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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 (NH₃) 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. NH₃ measurements were performed using Fourier transform infrared spectroscopy and the federal test procedure (FTP) driving cycle. The FTP NH₃ 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, NH₃ 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. NH₃ emissions showed a strong cycle dependence, with increased emissions under more aggressive driving conditions. These results show that NH₃ emissions formed under more aggressive driving conditions should be considered in the development of NH₃ emission factors. The onset of NH₃ emissions typically occurred after catalyst light-off, near when the catalyst reached its equilibrium temperature. Initial studies showed that NH₃ emissions increased as the sulfur content in the fuel was decreased. © 2002 Elsevier Science Ltd. All rights reserved.

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 μm in diameter (PM₂.₅) and the compounds that are precursors to ambient PM formation. Ammonia (NH₃) 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 (NH₄NO₃) or ammonium sulfate ((NH₄)₂SO₄). Analysis of ambient PM indicates that ammonium composed from 14.0% to 17.0% of the PM₂.₅ 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 NH₃ 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 NH₃ (Shelef and Gandhi, 1972a, b). More recent studies have indicated that NH₃ emissions from vehicles may be greater than the current emission inventories indicate, although there is a wide range of estimates for NH₃ 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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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. 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.

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>
<thead>
<tr>
<th>Manufacturer</th>
<th>Passenger car</th>
<th>LD truck</th>
</tr>
</thead>
<tbody>
<tr>
<td>GM</td>
<td>3</td>
<td>9</td>
</tr>
<tr>
<td>Ford</td>
<td>4</td>
<td>5</td>
</tr>
<tr>
<td>Chrysler</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>Honda</td>
<td>6</td>
<td>0</td>
</tr>
<tr>
<td>Toyota</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Nissan</td>
<td>2</td>
<td>0</td>
</tr>
<tr>
<td>Other</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

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 1/4-in heated sampling line (110°C) with a PTFE core and provides data once every 3 s. The minimum detection limit for NH₃ is 4 mg mi⁻¹ over the FTP cycle. The FTIR was calibrated for NH₃ 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 NH₃ 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 51, 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 NH₃ 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 dimethyl disulfide, 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. NH₃ emissions ranged from <4 to 177 mg mi⁻¹ with an average of 54 mg mi⁻¹. Results of replicate NH₃ measurements are presented in Table 3. In general, the replicates showed repeatability within 10–20% for NH₃ emissions, although for one vehicle the variability appeared to be considerably greater. For the one vehicle showing the greatest variability for NH₃ 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 NH₃ emissions profiles for the two tests were also qualitatively similar, varying primarily in the magnitude of the NH₃ emissions for each test.

The NH₃ 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 NH₃ emissions as a function of vehicle certification category are presented in Fig. 1 along with average emissions for NMHC, CO, and NOₓ. These results show that the emission levels for regulated pollutants have decreased significantly over the years. Overall, NH₃ 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 NH₃ emissions.

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Average FTP emission results</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>NMHC (g mi⁻¹)</td>
</tr>
<tr>
<td>Average</td>
<td>0.413</td>
</tr>
<tr>
<td>Median</td>
<td>0.156</td>
</tr>
<tr>
<td>High</td>
<td>4.385</td>
</tr>
<tr>
<td>Low</td>
<td>0.031</td>
</tr>
<tr>
<td>STD</td>
<td>0.758</td>
</tr>
</tbody>
</table>

< MDL = below minimum detection limit, MPG = miles per gallon.
emissions for different technology categories. It also should be noted that most of the LEV and ULEV vehicles did have NH₃ emissions near the detection limit. Above the 10 mg mi⁻¹ level, there was considerable range in the emission levels, indicating that NH₃ emissions can vary significantly even for vehicles with similar vehicle technology and control strategies. This could be due in part to the fact that NH₃ emissions are not regulated. The individual vehicle data for the highest NH₃ emitting vehicles are presented in Table 4. These data show that, in some cases, NH₃ emissions can have emissions similar to those of other regulated pollutants such as NMHC and NOₓ. The data also show that the highest NH₃ emitters came from range of vehicle technology categories including Tier 0, Tier 1 and TLEV.

These NH₃ emission results can be compared qualitatively to the results of other studies. NH₃ 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 NH₃ 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 NH₃ emissions rate of 79 ± 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 ± 4.3 mg mi⁻¹), a 1995 study by Moeckli et al. (1996) in Switzerland (24 ± 6 mg mi⁻¹), and a 1981 study by Pierson and Brachaczek (1983) in the Allegheny Tunnel in Pennsylvania (2.1 ± 5.6 mg mi⁻¹ [for NH₃ + NH₄⁺]). The studies by Moeckli et al. and Pierson and Brachaczek both included considerably higher percentages of non-catalyst vehicles, probably contributing the lower NH₃ 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 NH₃ 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>
<thead>
<tr>
<th>Cycle</th>
<th>NH₃ emissions (mg mi⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Test 1</td>
<td>Test 2</td>
</tr>
<tr>
<td>1992 Tier 0 PC FFV FTP</td>
<td>118</td>
</tr>
<tr>
<td>1992 Tier 0 PC FFV US06</td>
<td>196</td>
</tr>
<tr>
<td>1993 Tier 0 PC FTP</td>
<td>36</td>
</tr>
<tr>
<td>1989 Tier 0 Van FTP</td>
<td>64</td>
</tr>
<tr>
<td>1989 Tier 0 PC FTP</td>
<td>&lt;MDL</td>
</tr>
</tbody>
</table>

PC = passenger car, FFV = flexible fuel vehicle.
hot 505 cycle as opposed to a full cold start FTP. NH₃ 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⁻¹ for US Tier 1 vehicles. Overall, the results of our study are qualitatively consistent with those of previous studies showing that while NH₃ emission levels from vehicles are below those of the regulated pollutants, they can still make an important contribution to the overall inventory.

