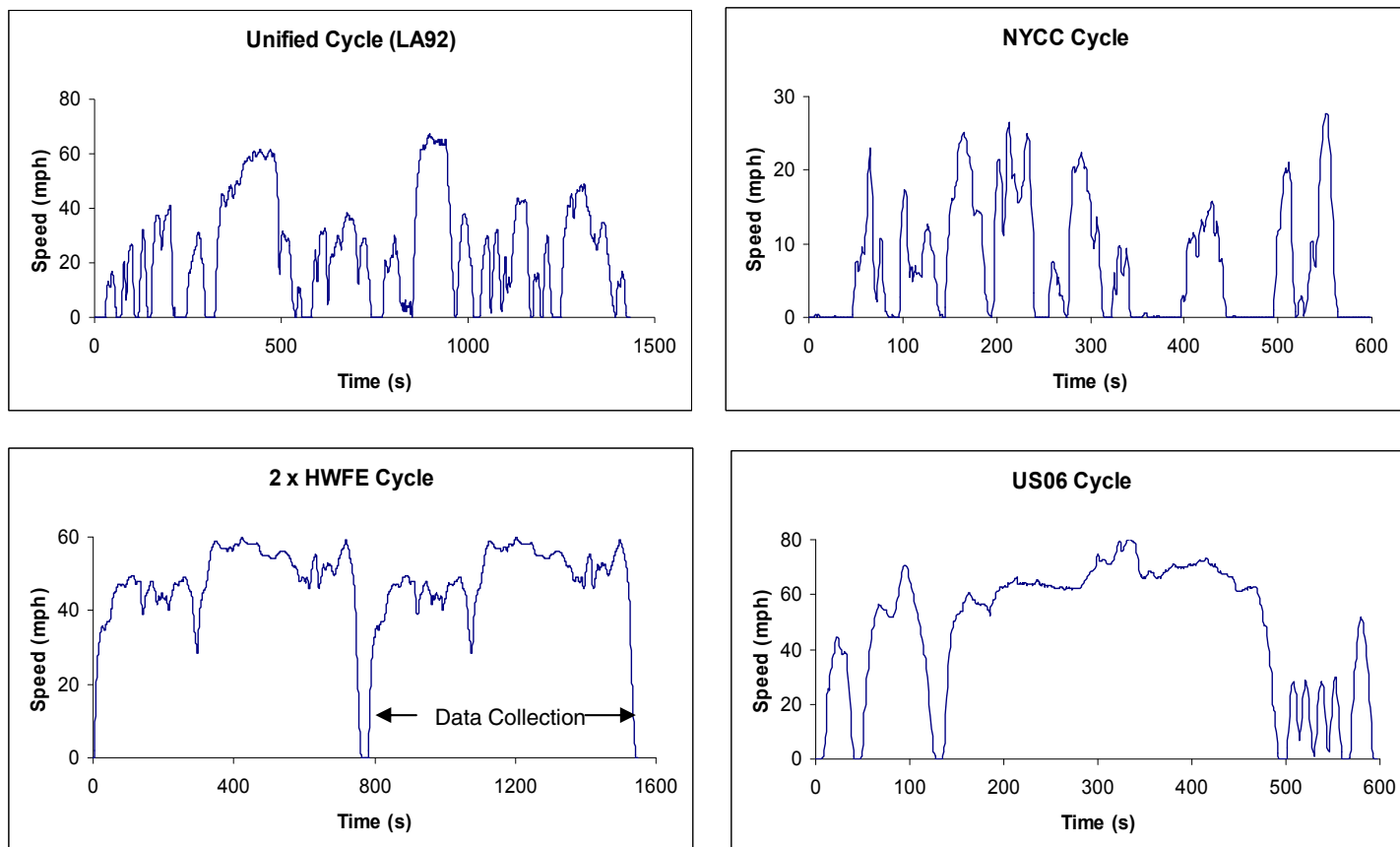


Figure 1. Driving Cycles Employed

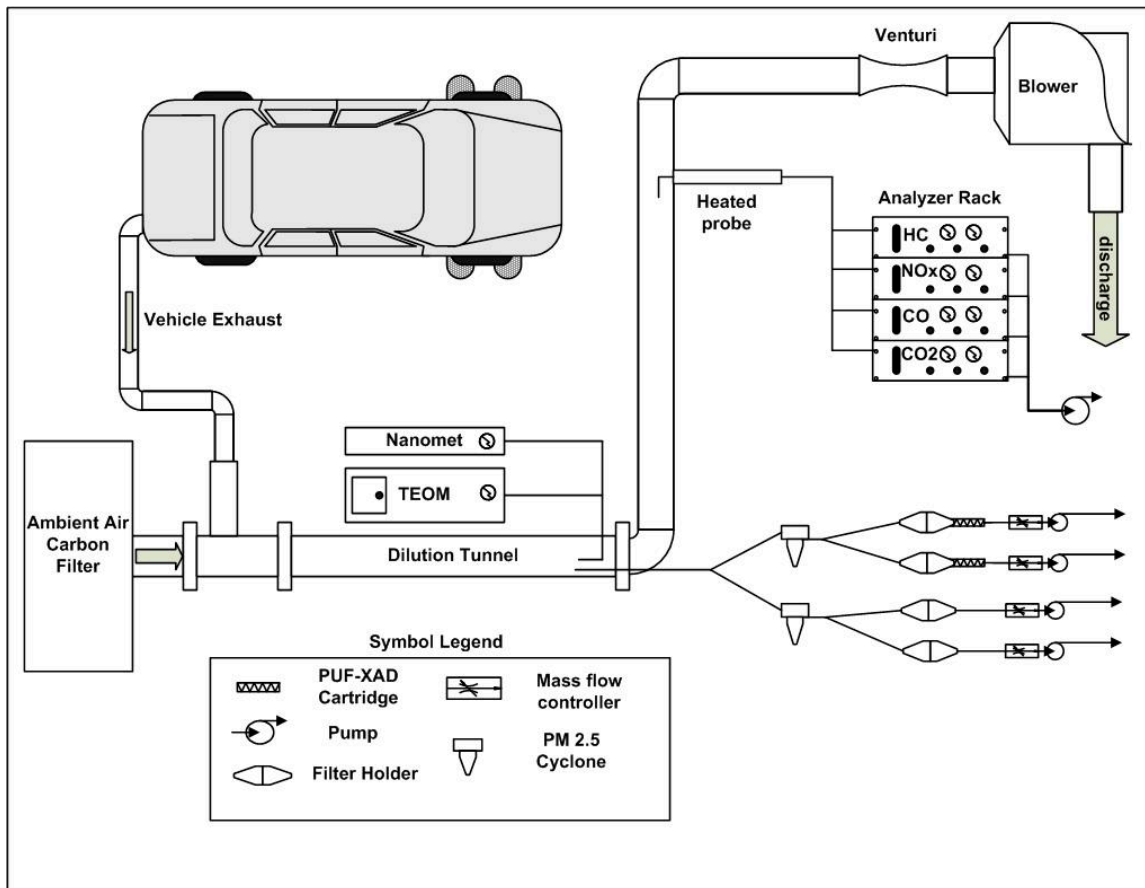


Figure 2. Vehicle Testing and Sampling Diagram

Compounds	Analysis Method	Instrument	Sample Collection
Carbon Monoxide (CO)	Non-Dispersive Infrared (NDIR)	California Analytical Instruments Model 240	Continuous
Carbon Dioxide (CO ₂)	Non-Dispersive Infrared (NDIR)	California Analytical Instruments Model 240	Continuous
Oxides of Nitrogen (NO _x)	Heated Chemiluminescence	California Analytical Instruments Model 600-HCLD	Continuous
Total Hydrocarbons (THC)	Heated Flame Ionization (HFID)	California Analytical Instruments Model 300-HFID	Continuous
Particulate Matter (PM)	PM _{2.5} Gravimetric Procedure	Sartorius SC2 (-OCE) Microbalance	Pallflex™ Filters TX40HI20
	PM (no particle size limitation) Tapered Element Oscillating Microbalance (TEOM)	Rupprecht & Patashnick (now Thermo Scientific) Diesel Particulate Monitor Series 1105	Continuous
Elemental & Organic Carbon (EC/OC)	NIOSH 5040 Thermal Optical Transmittance	Sunset Laboratory, Inc. OCEC Carbon Aerosol Instrument	Pallflex™ Filters TISSUQUARTZ

Table 2. Emission Measurement

For each vehicle emission test, four replicate PM_{2.5} filter samples were collected using two 47 mm Pallflex™ TX40HI20 Teflon coated glass fiber filters and two 47 mm Pallflex™ TISSUQUARTZ filters simultaneously with the employment of two PM_{2.5} cyclones in front of the filter holders (see Figure 2). The two fiber filters were used for gravimetric PM_{2.5} mass measurement, and the two

quartz filters for PM_{2.5} organic marker and EC/OC analyses. Prior to the test, all filters were conditioned for at least 60 min in an environmental chamber maintained at 45±2% relative humidity and 75±1°F. After stabilization, the filters were weighed by a microgram balance (Sartorius SC2-OCE) housed in the environmental chamber. Weighed filters were stored in

covered petri dishes in the environmental chamber until use when filters were placed in a sealed stainless steel filter holder assembly that in turn was placed into the AEL PM_{2.5} sampling system. After sampling over multiple driving cycles to collect sufficient PM_{2.5} for chemical analysis, filters were returned to the environmental chamber and conditioned for at least 60 minutes before being reweighed. The net PM_{2.5} mass, sampling flow rate and CVS flow were used to calculate the vehicle PM_{2.5} emission rate.

