

On-road tailpipe characterization of exhaust ammonia emissions from in-use light-duty gasoline motor vehicles



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ABSTRACT

A Portable Emissions Monitoring System (PEMS) has been used to estimate ammonia emission rates from a representative fleet of 47 in-use light-duty gasoline motor vehicles over 145 on-road Real Driving Emissions (RDE) tests. The PEMS modules were carried onboard the tested vehicles and were wired such that their ceramic exhaust emission sensors were mounted directly in the tailpipe. The on-road RDE tests were conducted over an urban testing route that included residential and highway roads, uphill and downhill road segments, stop signs, traffic lights, and a school zone with a reduced speed limit. The entire vehicle test sample had an average ammonia emission rate of 114.7 mg/mile \pm 135.3 (StD). This would yield an estimated 2909 metric tons per year of NH₃ emissions from the Wasatch fleet on-road gasoline motor vehicles. Old vehicles with aged Three-Way Catalyst (TWC) converters had higher NH₃ emissions rates than newer vehicles with newer TWC converters. For instance, Tier 0, Tier I, NLEV, Tier II and Tier III vehicles had average emission rates of 563.1, 177.8, 213.6, 94.4 and 18.9 mg/mile, respectively. Carbon monoxide, and nitrogen oxides had a strong correlation with ammonia emission rates, with $r \geq 0.70$. A Moderate correlation was found with vehicles' mileage ($r = 0.6$), model year ($r = -0.5$), engine displacement ($r = 0.4$), and number of cylinders ($r = 0.4$). The outcomes highlight vehicle contributions to the atmospheric NH₃ inventory and the impact of vehicle characteristics and ammonia precursor concentrations on ammonia emission rates from gasoline vehicles.

Authorship statement

All persons who have made substantial contributions to the work reported in the manuscript (e.g., technical help, writing and editing assistance, general support), but who do not meet the criteria for authorship, are named in the Acknowledgements and have given us their written permission to be named.

1. Introduction

Ammonia (NH₃) is a colorless reactive gas with a sharp characteristic odor. It is an inorganic chemical compound composed of a single nitrogen atom (N) covalently bonded to three atoms of hydrogen (H). Ammonia is one of the most abundant alkaline gases in the atmosphere

and is the third most abundant nitrogen-containing atmospheric compound after nitrogen (N₂) and nitrous oxide (N₂O) (Kean et al., 2000). Because of that, ammonia plays a key role in atmospheric chemistry. For instance, it contributes to forest decline and vegetation damage, visibility problems and formation of photochemical smog, dry and wet deposition, and the eutrophication process in lakes (Behera et al., 2013; Moeckli et al., 1996). Nevertheless, the biggest concern regarding atmospheric NH₃ is its contribution to the formation of fine secondary PM_{2.5} such as ammonium sulfate ((NH₄)₂SO₄) and ammonium nitrate (NH₄NO₃). PM_{2.5} particles are particles with an aerodynamic diameter of less than 2.5 μm (Abualqumboz et al., 2017; Nazif et al., 2016, 2018). Because of their small size, secondary (NH₄)₂SO₄ and NH₄NO₃ can penetrate deeply into human lungs and reach lung alveoli, which can result in several respiratory and cardiovascular diseases (Chaney et al.,

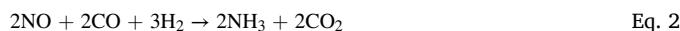
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2017; Landrigan, 2017; Kampa and Castanas, 2008). Previous studies have shown that PM_{2.5} particles along the Wasatch Front and in the Cache County region in the U.S. (United States) State of Utah, where this study was conducted, are comprised mostly of secondary aerosols, including ammonium chloride (NH₄Cl) and (NH₄)₂SO₄ (Kelly et al., 2017). Baasandorj et al. (2018) and Martin et al. (2016) also reported that NH₄NO₃ makes up the bulk of the PM_{2.5} particles along the Wasatch Front region, accounting for approximately 90% of their total mass.

Atmospheric NH₃ has long been known to be an emission from biological processes in soil, biomass burning, ammonia-based chemical fertilizers, sewage treatment plants, and animal waste decay processes (Behera et al., 2013; Chang et al., 2016). However, early studies indicated that substantial amounts of atmospheric NH₃ could be attributed to gasoline motor vehicle fleets following the introduction of Three-Way Catalyst (TWC) converters in 1981 (Bradow and Stump, 1977; Cadle et al., 1979; Cadle and Mulawa, 1980; Smith and Carey, 1982; Urban and Garbe, 1979). Three-way catalyst converters are small canisters containing a series of ceramic screens coated with rare earth metals including palladium (Pd), platinum (Pt) and rhodium (Rh). They are designed to control exhaust emissions of carbon monoxide (CO), unburned hydrocarbons (HC), and nitrogen oxides (NO_x) from gasoline motor vehicles. Three-Way Catalyst converters remove these pollutants from the exhaust from fuel combustion by simply converting them into less-harmful compounds such as CO₂, H₂O and N₂ (de Nevers, 2010). This is usually done through a set of simultaneous chemical oxidation/reduction reactions on their surfaces. The Pd, Pt and Rh rare earth metals coating the TWC converter surfaces are chemical catalysts that significantly accelerate the oxidation/reduction reactions by reducing the activation energy barriers of these reactions (Heck et al., 2009). The TWC converters oxidize CO and unburned HC compounds to CO₂ and H₂O, and reduce NO_x molecules such as nitrogen monoxide (NO) to N₂ gas (Farrauto et al., 2019; Johnson, 2016; Johnson and Joshi, 2018). The over-reduction of NO compounds at the surfaces of TWC converters beyond the formation of molecular N₂ gas can result in NH₃ formation, primarily linked to the reaction of NO and H₂ (Wang et al., 2015). The process normally starts with a water-gas shift reaction between CO and H₂O compounds that causes CO₂ and H₂ to be produced (Eq. (1)), and then NH₃ forms as an outcome of the reaction between the produced H₂ gas and NO compounds (Eq. (2)-Eq. (3)).



