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Estimates of the emission rates of ammonia from light-duty vehicles using standard chassis dynamometer test cycles

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Abstract

Emissions rates of ammonia (NH_3) are reported for a fleet of 39 in-use light-duty gasoline-fueled vehicles. The fleet consisted of both light-duty passenger vehicles and light-duty trucks with various levels of emission control technologies, ranging from non-catalyst vehicles to those that were certified at the ULEV standard for California. NH_3 measurements were performed using Fourier transform infrared spectroscopy and the federal test procedure (FTP) driving cycle. The FTP NH_3 emission rate for this fleet of vehicles averaged $54\,mg\,mi^{-1}$ with a range from <4 to $177\,mg\,mi^{-1}$. For this fleet of vehicles, NH_3 emissions did not decline as significantly as the regulated pollutants with improvements in emission control technology. A subset of 5 vehicles was tested over the US06, the New York City Cycle (NYCC), and a high-speed freeway cycle for comparison with the FTP cycle. NH_3 emissions showed a strong cycle dependence, with increased emissions under more aggressive driving conditions. These results show that NH_3 emissions formed during more aggressive driving conditions should be considered in the development of NH_3 emission factors. The onset of NH_3 emissions typically occurred after catalyst light-off, near when the catalyst reached its equilibrium temperature. Initial studies showed that NH_3 emissions increased as the sulfur content in the fuel was decreased.

Keywords: Automotive exhaust; Automotive emissions; Vehicle emissions; Chassis dynamometer; Emission rates; Ammonia

1. Introduction

There is increasing concern regarding the adverse health effects associated with airborne particulate matter that is $<2.5\,\mu m$ in diameter ($PM_{2.5}$) and the compounds that are precursors to ambient PM formation. Ammonia (NH_3) is one compound that has received attention as it is known to contribute to the production of secondary PM in the form of ammonium nitrate (NH_4NO_3) or ammonium sulfate ($(NH_4)_2SO_4$). Analysis of ambient PM indicates that ammonium composed from 14.0% to 17.0% of the $PM_{2.5}$ mass at various locations within the South Coast Air Basin (SCAB), which includes Los Angeles and the surrounding metropolitan area (Kim et al., 2000).

The identification of NH_3 in vehicle exhaust dates back to the 1970s (Bradow and Stump, 1977; Cadle et al., 1979; Cadle and Mulawa, 1980; Smith and Carey, 1982; Urban and Garbe, 1979). Early studies showed that reactions over the catalyst surface could result in the formation of NH_3 (Shelef and Gandhi, 1972a, b). More recent studies have indicated that NH_3 emissions from vehicles may be greater than the current emission inventories indicate, although there is a wide range of estimates for NH_3 emissions rates for mobile sources. These include studies in tunnels (Fraser and Cass, 1998; Gertler et al., 2002; Kean et al., 2000; Moeckli et al., 1996), remote sensing studies (Baum et al., 2000, 2001), some limited chassis dynamometer measurements

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(Graham, 1999; Baronick et al., 2000; Volkswagen, 1989), and studies using dedicated vehicles (Shores et al., 2000).

At present, it is estimated that NH_3 emissions from mobile sources are the third-largest source and account for $\sim 18\%$ of the inventory in the SCAB (Chitjian et al., 2000). This estimate is based on tunnel measurements made by Fraser and Cass (1998) rather than on direct measurement of tailpipe emissions. Using a fleet average NH_3 emission factor of 98 mg mi^{-1}, Chitjian et al. (2000) estimated that mobile sources account for 33 tons per day in SCAB. For comparison, livestock and poultry waste is largest single source, and is estimated to be 60 tons per day in SCAB (Chitjian et al., 2000). Soil surface emissions are the second-largest source of NH_3 emissions, contributing 34 tons per day in SCAB.

More needs to be known about NH_3 emission rates from mobile sources and the factors that may influence these emission rates. The purpose of this study was to quantify the NH_3 emission rate for a fleet of in-use vehicles and assess the effect of various levels of emission control technology and driving cycles on NH_3 emissions. The fleet consisted of 39 in-use, gasoline-fueled light-duty passenger vehicles and light-duty trucks. Each vehicle was tested over the United States (US) Federal Test Procedure (FTP) cycle. The fleet included vehicles certified to California's Low Emission Vehicle standards that will represent a larger portion of the in-use fleet in the next 5–10 yr. A subset of 5 of these vehicles was also tested over the US06, New York City Cycle (NYCC), and a high-speed freeway cycle for comparison. Additional experiments were also conducted to evaluate the repeatability of NH_3 measurements and the impact of fuel sulfur levels on NH_3 emissions. NH_3 measurements were done using a Fourier Transform Infrared spectrometer (FTIR), which can measure mass emission rates of compounds such as NH_3 in near real-time (Butler et al., 1981). In addition to NH_3, the emission rates of the regulated pollutants were also determined using standard analytical procedures.