NH₃ modal emissions plotted against vehicle speed are presented in Fig. 4. Similar to other regulated pollutants, NH₃ emissions were found to increase significantly over the more aggressive driving cycles. This is true even for vehicles that have relatively low levels of NH₃ emissions over the FTP. The results are consistent with previous studies, which have shown that NH₃ emissions can increase significantly under more aggressive operating conditions (Cadle and Mulawa, 1980; Shores et al., 2000). These results also show that NH₃ emissions formed under more aggressive operating conditions need to be included in the development of NH₃ emissions factors for inventory models.

NH₃ 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 NH₃ emissions throughout the driving cycle. The modal data show that the onset of NH₃ emissions occurs after catalyst light-off, consistent with the formation of NH₃ over the catalyst surface. Experiments conducted on a separate vehicle where the catalyst bed temperature was monitored indicate that NH₃ 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 NH₃ emissions were observed for each test with the lower fuel sulfur level. Since NH₃ 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.

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

<table>
<thead>
<tr>
<th>Vehicle</th>
<th>Cycle</th>
<th>S level</th>
<th>THC (g mi$^{-1}$)</th>
<th>NMHC (g mi$^{-1}$)</th>
<th>CO (g mi$^{-1}$)</th>
<th>NO$_x$ (g mi$^{-1}$)</th>
<th>NH$_3$ (g mi$^{-1}$)</th>
<th>Fuel economy</th>
<th>MPG</th>
</tr>
</thead>
<tbody>
<tr>
<td>1992 Tier 0 FFV PC</td>
<td>FTP</td>
<td>30</td>
<td>0.177</td>
<td>0.138</td>
<td>2.791</td>
<td>0.176</td>
<td>0.118</td>
<td>21.34</td>
<td></td>
</tr>
<tr>
<td></td>
<td>FTP</td>
<td>30</td>
<td>0.152</td>
<td>0.119</td>
<td>2.364</td>
<td>0.167</td>
<td>0.119</td>
<td>21.21</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td></td>
<td>0.165</td>
<td>0.129</td>
<td>2.578</td>
<td>0.172</td>
<td>0.119</td>
<td>21.28</td>
<td></td>
</tr>
<tr>
<td></td>
<td>FTP</td>
<td>330</td>
<td>0.221</td>
<td>0.161</td>
<td>3.250</td>
<td>0.226</td>
<td>0.086</td>
<td>21.14</td>
<td></td>
</tr>
<tr>
<td></td>
<td>US06</td>
<td>30</td>
<td>0.225</td>
<td>0.164</td>
<td>9.984</td>
<td>0.619</td>
<td>0.195</td>
<td>20.12</td>
<td></td>
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<tr>
<td></td>
<td>Average</td>
<td></td>
<td>0.289</td>
<td>0.227</td>
<td>11.870</td>
<td>0.599</td>
<td>0.224</td>
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</tr>
<tr>
<td></td>
<td>US06</td>
<td>330</td>
<td>0.322</td>
<td>0.246</td>
<td>13.184</td>
<td>0.805</td>
<td>0.161</td>
<td>20.09</td>
<td></td>
</tr>
<tr>
<td>1997 TLEV PC</td>
<td>FTP</td>
<td>30</td>
<td>0.054</td>
<td>0.051</td>
<td>0.514</td>
<td>0.058</td>
<td>0.038</td>
<td>28.90</td>
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<tr>
<td></td>
<td>FTP</td>
<td>330</td>
<td>0.061</td>
<td>0.057</td>
<td>0.596</td>
<td>0.060</td>
<td>0.005</td>
<td>28.74</td>
<td></td>
</tr>
<tr>
<td></td>
<td>US06</td>
<td>30</td>
<td>0.085</td>
<td>0.064</td>
<td>11.710</td>
<td>0.225</td>
<td>0.237</td>
<td>24.88</td>
<td></td>
</tr>
<tr>
<td></td>
<td>US06</td>
<td>330</td>
<td>0.093</td>
<td>0.074</td>
<td>10.407</td>
<td>0.216</td>
<td>0.146</td>
<td>25.67</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 5. NH$_3$ vs. speed for 1999 Tier 1 PC over the US06 cycle.
Initial studies showed that NH₃ emissions increased as the sulfur content in the fuel was decreased over the FTP and US06 cycles.

Acknowledgements

The authors acknowledge the contribution and support of Dave Martis, Joseph Calhoun, Ross Rettig and Joe Valdez of the Vehicle Emissions Research Laboratory (CE-CERT) who performed the emissions testing on the vehicles. We thank Carl Fulper, Carl Scarbro, Kent Helmer, and John White of the United States Environmental Protection Agency for their assistance in the development of the technical program and comments on the manuscript. We also thank the United States Environmental Protection Agency for its financial technical support of this project under cooperative agreement No. CX827692-01-0.

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