The production of NH₃ over TWC converters surfaces can increase further when vehicle engines run at rich air/fuel conditions (Heeb et al., 2006, 2008). These conditions favor reducing processes on the surfaces of TWC converters and also cause formation of higher levels of pre-catalyst CO emissions, resulting in more H₂ gas. Rich air/fuel conditions may occur in cases of aggressive driving (Heeb et al., 2008; Huai et al., 2003) or with malfunctioning oxygen (lambda) sensors that no longer maintain the air/fuel ratio at stoichiometric conditions (Cadle and Mulawa, 1980; Fraser and Cass, 1998; Urban and Garbe, 1979). The condition (age) of TWC converters can also impact NH₃ production rates on their surfaces. For instance, Wang et al. (2019) showed that aging a Pd/Rh TWC converter significantly increased the amount of NH₃ formed on its surface (Durbin et al., 2004). also showed increasing NH₃ emissions for aged catalyst for a fleet of 12 vehicles. Other factors that might affect NH₃ production over TWC converter surfaces include vehicle and engine specifications, state of operation, ambient temperature, and engine temperature (Czerwinski et al., 2016; Moeckli et al., 1996; Padeste and Baiker, 1994).

Accurate quantification of raw exhaust NH₃ concentrations would result in a better understanding of vehicle contributions to the atmospheric particulate matter inventory. It would also help regulators and

stakeholders to better plan, facilitate and implement effective secondary PM_{2.5} reduction strategies, especially in areas like the Wasatch Front and Cache County regions where these pollutants occur at risky levels. Ammonia concentrations from gasoline motor vehicles have been estimated using on-road tunnel measurement studies (Emmenegger et al., 2004; Fraser and Cass, 1998; Kean et al., 2000, 2009; Liu et al., 2014; Moeckli et al., 1996; Pierson and Brachaczek, 1983), chassis dynamometer studies (Borsari and Assunção, 2017; Durbin et al., 2002, 2004; Heeb et al., 2006, 2008; Huai et al., 2003, 2004, 2005; Livingston et al., 2009; Mohn et al., 2004; Sutton et al., 2000), and studies using remote sensing instruments (Baum et al., 2000, 2001; Burgard et al., 2006; Zhang et al., 2021) that are placed near the roads to measure ammonia concentrations based on ammonia's absorption effect on a spectrum for a specific wavelength. On-road tunnel studies estimate ammonia emission rates from in-use motor vehicles depending on field measurements inside highway tunnels, whereas chassis dynamometer studies rely on having different types of vehicles with specific characteristics operated on dynamometers over different driving schedules and cycles. Recent studies, such as (Mendoza-Villafuerte et al., 2017), have used onboard Portable Emissions Monitoring System (PEMS) for measuring NH₃ exhaust emissions of a heavy-duty motor vehicle.

In this study, a representative test sample of 47 in-use light-duty (LD) gasoline motor vehicles was used to estimate the average NH₃ emission rates of the on-road LD gasoline motor vehicles fleet of the Wasatch Front and Cache county region in the north-central part of the U.S. state of Utah. Ammonia concentrations of the test sample vehicles were measured using a PEMS over 145 on-road Real Driving Emissions (RDE) tests. The RDE tests were conducted on a 5.3-mile urban testing cycle designed using the local road network in the City of Logan located at the north end of the Cache County. The test sample had the same federal emission standards (Tier levels) distribution as the overall on-road gasoline motor vehicle fleet of the Wasatch Front and Cache County region. The federal emission standards classification designed by the U. S. EPA was used to replicate the on-road gasoline vehicles fleet because the agency normally assigns each major revision of on-road vehicle tailpipe and evaporative emission standards to a "Tier level". The plots presented in Supplementary 1 clearly show how gasoline motor vehicles with different emission standards would cause different emission rates of ammonia into the atmosphere. The raw data and the R Jupyter notebook used to analyze the raw data can be found in this online resource (Abualqumboz et al., 2022) that has been created on the HydroShare collaborative website (<https://www.hydroshare.org>). HydroShare is an online, open-source environment for sharing hydrologic data and models.