2. Experimental procedures

2.1. Description of vehicle fleet

The 39 gasoline-fueled vehicles were recruited from several sources, including private owners, the University of California at Riverside campus fleet, and rental car companies. A breakdown of the vehicles by manufacturer is provided in Table 1. The vehicle fleet corresponds to reasonable distribution of the major manufacturers and vehicle types, although the study focused primarily on newer cars. All but 5 of the vehicles are 1990 and newer model years. For the 1990 and newer vehicles, the average age of the vehicle fleet was 1996.

Table 1
List of vehicles by manufacturer

Manufacturer	Passenger car	LD truck
GM	3	9
Ford	4	5
Chrysler	3	1
Honda	6	0
Toyota	2	2
Nissan	2	0
Other	1	1

The fleet also included a range of different emission control technology levels including 14 pre-Tier 1 vehicles, 11 Tier 1 vehicles, 8 transitional low-emission vehicles (TLEVs), 1 national low-emission vehicle (NLEV), 2 low-emission vehicles (LEVs), and 3 ultra low-emission vehicles (ULEVs).

2.2. Protocol for vehicle testing

All vehicles were tested over one FTP to obtain mass emission rates for total hydrocarbons (THC), non-methane hydrocarbons (NMHC), carbon monoxide (CO), nitrogen oxides (NO_x), and NH_3. The FTP is a three-phase cycle designed to represent emissions under cold start conditions, hot stabilized operating conditions over an urban route, and hot start conditions. Replicate FTPs were performed on 4 of these vehicles. A subset of 5 vehicles was also tested over the US06, New York City Cycle (NYCC), and a high-speed freeway cycle. The US06 test is designed to be representative of more aggressive, high-speed driving. It has been incorporated into the supplemental certification procedures for light-duty vehicles to represent behavior that is not included in the FTP (Code of Federal Regulations, 2001). The NYCC simulates low-speed urban driving with frequent stops. The high-speed freeway cycle is a facility cycle designed to represent higher speed operation on a freeway (Brzezinski et al., 1999). For two vehicles, some initial tests were also conducted to evaluate the potential impact of fuel sulfur levels on NH_3 emissions.

All tests were conducted in CE-CERT's Vehicle Emission Research Laboratory (VERL) equipped with a Burke E. Porter 48-in single-roll electric dynamometer. Sampling was conducted using VERL's 10-in diameter dilution tunnel and tunnel flow rates of 350 standard cubic feet per minute (SCFM). Since NH_3 is a relatively reactive compound, a heating pad maintained at a temperature of 120°C was wrapped around the transfer tube for some of the experiments to minimize the loss of NH_3 through the sampling system. A comparison of tests run with and without the heating pad showed no difference in the observed NH_3 emission levels, however.

NH_3 emissions were measured using a Pierburg AMA/Mattson FTIR system. The FTIR samples from the dilution tunnel through a $\frac{1}{4}$-in heated sampling line (110°C) with a PTFE core and provides data once every 3 s. The minimum detection limit for NH_3 is 4 mg mi^{-1} over the FTP cycle. The FTIR was calibrated for NH_3 using standard calibration gases from Scott Specialty Gases at levels comparable to what is expected in the diluted exhaust (~10 ppm). The gases were certified from the producer with an accuracy of ±5%, although others have suggested that it is difficult to achieve uncertainties of <10% for NH_3 calibration gases (Marrin, 2001). To adjust the modal emissions data to correct the residence time in the FTIR cell, a well-mixed flow cell model was used. Specifically, the absorption cell for the FTIR has a volume of 5 l, and the residence time in the cell is ~10 s. A 3-s average was applied to the data prior to using the well-mixed flow cell model. The data were also shifted to account for the approximately 17-s delay between the time the exhaust gases are emitted from the tailpipe and when they are sampled by the FTIR. The use of a well-mixed flow cell model for analysis of modal emissions data is described in greater detail by Truex et al. (2000). Regulated pollutants were measured using the standard techniques as outlined in the Code of Federal Regulations (2001).