Propane injection tests were performed prior to the start of testing, and at several intervals throughout the study to verify CVS flow and analyzer concentration measurements. Gas analyzers were calibrated monthly, and gas analyzer zero/span settings were verified daily prior to emission testing.

ELEMENTAL AND ORGANIC CARBON MEASUREMENT

Elemental carbon (EC) and organic carbon (OC) mass in vehicle exhaust PM_{2.5} was measured by Sunset Laboratory, Inc., (Tigard, OR) using the U.S. National Institute for Occupational Safety and Health (NIOSH) Method 5040, an evolved gas thermal optical transmittance method (12). Exhaust PM_{2.5} was collected on 47 mm Pallflex™ TISSUQUARTZ filters that were precleaned prior to sampling by baking at 500°C for 8 hours in individual aluminum foil envelopes and then stored in the envelopes until actual use. Exhaust PM_{2.5} typically was collected over a four cycle suite of tests that comprised, on average, 76.48 miles of driving. After sample collection at AEL, filters were placed in glass petri dishes that were then sealed with Teflon™ tape and shipped to Rutgers University. At Rutgers, the filters were stored at -20°C until preparation for chemical analysis. An aliquot was removed from each filter with a 1 cm² filter punch and stored in a foil-lined petri dish. The filter punches were sent to Sunset Laboratory, Inc., via overnight shipping for EC, OC, and total carbon (TC) analysis.

Mass concentrations for EC and OC in the aliquot were reported in micrograms per square centimeter (µg/cm²) with an uncertainty of 6% (Sunset Laboratory, 2006). Filter loading was assumed uniform and the total filter mass concentrations for EC and OC were calculated from the aliquot EC and OC mass concentrations and converted to mass per vehicle mile driven units. In terms of mass per vehicle mile, blank values for the TISSUQUARTZ filters were estimated be 0.02 to 0.06 µg/mi OC and not detected to 0.02 µg/mi EC. The CVS tunnel contribution (tunnel wall debris from prior testing) was estimated from two tunnel blank filters to be 0.43 to 0.91 µg/mi OC and up to 0.02 µg/mi EC for the period when gasoline vehicles were being tested. During the diesel vehicle testing period, the tunnel contribution to filter OC and EC increased slightly.

RESULTS AND DISCUSSIONS

COMPARISON OF COLD AND WARM START EMISSIONS

For each vehicle on each test day, a cold start LA92 driving cycle was attempted as the first test of the day. The LA92 was repeated as the first test of the afternoon after an engine off period of 30 to 60 minutes, for which the engine and catalyst temperatures were likely warmer than in first morning test. Same vehicle THC, CO, NO_x, and CO₂ emissions for LA92 first morning (cold start) and first afternoon (warm start) tests are presented in Figure 3 and the Appendix. PM_{2.5} was not collected for individual driving cycles; therefore, cold start vs. warm start data is not available for PM_{2.5}.

As expected, higher emissions and lower fuel economies were generally observed with cold start compared to warm start tests.

Average cold start THC g/mi was 1.9 times the average warm start THC g/mi. The maximum observed cold start to warm start THC g/mi ratio was 3.2. For CO g/mi, the cold start average also was 1.9 times the warm start average and the maximum observed cold/warm ratio was 5.8. For NO_x g/mi, the cold start average was 1.4 times the warm start average and the maximum observed cold/warm ratio was 2.0. CO₂ exhibited much less difference between cold and warm start; the cold start average was only 1.08 times the warm start average and the maximum observed cold/warm ratio was 1.3.

EMISSIONS OVER WARM START DRIVING CYCLES

Warm start emissions of THC, CO, NO_x, CO₂, and fuel economies from four driving cycles are listed in the Appendix.

1. THC EMISSIONS: Figure 4 presents THC emissions for all vehicles over all driving cycles except the cold start LA92. Data magnification for model year 1997 and later gasoline vehicles is provided by the insert graph.