2. Study Area-Wasatch Front and Cache County, UT

The Wasatch Front and Cache County region is located in the north-central part of the U.S. state of Utah and represents the state's prime arable land. It is where more than 80% of Utah's population resides. It is currently the third-fastest growing region in the nation and is projected to double in population size by 2065 (Woods, 2020). According to the U. S. EPA, the six counties of the Wasatch Front, Box Elder, Davis, Salt Lake, Tooele, Utah, and Weber and the Cache County were designated as non-attainment areas for PM_{2.5} in 2020 (EPA, 2022). Additionally, in January 2004, Cache County (Also known as Cache Valley) had the worst ever PM_{2.5} pollution episode in the United States. The PM_{2.5} concentration was recorded at 132.5 µg/m³, which is almost four times the EPA National Ambient Air Quality Standard (NAAQS) of 35 µg/m³ (Malek et al., 2006). Alarming pollution episodes of PM_{2.5} particles in northern Utah counties are closely related to formation of Persistent Cold Air Pools (PCAPs) that trap and accumulate urban particulate emissions within a shallow boundary layer near the earth's surface (Baasandorj et al., 2018). For instance, the nation's worst PM_{2.5} pollution episode in 2004 occurred mainly because of a strong inversion that was created due to the Cache County's geographical, meteorological,

and environmental characteristics. The inversion was further enhanced by a prolonged high atmospheric surface pressure event and because of the Cache County snow-covered surface that caused atmospheric temperatures to significantly decline and also caused the reflection of solar radiation to increase to about 80% (Malek et al., 2006).

According to the Utah Division of Motor Vehicles (DMV), the Wasatch Front counties and the Cache County had approximately 1,826,584 registered LD gasoline motor vehicles with a Gross Vehicles Weight Rating (GVWR) up to 12,000 pounds as of February 2019. For this research, these on-road gasoline vehicles were first assigned to the appropriate EPA emission standard (tier level) based on their model year (MY), as shown by the green bars in Fig. 1. The tier levels were Pre-Tier 0 (MY \leq 1980), Tier 0 (MY from 1981 to 1993), Tier I (MY from 1994 to 2000), National Low Emission Vehicle (NLEV) (MY from 2001 to 2003), Tier II (MY from 2004 to 2016) and Tier III (MY from 2017 to 2025). After that, a representative sample of $n = 47$ LD gasoline vehicles was selected to represent the on-road LD gasoline vehicle fleet. The test sample vehicles had the same tier-level distribution as the on-road LD gasoline vehicle fleet of the Wasatch Front and the Cache County, as illustrated in Fig. 1. The yellow bars represent the population of tested gasoline vehicles of each tier level. The model year of the tested vehicles was not pre-defined. The research team continued to collect and test vehicles until the total number of vehicles needed for each tier level was complete. Tested vehicles were collected from family members, friends, colleagues, the Utah Water Research Laboratory (UWRL) at Utah State University (USU), and USU Facilities. Fig. 1 also shows that no Pre-Tier 0 (MY \leq 1980) vehicles were tested. This is mainly because gasoline vehicles made on or before 1980 emit negligible amounts of NH₃ due to the absence of TWC converters (Cadle and Mulawa, 1980; Pierson and Brachaczek, 1983). Additionally, most Pre-Tier 0 vehicles are vintage vehicles that are rarely seen on the road.

3. Methodology

3.1. Real driving emissions (RDEs) tests

An urban on-road 5.3-mile driving cycle was designed on the local road network within the City of Logan, Utah, on which to conduct the Real Driving Emissions (RDE) tests using the vehicles in the test sample. The test cycle was coded as UWRL-UDTC (The Utah Water Research Laboratory Urban Driving Test Cycle). The UWRL-UDTC is shown in Fig. 2. The same testing cycle was recently used by (Khader and Martin, 2019). The UWRL-UDTC included residential and highway roads with 25, 40, and 45 mph speed limit areas. The cycle had variable grades, including low and steep uphill and downhill road segments. The cycle had three traffic lights, and two 4-way stop signs, where vehicles had to come to a complete stop and yield to vehicles arriving first at the stop sign. The route also included a 2-way stop sign that requires vehicles to completely stop and yield to crossing traffic. Lastly, the cycle included many pedestrian crossings and a school zone with a lower speed limit

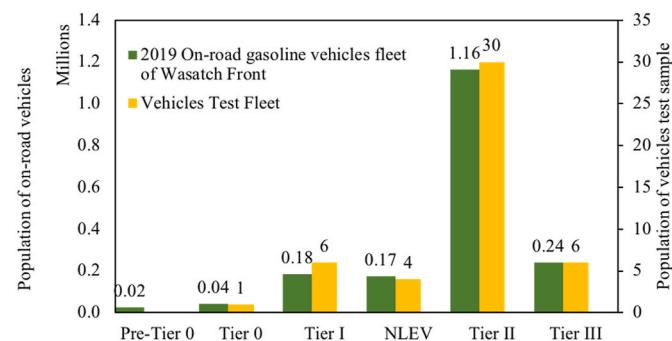


Fig. 1. Vehicular profile of the Wasatch Front and current study gasoline vehicles.

from 40 to 25 mph. Therefore, the calculated NH₃ emission rates represent the effect of many driving conditions, including stop-and-go, high-speed highway and low-speed urban driving, acceleration/deceleration, and uphill and downhill driving conditions. The RDEs tests started and ended at the Utah Water Research Laboratory (UWRL).