All but 5 vehicles were tested with the gasoline in the tank at the time the vehicle was procured for testing. Since the specifications for California Phase 2 gasoline are relatively stringent and must provide equivalent emissions under California's Predictive Model, any effects on regulated emissions due to testing with in-tank fuel should be negligible. The sulfur level in the fuel, which some studies have suggested could affect NH_3 emissions (Gandhi and Shelef, 1991), is also limited within a narrow range in California and typically averages between 20 and 25 ppmw in the SCAB (Brisby, 2001). The other vehicles were tested on a certification grade California Phase 2 fuel (2 vehicles) and industry average RFA gasoline (3 vehicles). The industry average RFA gasoline is the base fuel used in studies for the Auto/Oil Air Quality Improvement Research Program (AQIRP) (Hochhauser et al., 1991). For two vehicles, tests were also conducted at two different sulfur levels. These fuels were certification grade California Phase 2 gasoline with nominal sulfur levels of 30 and 330 ppmw. The 330 ppmw California Phase 2 fuel was produced by adding a three-component mixture composed of dimethyldisulfide, thiophene, and benzothiophene to the 30 ppmw base California Phase 2 fuel. These fuels were obtained from Philips Petroleum Chemical Company in Borger, TX. The vehicles for the fuel sulfur tests were preconditioned using procedures used in previous AQIRP programs (Burns et al., 1991).

3. Emissions test results

A summary of the FTP emission results is provided in Table 2 for the 39-vehicle fleet. NH_3 emissions ranged from <4 to 177 mg mi^{-1} with an average of 54 mg mi^{-1}. Results of replicate NH_3 measurements are presented in Table 3. In general, the replicates showed repeatability within 10–20% for NH_3 emissions, although for one vehicle the variability appeared to be considerably greater. For the one vehicle showing the greatest variability for NH_3 emissions, the regulated emissions for these tests all showed repeatability within 15% or better, indicating good repeatability in the testing procedures. The second-by-second NH_3 emissions profiles for the two tests were also qualitatively similar, varying primarily in the magnitude of the NH_3 emissions for each test.

The NH_3 emissions as a function of vehicle certification category were as follows: 12 mg mi^{-1} for pre-1990 vehicles, 72 mg mi^{-1} for 1990 and newer Tier 0 vehicles, 79 mg mi^{-1} for Tier 1 vehicles, 49 mg mi^{-1} for the TLEV vehicles, 56 mg mi^{-1} for a 49 state NLEV vehicle, 4 mg mi^{-1} for the LEV vehicle and 25 mg mi^{-1} for the ULEV vehicles. The average NH_3 emissions as a function of vehicle certification category are presented in Fig. 1 along with average emissions for NMHC, CO, and NO_x. These results show that the emission levels for regulated pollutants have decreased significantly over the years. Overall, NH_3 emissions did not decline as significantly for the range of technology categories tested. It is important to note, however, that only a few vehicles were tested for the low-emission vehicle technology categories and that more data would be needed to provide a more definite comparison of NH_3

Table 2
Average FTP emission results

	NMHC (g mi^{-1})	CO (g mi^{-1})	NO_x (g mi^{-1})	NH_3 (g mi^{-1})	Fuel economy MPG
Average	0.413	6.89	0.573	0.054	21.53
Median	0.156	2.30	0.258	0.046	20.65
High	4.385	117.0	3.709	0.177	42.25
Low	0.031	0.37	0.058	<MDL	11.49
STD	0.758	18.6	0.733	0.053	6.4

<MDL = below minimum detection limit, MPG = miles per gallon.

Table 3
Replicate ammonia FTP emissions

	Cycle	NH_3 emissions ($mg\,mi^{-1}$)	
		Test 1	Test 2
1992 Tier 0 PC FFV	FTP	118	119
1992 Tier 0 PC FFV	US06	196	224
1993 Tier 0 PC	FTP	36	18
1989 Tier 0 Van	FTP	64	52
1989 Tier 0 PC	FTP	<MDL	<MDL

PC = passenger car, FFV = flexible fuel vehicle.