As expected, THC generally decreases with increasing vehicle model year. Typically, THC g/mi emissions were highest over the NYCC (but sometimes highest for the US06 or LA92) and always lowest over the HWFE.

The 1990 Dodge Ram van driven over the NYCC produced the highest observed THC emissions, 5.87g/mi. In contrast, the 2004 Honda Civic hybrid exhibited the lowest NYCC THC emissions, 0.007 g/mi, and lowest THC emissions across all cycles. Over the other three driving cycles, the 1990 Dodge Ram THC emissions ranged from 1.25 to 2.54 g/mi. The ratio of 1990 Dodge Ram to 2004 Honda Civic hybrid THC emissions over the four driving cycles varied from about 90 to over 800.

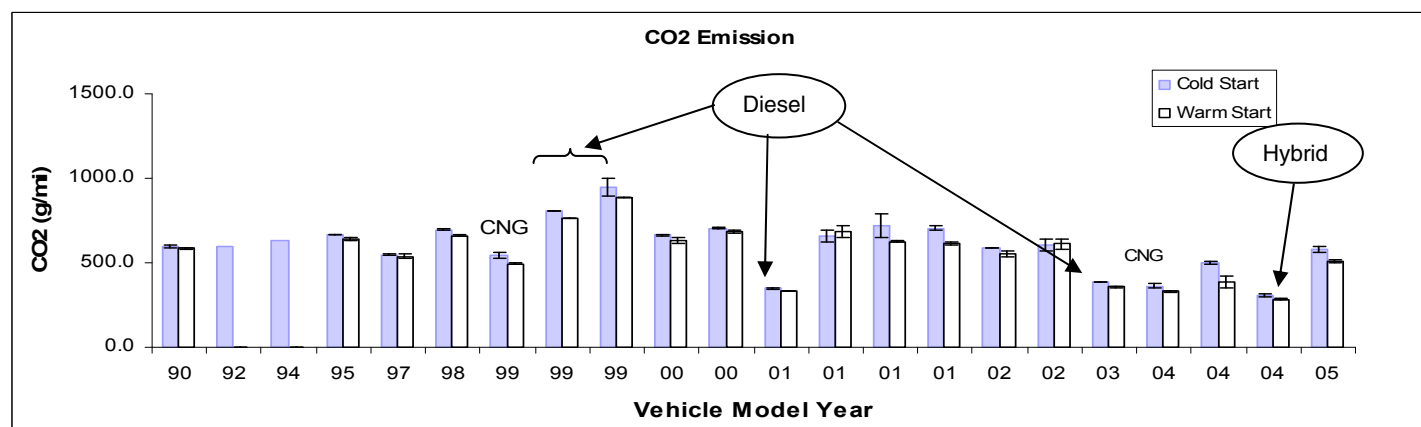
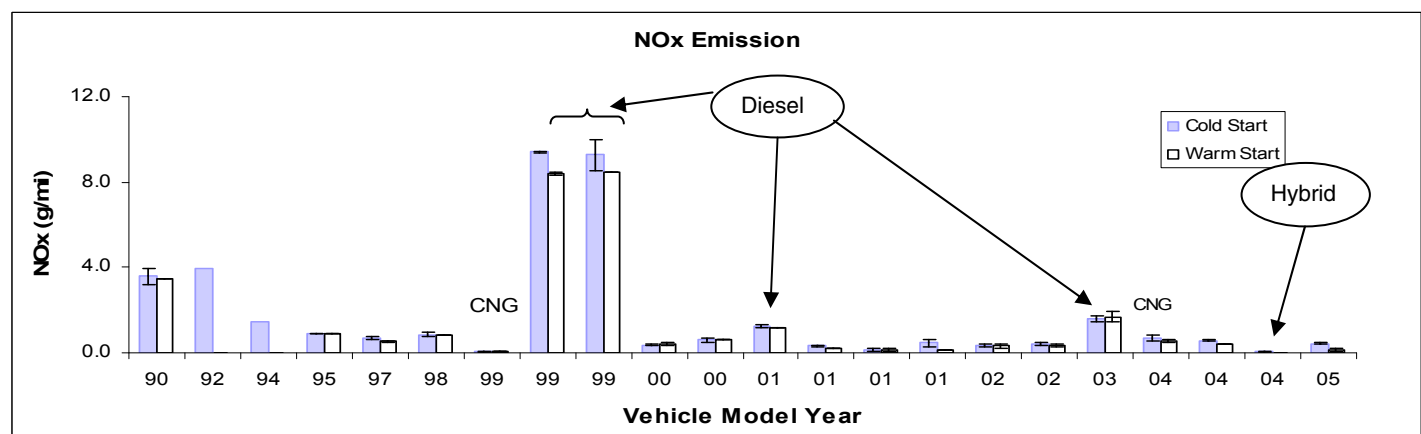
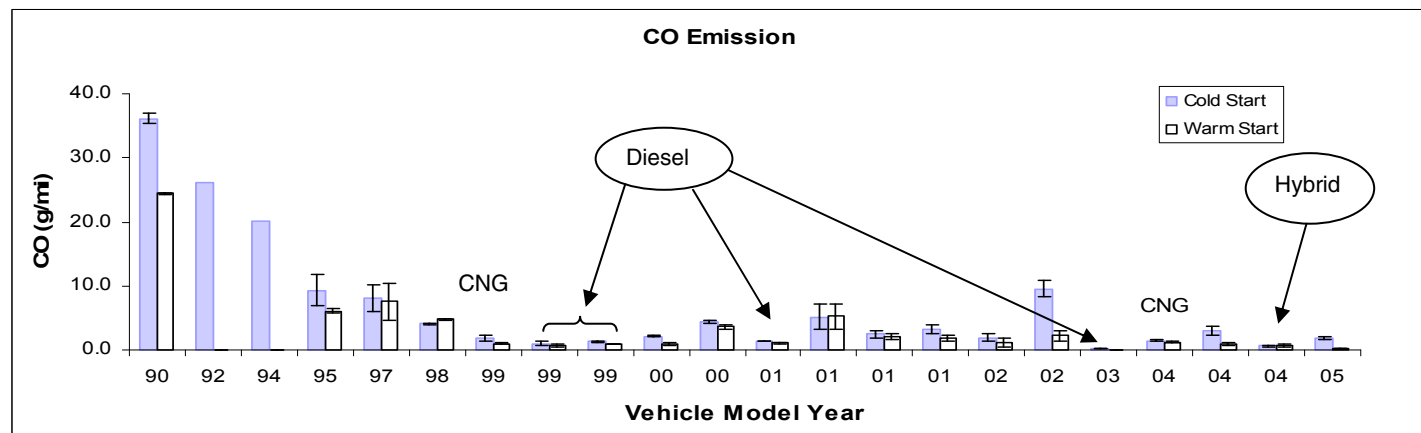
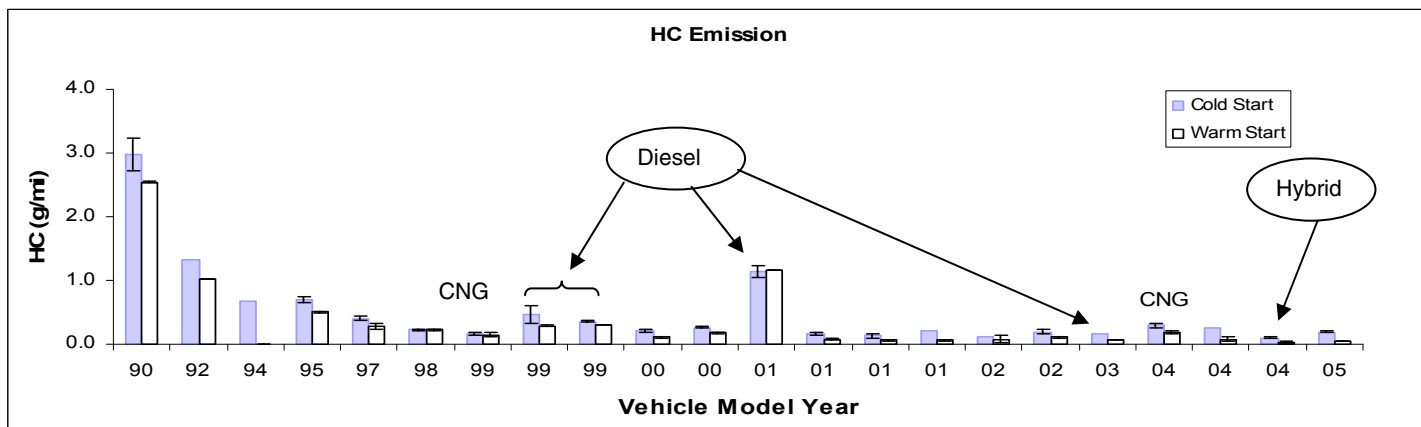


Figure 3. Comparison of Cold and Warm Start Emissions with LA92 Cycle

NOTE: Vertical bars represent 1 SD for multiple measurement data