Tested vehicles were driven to the UWRL and then parked to be prepared for the RDE tests and to be equipped with the testing instruments. The engine of vehicles was turned off during the preparation process that took about 45 min. This may have allowed vehicles' engine and TWC converters to cool off a little bit before carrying out the first on-road RDE test. This might explain having higher NH₃ emission rates in the first lap than in the second and third laps (Supplementary 2). The preparation process included instrument calibration, installation on vehicles and warm-up, collection of vehicles specifications (type, make, model, model year, mileage reading, engine displacement, number of cylinders, gross vehicle weight rating (GVWR) and tailpipe diameter), documentation of vehicle's owner name and contact information, taking pictures of the tested vehicles and lastly, measurement of atmospheric temperature and pressure at the time of testing. Triplicate, consecutive RDE tests were conducted for all tested vehicles. The triplicate RDEs of each vehicle were only a few seconds apart from each other. The mean values of vehicle speed (m/s), engine revolution per minute (RPM), acceleration (m/s^2) and deceleration (m/s^2) of all tested vehicles were consistent over the consecutive triplicate laps (Supplementary 3). The RDE tests were conducted between January and September of 2020. The researcher drove the tested vehicles over the testing cycle only when vehicle owners were not available to drive the test cycle themselves. In those cases, the owners delivered their vehicles to the UWRL and gave permission to the researcher to drive the vehicles over the designed on-road urban testing cycle. Vehicle owners who participated in the on-road tests were accompanied by the researcher and were only asked to drive normally and adhere to traffic rules. It's worth mentioning that none of the tested vehicles had a check engine light illuminated on their dashboard at the time of testing, which suggests that their oxygen (lambda) sensors were not faulty.

3.2. Equipment

Ammonia and NO_x concentrations were measured in parts per million (ppm) using the portable ECM (Engine Control and Monitoring) miniPEMS. The ECM miniPEMS had two modules for measuring exhaust NH₃ and NO_x concentrations: NO_xCANT and NO_xCANf. The NO_xCANT and NO_xCANf modules were wired such that their ceramic exhaust emission sensors were mounted in the tested vehicle's engine exhaust (Fig. 3). The stainless-steel tube holding the sensors was inserted 5 inches inside the vehicle's tailpipe and extended for about 10 inches into the atmosphere to avoid ambient air interference. In addition to the NH₃ and NO_x exhaust emissions, carbon monoxide (CO) concentrations in vehicles exhaust were also measured using an Applus Autologic 5-Gas Portable Vehicle Gas Analyzer (model 310-0220) (Fig. 3). The concentrations of CO were measured in percentages and then multiplied by 10,000 to convert them into ppm. The 5-gas analyzer interfaced to laptop computers via serial ports to collect emissions data every second. The lag-time between the 5-Gas analyzer and the ECM miniPEMS was 6 s in favor of the ECM miniPEMS. The ECM miniPEMS was also used to retrieve various parameters data such as vehicle speed, the engine revolution per minute (RPM) readings, and engine load percentages from the vehicle's OBDII port using several OBD readers. The vehicle exhaust temperature was also measured by the ECM miniPEMS using Type K thermocouples. The ECM miniPEMS collected the concentrations of NH₃ and NO_x compounds as well as vehicle speed, engine RPMs and load and exhaust temperature information every 0.1 s. The data, however, were averaged over a 1-s time period.

The NO_xCANT sensor measured the concentration of both NH₃ and NO_x, whereas the NO_xCANf sensor measured only NO_x concentrations,

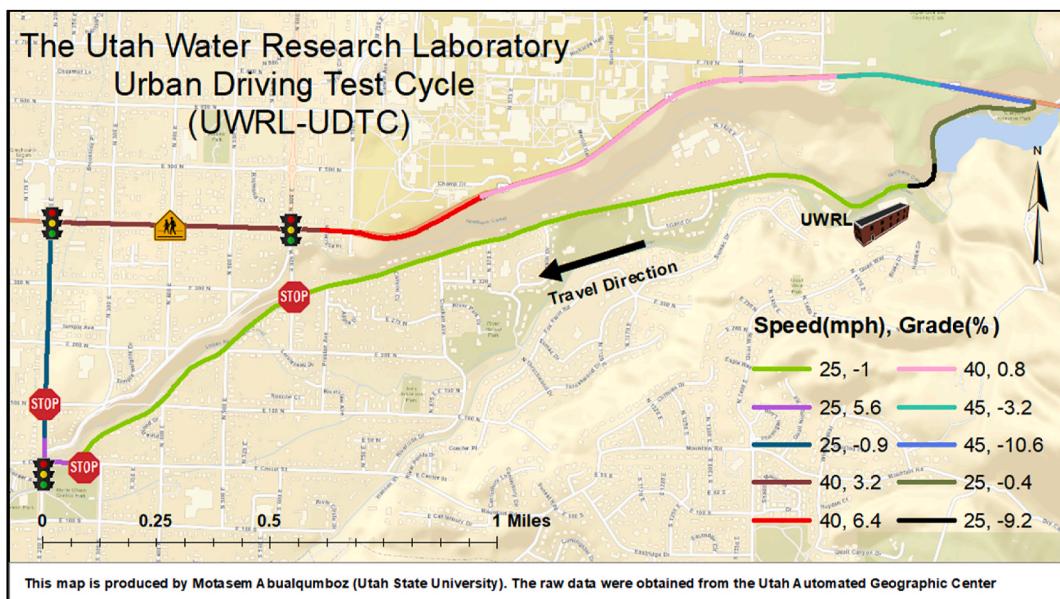


Fig. 2. The Utah water research laboratory urban driving test cycle (UWRL-UDTC).