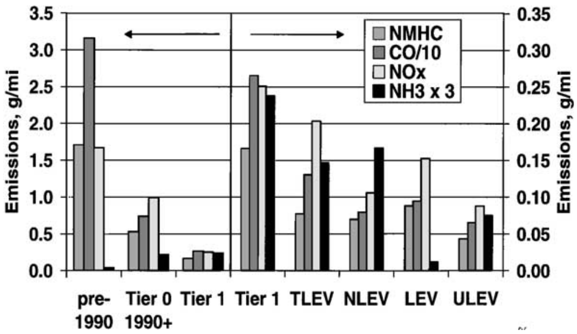

Fig. 1. Average emission results for NMHC, CO, NO_x, and NH_3.

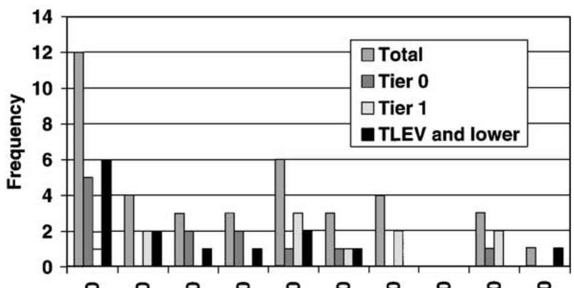

Fig. 2. Histogram of the NH_3 emissions.

emissions for different technology categories. It also should be noted that most of the LEV and ULEV vehicles did have NH_3 emissions near the detection limit.

A histogram of the NH_3 emissions is presented in Fig. 2. These data show that NH_3 emissions for 12 of the 39 vehicles were $10\,mg\,mi^{-1}$ or less. The lowest NH_3 emitting vehicles included vehicles in each different technology category. NH_3 emissions were below the detection limit for the only non-catalyst vehicle tested, consistent with the idea that NH_3 emissions are formed over the catalyst. Above the $10\,mg\,mi^{-1}$ level, there was considerable range in the emission levels, indicating that NH_3 emissions can vary significantly even for vehicles with similar vehicle technology and control strategies. This could be due in part to the fact that NH_3 emissions are not regulated. The individual vehicle data for the highest NH_3 emitting vehicles are presented in Table 4. These data show that, in some cases, NH_3 emissions can have emissions similar to those of other regulated pollutants such as NMHC and NO_x. The data also show that the highest NH_3 emitters came from range of vehicle technology categories including Tier 0, Tier 1 and TLEV.

These NH_3 emission results can be compared qualitatively to the results of other studies. NH_3 emissions have been measured as part of several tunnel studies. In this regard, it is important to note that vehicles measured in tunnel studies are generally operating under more steady state operating conditions than during the FTP. Other differences can include fleet composition and different fuels. Fraser and Cass (1998) conducted a tunnel study in Van Nuys, CA, in 1993, finding an NH_3 emission rate of $98\,mg\,mi^{-1}$ using the carbon balance approach. The Fraser and Cass study was conducted before the introduction of the California Phase 2 fuel used in this study, and the fleet had an older average model year of 1986. Reviewing these results, Gertler et al. (2001) also suggested that this result may be too high and that a better estimate based on this 1993 data may be $48\,mg\,mi^{-1}$. Kean et al. (2000) measured an NH_3 emissions rate of $79 \pm 4.3\,mg\,mi^{-1}$ in a 1999 tunnel study in the Caldecott tunnel in the San Francisco Bay area. This study represented a more modern fleet probably more similar to the fleet in the present study with vehicles operating on California Phase 2 fuel. Some other tunnel studies have measured lower rates including a 1999 study by Gertler et al. (2001) in the Tuscarora tunnel in Pennsylvania ($15.1 \pm 4.3\,mg\,mi^{-1}$), a 1995 study by Moeckli et al. (1996) in Switzerland ($24 \pm 6\,mg\,mi^{-1}$), and a 1981 study be Pierson and Brachaczek (1983) in the Allegheny Tunnel in Pennsylvania ($2.1 \pm 5.6\,mg\,mi^{-1}$ [for $NH_3 + NH_4^+$]). The studies by Moeckli et al. and Pierson and Brachaczek both included considerably higher percentages of non-catalyst vehicles, probably contributing the lower NH_3 emission rates. Gertler et al. suggested their lower emission rates could be due to the newer, better maintained vehicles, higher average speeds, or lack of accelerations/decelerations observed in the Tuscarora tunnel.

Researchers at Environment Canada have also conducted chassis dynamometer tests to measure NH_3 emissions on a fleet of 75 in-use Canadian and United States (US) vehicles (Graham, 1999). These tests were conducted using Canadian in-tank fuel and over only a

Table 4
Individual vehicle data for the highest NH_3 emitting vehicles

	NMHC ($mg\,min^{-1}$)	NO_x ($mg\,mi^{-1}$)	NH_3 ($mg\,mi^{-1}$)	Fuel economy MPG
1996 TLEV SUV	78	535	177	20.70
1995 Tier 1 PC	244	235	157	30.74
1993 Tier 0 PC	866	1394	155	24.57
1996 Tier 1 PC	130	123	144	35.98
1992 Tier 0 FFV PC	129	172	119	21.28
1997 Tier 1 Van	215	412	118	13.98
1991 Tier 0 PC	295	1812	111	42.25
1996 Tier 1 PC	273	258	109	20.70
1993 Tier 1 SUV	161	410	95	17.18
1998 TLEV LDT	87	233	88	17.66

SUV = sport utility vehicle, LDT = light-duty truck.

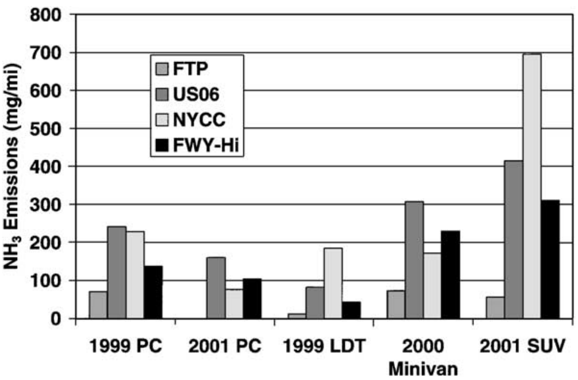

Fig. 3. NH_3 emissions for FTP, US06, NYCC and high-speed freeway cycles.

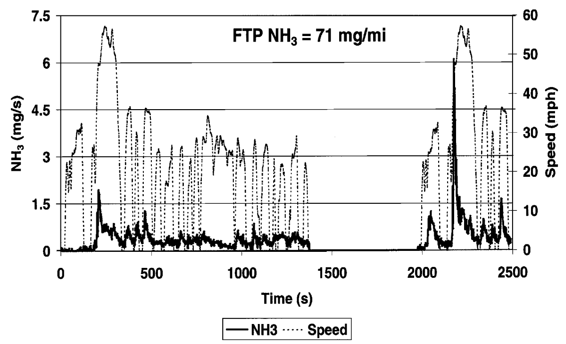

Fig. 4. NH_3 vs. speed for 1999 Tier 1 PC over the FTP cycle.

hot 505 cycle as opposed to a full cold start FTP. NH_3 emissions showed a range from $<1\,mg\,mi^{-1}$ to nearly $300\,mg\,mi^{-1}$ with average emissions by vehicle class of $21\,mg\,mi^{-1}$ for Canadian Tier 0 vehicles, $73\,mg\,mi^{-1}$ for US Tier 0 vehicles, and 65 mg/mi for US Tier 1 vehicles. Overall, the results of our study are qualitatively consistent with those of previous studies showing that while NH_3 emission levels from vehicles are below those of the regulated pollutants, they can still make an important contribution to the overall inventory.

NH_3 emissions for the US06, NYCC, and the high-speed freeway cycles are presented in Fig. 3. Similar to other regulated pollutants, NH_3 emissions were found to increase significantly over the more aggressive driving cycles. This is true even for vehicles that have relatively low levels of NH_3 emissions over the FTP. The results are consistent with previous studies, which have shown that NH_3 emissions can increase significantly under more aggressive operating conditions (Cadle and Mulawa, 1980; Shores et al., 2000). These results also show that NH_3 emissions formed under more aggressive operating conditions need to be included in the development of NH_3 emissions factors for inventory models.

NH_3 modal emissions plotted against vehicle speed are presented in Fig. 4 for the FTP and in Fig. 5 for the more aggressive US06 for one of the vehicles. The modal emissions show the transient nature of the NH_3 emissions throughout the driving cycle. The modal data show that the onset of NH_3 emissions occurs after catalyst light-off, consistent with the formation of NH_3 over the catalyst surface. Experiments conducted on a separate vehicle where the catalyst bed temperature was monitored indicate that NH_3 emissions tend to occur as the catalyst gets closer to its equilibrium temperature rather than during the initial portion of catalyst light-off.