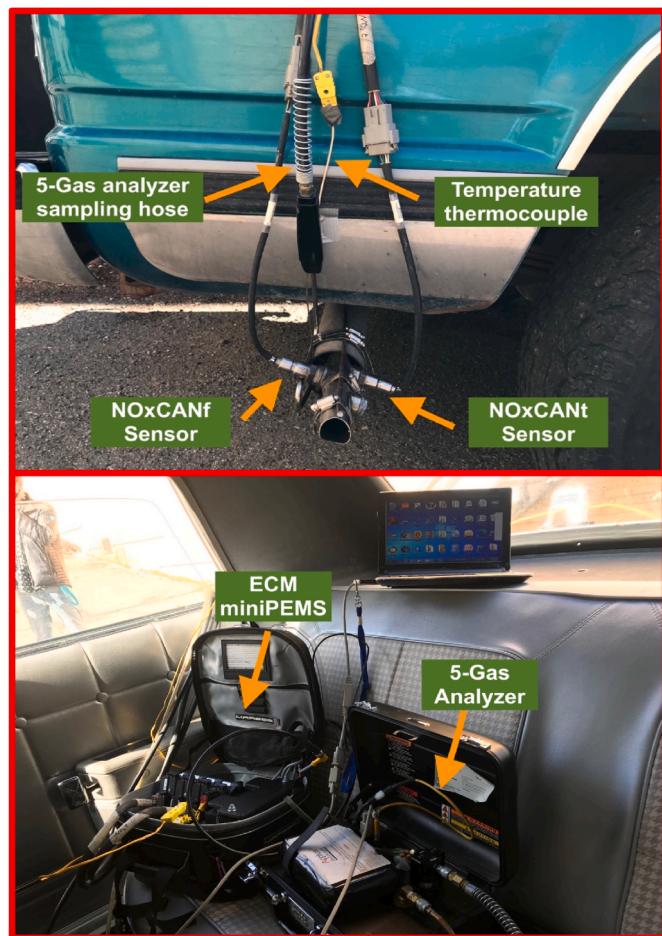


Fig. 3. The ECM miniPEMS and 5-Gas analyzer instruments.

as it was equipped with an acid filter to absorb NH₃ emissions. Ammonia concentrations were obtained by subtracting the NO_xCANf readings from the NO_xCANt readings. The acid filter had a high concentration of phosphoric acid (H₃PO₄). A new filter was used for each vehicle, despite

the fact that the ECM company recommendations stated that each filter could be used for more than one vehicle. Moreover, a couple of acid filters were tested after the on-road RDE tests were conducted to check that the H₃PO₄ acid was not totally consumed during the tests. The NO_xCANt and NO_xCANf modules instantaneously measure NH₃ and NO_x concentrations once the exhaust emissions touch their ceramic exhaust sensors. This overcomes the problem of NH₃ adsorption/desorption to sampling tubes and tunnels walls because of the sticky nature of ammonia. This problem was reported by several on-road tunnel studies including (Heeb et al., 2006, 2008; Mohn et al., 2004). The instantaneous measurement of NH₃ and NO_x concentrations also prevents the loss of these gases due to potential reactions with acids present in the ambient air or in emissions such as H₂SO₄ from on-road diesel motor vehicles (Pierson and Brachaczek, 1983; Trux et al., 1980). The effect of background ambient concentrations of ammonia reported particularly by on-road in-tunnel studies is also avoided by the raw, direct and instantaneous NH₃ measurement.

The NO_xCANt and NO_xCANf sensors are oxygen (O₂) pumping type sensors with two cavities. Exhaust emissions diffuse first into the 1st cavity where the O₂ pumping current (Ip1) is controlled to obtain a 0% oxygen condition in the cavity. The first cavity electrochemically pumps O₂ out of the exhaust emissions so it does not interfere with the NO_x measurement in the second cavity. The O₂ pumping current (Ip1) is proportional to O₂ concentration in the exhaust. Exhaust gases from the 1st cavity next diffuse into the 2nd cavity, where oxygen molecules are stripped from O₂ and NO_x compounds. The Ip2 current is proportional to the concentration of O₂ and NO_x in the cavity. Since the O₂ percentage is reduced to near zero in the 1st cavity, the Ip2 would largely be due to NO_x emissions. Hence, the Ip2 is used to determine the amount of NO_x compounds in the exhaust emissions, as it would be equivalent to their concentration. The NO_xCANt and NO_xCANf sensors were factory-calibrated and again calibrated at the UWRL using a certified NH₃ standard calibration gas from Airgas Specialty Gases (Fig. 4). The ECM miniPEMS was also frequently calibrated against the bench-scale Picarro cavity ringdown spectrometer (Model G2103) (Fig. 4). The Picarro base station NH₃ analyzer is a reliable and accurate instrument and is usually calibrated using a couple of standard gases. Additionally, the Excel sheets obtained from the ECM miniPEMS were checked for error codes that may have recorded during the RDEs tests. Some parameters such as vehicle speed were also obtained from the ECM miniPEMS and vehicles OBDII and compared together. Similar to the ECM miniPEMS, the 5-Gas

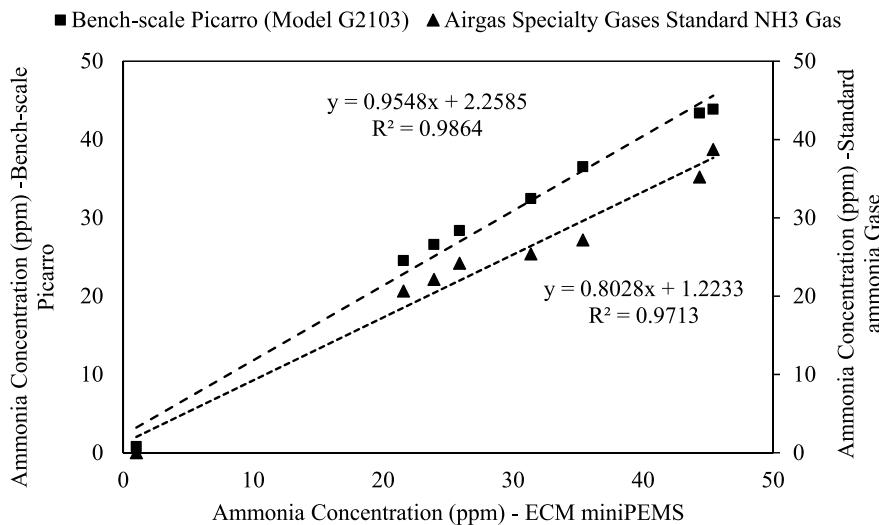


Fig. 4. ECM miniPEMS calibration using the bench-scale Picarro (Model G2103) ammonia analyzer and the Airgas Specialty Gases standard NH₃ gas.

analyzer was calibrated frequently using a certified blend gas from Airgas Specialty Gases.

3.3. Calculation of gases emission rates (mg/mile)

3.3.1. The 1-s averaged gases mass concentrations (mg/m³)

The 1-s averaged tailpipe mixing ratios (ppm) obtained from the ECM miniPEMS and the 5-Gas analyzer were converted to mass concentrations (g/L) using the ideal gas law shown in Eq. (4). The ideal gas constant (R) was taken as 62.36 mmHg-L/gmol-K, with the atmospheric pressure measured at the time of each test using a mercury barometer placed inside the Air Quality Lab at the UWRL. The exhaust temperature was the 1-s averaged exhaust temperature values that were measured during the ~600-s on-road RDE tests using a Type K thermocouple (Fig. 3). The calculated mass concentrations were multiplying by 10⁶ to convert them from grams per liters (g/L) to milligrams per cubic meters (mg/m³).

$$\text{Mass concentration } \left(\frac{\text{g}}{\text{L}} \right) = \frac{\text{Tailpipe mixing ratio (ppm)} \times \text{MW} \left(\frac{\text{g}}{\text{mol}} \right) \times \text{Atmpressure (mmHg)}}{\text{Exhaust temprature (K)} \times \text{Ideal gas constant (R)} (\text{mmHg-L/gmol-K}) \times 10^6} \quad \text{Eq. 4}$$

3.3.2. The volume of exhaust emission (V_{total})

The 1-s averaged exhaust mass flow rates (\dot{m}_{Exhaust}) were estimated using the mass balance approach that sums the mass flow rate of intake air (\dot{m}_{Air}) and the mass flow rate of fuel ($\dot{m}_{\text{air}}/\text{Air-Fuel-Ratio}$) (Eq. (5)). This method has been used by several studies including (Bodisco et al., 2019; Prakash and Bodisco, 2019; Yang et al., 2021). The exhaust emission volume (V_{Total}) was then calculated from the exhaust mass flow rate (\dot{m}_{Exhaust}) using the ideal gas law. The calculated mass concentrations using Eq. (4) were multiplied by the exhaust emission volume (V_{Total}) to get the mass (mg) of each pollutant. The masses calculated at all seconds of the ~600-s on-road RDE tests were then summed and divided by the length of the UWRL-UDTC, 5.3 miles, to get the emission rates (mg/mile) for all of the gases. The data were then analyzed using the RStudio statistical tools.

$$\dot{m}_{\text{exhaust}} \approx \dot{m}_{\text{air}} + \frac{\dot{m}_{\text{air}}}{\text{Air - Fuel - Ratio}} \quad \text{Eq. 5}$$

As shown in Eq. (6) (Ahmadipour et al., 2019; Cai et al., 2014; Katsoyiannis, 2014; Katsoyiannis et al., 2012), the flow rate of the

engine's intake air ($Q_{\text{Intake Air}}$) corresponding to each of the 1-s averaged RPM readings was estimated using vehicles' engine displacement and the 1-s averaged engine speed (RPM) that were obtained during the ~600-s on-road RDE tests. The flow rates were then converted from cubic feet per minute (ft³/min) to cubic meters per second (m³/s) flow rates. The volumetric efficiency was assumed as 85% as recommended by (Sandhu and Frey, 2013; Yang et al., 2021) when it can be measured directly. The flow rate of engine's intake air ($Q_{\text{Intake Air}}$) was then converted into mass flow rates (\dot{m}_{Air}) using the ideal gas law. The atmospheric pressure and temperature were measured at the time of each test. The air-to-fuel ratios were measured during the ~600-s on-road RDE tests by the ECM miniPEMS. The raw data files and all the calculations and data analyses can be found in the Jupyter notebook included in the online resource (Abualqumboz et al., 2022).

$$\begin{aligned} Q_{\text{Intake Air}} \left(\frac{\text{ft}^3}{\text{min}} \right) &= \\ \text{Engine Displacement (inch}^3\text{)} \times \text{Engine Speed (RPM)} \times \text{Volumetric Efficiency (\%)} \\ &= 3456 \end{aligned} \quad \text{Eq. 6}$$