Initial tests were conducted on two vehicles with gasoline containing sulfur levels of 30 and 330 ppmw over the FTP and US06. The results of these tests are presented in Table 5. Although the data are limited, for each of the vehicle/cycle combinations, higher NH_3 emissions were observed for each test with the lower fuel sulfur level. Since NH_3 is primarily formed over the catalyst, these results suggest that sulfur could inhibit NH_3 formation on the catalyst by poisoning reaction sites for NH_3 formation (Gandhi and Shelef, 1991). Early engine dynamometer and simulated exhaust gas experiments have shown that increasing SO_2 concentrations in the exhaust can suppress the formation of NH_3 (Gandhi et al., 1977; Summers and Baron, 1979). Other chassis dynamometer measurements on two vehicles, however, showed that decreasing fuel sulfur content resulted in lower NH_3 emissions for one vehicle and had little effect on NH_3 emissions for the second vehicle (Baronick et al., 2000). The effect of gasoline sulfur levels on NH_3 emissions will be investigated more extensively in future studies in our laboratory.

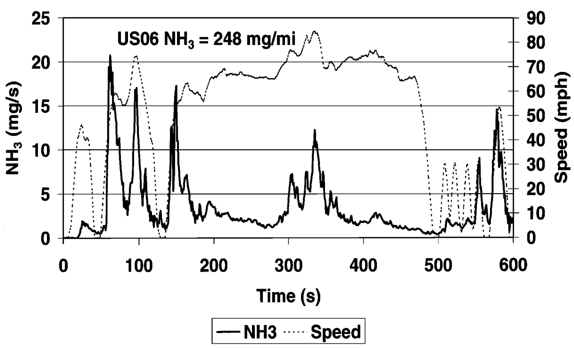

Fig. 5. NH_3 vs. speed for 1999 Tier 1 PC over the US06 cycle.

4. Summary and conclusions

For the present program, a total of 39 in-use light-duty gasoline-fueled vehicles were tested over the FTP with emissions measured for NH_3 using an FTIR. A subset of 5 vehicles was also tested over the US06, NYCC, and high-speed freeway cycle. The major results of this study are:

- NH_3 FTP emissions for the vehicles tested ranged from <4 to $177\,mg\,mi^{-1}$ and averaged $54\,mg\,mi^{-1}$. Of the 39 test vehicles, 12 had FTP NH_3 emissions below $10\,mg\,mi^{-1}$ while the emissions from the remaining vehicles varied significantly depending on the specific vehicle.
- NH_3 emissions did not decline as significantly as those of the regulated pollutants with progressive improvements in emission control systems. Additional data for low-emission technology categories should be obtained, however, to provide a more definitive comparison between different technology categories.
- NH_3 emission levels increased significantly for the more aggressive US06, NYCC, and high-speed freeway cycles. These results show that in the development of NH_3 emission factors, the contribution of NH_3 emissions formed under more aggressive driving conditions should be considered.
- Modal emissions measurements showed that the onset of NH_3 emissions typically occurred after catalyst light-off and near when the catalyst reached its equilibrium temperature.

Table 5
Emissions of NH_3 and regulated components at two different sulfur levels

Vehicle	Cycle	S level	THC ($g\,mi^{-1}$)	NMHC ($g\,mi^{-1}$)	CO ($g\,mi^{-1}$)	NO_x ($g\,mi^{-1}$)	NH_3 ($g\,mi^{-1}$)	Fuel economy MPG
1992 Tier 0 FFV PC	FTP	30	0.177	0.138	2.791	0.176	0.118	21.34
	FTP	30	0.152	0.119	2.364	0.167	0.119	21.21
		Average	0.165	0.129	2.578	0.172	0.119	21.28
	FTP	330	0.221	0.161	3.250	0.226	0.086	21.14
	US06	30	0.225	0.164	9.984	0.619	0.195	20.12
	US06	30	0.289	0.227	11.870	0.599	0.224	20.02
		Average	0.257	0.196	10.927	0.609	0.210	20.07
	US06	330	0.322	0.246	13.184	0.805	0.161	20.09
1997 TLEV PC	FTP	30	0.054	0.051	0.514	0.058	0.038	28.90
	FTP	330	0.061	0.057	0.596	0.060	0.005	28.74
	US06	30	0.085	0.064	11.710	0.225	0.237	24.88
	US06	330	0.093	0.074	10.407	0.216	0.146	25.67

- Initial studies showed that NH_3 emissions increased as the sulfur content in the fuel was decreased over the FTP and US06 cycles.

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