4. Results and discussion

4.1. Vehicle test fleet

A total of 145 on-road RDE tests were conducted using 47 LD gasoline motor vehicles with GWVR up to 12,000 pounds. The characteristics and the emission rates of NH₃, NOx and CO of tested gasoline motor vehicles are shown in Table 1. The characteristics included vehicle type, make, model, model year, EPA Emission Standard Tier, engine size, number of cylinders, vehicle GVWR and odometer reading. The 5-Gas analyzer did not produce useable data during the RDE tests of four vehicles (Vehicles # 9, 13, 14 and 34 in Table 1). The data in Table 1 also show that the on-road RDE tests were repeated for the 2007 RAM 1500 Dodge Light-Duty Truck (LDT) and the 2006 Matrix Toyota Passenger Car (PC). The observed variability in NH₃ emission rates between the repeated tests was statistically insignificant at the 95% confidence level, which may highlight good repeatability in the testing procedure. The p-value of the t-test that measured the difference in NH₃ factors for the 2007 Dodge tests was 0.27. This is larger than the tail area probability of the 95% confidence level of $\alpha = 0.05$. Thus, NH₃ emission rates of the two repeated tests are considered as one group statistically. The repeated tests of the 2006 Matrix Toyota were also treated as one group,

Table 1

Characteristics of tested gasoline motor vehicles and their post-catalyst NH₃, NO_x, CO, HC and CO₂ emission rates (mg/mile). PC = Passenger Car, MPV = Multi-Purpose Vehicle, LDT = Light-Duty Truck. NLEV = National Low Emission Vehicle.

NO	Driver	Type	Make	Model	Year	Emission Standard Tier	Engine Displacement (L)	GVWR (US lbs.)	Mileage (mile)	NH ₃	NO _x	CO
1	Researcher	LDT	Chevrolet	CK 1500	1993	Tier 0	5.7	8	284,117	563.1	797.2	23134.4
2	Researcher	PC	Subaru	Legacy	1995	Tier I	2.2	4	161,603	275.1	209.8	6846.7
3	Owner	LDT	Ford	Ranger XLT	1997	Tier I	2.3	4	4740	184,545	122.6	312.4
4	Researcher	MPV	Pontiac	Trans Sport	1998	Tier I	3.4	6	5357	200,624	201.0	408.3
5	Researcher	MPV	GMC	Suburban	1999	Tier I	5.7	8	7300	160,714	277.1	536.3
6	Owner	PC	Honda	Accord	2000	Tier I	3	6	4235	188,405	126.3	61.9
7	Owner	PC	Mercedes-Benz	SLK 230	2000	Tier I	2.3	4	3485	171,099	65.0	29.5
8	Owner	MPV	Chevrolet	Tahoe	2002	NLEV	5.3	8	6900	267,031	415.1	586.1
9	Owner	MPV	Honda	CR-V	2003	NLEV	2.4	4	4320	249,362	16.8	34.7
10	Researcher	PC	Nissan	Maxima	2003	NLEV	3.5	6	4295	221,711	404.9	232.8
11	Owner	PC	BMW	325i	2003	NLEV	2.5	6	4365	122,918	17.8	3.1
12	Owner	PC	Hyundai	Elantra	2004	Tier II	2	4	3880	137,724	70.3	21.5
13	Owner	PC	Subaru	Forester	2005	Tier II	2.5	4	4150	150,872	43.7	26.7
14	Owner	PC	Toyota	Matrix XR	2006	Tier II	1.8	4	3845	124,689	12.7	6.1
										124,817	30.4	36.0
15	Owner	LDT	Dodge	RAM 1500	2007	Tier II	5.7	8	6700	214,484	100.7	136.9
										216,506	128.5	136.3
16	Owner	MPV	Nissan	Pathfinder	2008	Tier II	4	6	6113	166,095	506.3	145.1
17	Owner	PC	Nissan	Sentra	2008	Tier II	2	4	3920	134,613	117.5	45.3
18	Owner	MPV	Chrysler	Grand Caravan	2009	Tier II	3.3	6	6050	40,680	65.4	12.3
19	Owner	MPV	Honda	Odyssey	2009	Tier II	3.5	6	5941	136,262	39.5	20.1
20	Researcher	MPV	Chrysler	Town & Country	2010	Tier II	3.8	6	6050	63,570	59.6	113.5
21	Researcher	PC	Hyundai	Sonata	2011	Tier II	2.4	4	4299	57,976	48.1	18.3
22	Researcher	LDT	Chevrolet	Colorado LT	2011	Tier II	3.7	5	5300	33,320	48.2	49.3
23	Owner	PC	Chevrolet	Malibu Lt	2011	Tier II	2.4	4	4419	192,228	200.1	16.7
24	Owner	PC	Subaru	Legacy	2012	Tier II	2.5	4	4435	165,808	256.6	34.0
25	Owner	PC	Nissan	Versa	2012	Tier II	1.6	4	3388	104,870	112.1	74.9
26	Owner	MPV	Kia	Sedona	2012	Tier II	3.5	6	5853	136,804	326.7	86.1
27	Owner	LDT	Ford	F 150	2012	Tier II	5	8	7350	126,928	216.4	136.1
28	Researcher	PC	Chevrolet	Impala	2012	Tier II	3.6	6	4564	30,145	20.9	8.3
29	Researcher	MPV	Chevrolet	Traverse	2012	Tier II	3.6	6	6459	53,982	73.2	13.2
30	Researcher	LDT	Chevrolet	Colorado LT	2012	Tier II	2.9	4	5000	13,602	47.3	28.2
31	Researcher	MPV	Toyota	Highlander	2013	Tier II	3.5	6	6000	50,432	12.3	13.0
32	Researcher	LDT	Toyota	Tacoma	2013	Tier II	4	6	5500	67,634	22.9	13.7
33	Researcher	LDT	Chevrolet	Silverado	2013	Tier II	4.8	8	6400	19,747	151.0	20.8
34	Researcher	MPV	Toyota	FJ Cruiser	2013	Tier II	4	6	5570	92,976	13.5	9.7
35	Owner	PC	Chrysler	200 S	2014	Tier II	2.4	4	4600	93,642	75.0	119.1
36	Owner	MPV	Nissan	Quest	2015	Tier II	3.5	6	5818	45,238	56.1	32.7
37	Researcher	LDT	Chevrolet	Colorado	2015	Tier II	2.5	4	5400	16,564	39.3	16.7
38	Owner	PC	Toyota	Corolla	2015	Tier II	1.8	4	3820	125,429	9.2	21.7
39	Researcher	MPV	Chevrolet	Equinox	2015	Tier II	2.4	4	5070	15,769	41.8	3.7
40	Owner	MPV	Jeep	Wrangler	2016	Tier II	3.6	6	4900	14,107	17.4	12.8
41	Owner	LDT	Chevrolet	Colorado	2016	Tier II	3.6	6	6000	15,536	59.1	18.6
42	Owner	LDT	Toyota	Tacoma	2018	Tier III	3.5	6	5600	5654	37.8	10.1
43	Researcher	PC	Subaru	Legacy	2018	Tier III	2.5	4	4519	33,374	7.9	4.3
44	Owner	MPV	Subaru	Cross Trek	2019	Tier III	2	4	4343	9365	15.8	2.7
45	Owner	LDT	Toyota	Tacoma	2019	Tier III	3.5	6	5600	3618	23.9	23.2
46	Owner	MPV	Chrysler	Pacifica	2019	Tier III	3.6	6	6055	8006	10.3	11.5
47	Owner	LDT	Ford	Ranger	2020	Tier III	2.3	4	6050	3439	17.7	3.8

as the *t*-test p-value of 0.43 was also higher than 0.05.

4.2. Ammonia emission rates of the entire vehicles test sample

A summary of averaged NH₃ emission rates from tested gasoline motor vehicles is presented in Fig. 5. The data show that NH₃ emission rates for the entire fleet averaged 114.7 ± 135.3 (StD) mg/mile and ranged between 7.9 and 563.1 mg/mile.

The estimated EPA NH₃ emission rates for LD gasoline motor vehicles range from 1.6 mg/mile to 516.6 mg/mile and have an average of 101.4 mg/mile (Huai et al., 2003). Roe et al. (2004) reported that the EPA recommended NH₃ emission rates obtained from the MOBILE6.2 Motor Vehicle Emissions Factor Model for LD catalyst-equipped gasoline motor vehicles is 101.7 mg/mile. The average NH₃ emission rates estimated in this study (114.7 mg/mile) is higher than the average EPA estimated NH₃ emission rate of 101.4 mg/mile by only 12%. The mean NH₃

emission rate estimated in this study was also consistent with previously measured NH₃ emission rates, which ranged from 37.5mg/mile (Czerwinski et al., 2016) to 132.8 mg/mile (Durbin et al., 2002) (Supplementary 4). The average emission rate of 114.7 mg/mile was also consistent with the outcomes of (Huang et al., 2018) who used the EPA's MOVES (MOtor Vehicle Emission Simulator) model to estimate ammonia emission rates for gasoline vehicles made in 1992, 1996, 2000, 2005 and 2010. The model estimated that NH₃ emission rates would range from 19 to 114.3 mg/mile.

The standard deviation and the 95% confidence level of NH₃ emission rates for the entire fleet were 135.3 and ± 38.9 mg/mile, respectively. The wide range of 555.2 mg/mile and the high standard deviation suggest high variability in NH₃ emission rates among the entire fleet. This is likely due to differences in vehicles characteristics and in NH₃ precursor concentrations, as clearly illustrated in Table 1. For instance, the minimum NH₃ averaged emission rate of 7.9 mg/mile was recorded

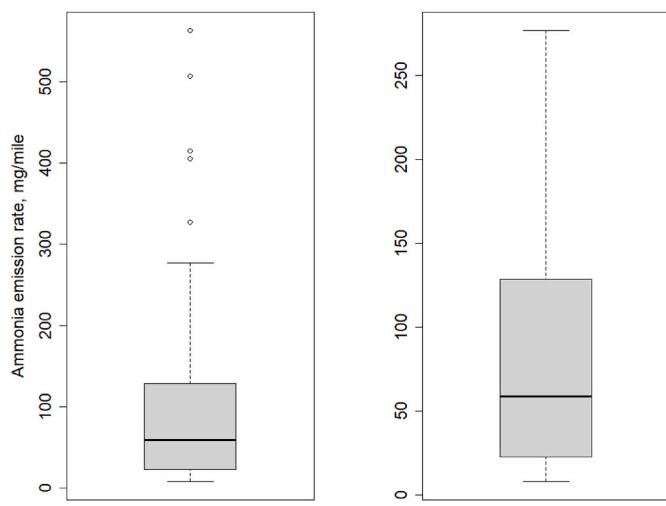


Fig. 5. Boxplot of NH₃ emission rates for the entire fleet of tested gasoline motor vehicles with and without displaying the extreme values.

for a Tier III PC vehicle (Vehicle # 45) that was driven for only 33,347 miles, whereas the maximum NH₃ averaged emission rate of 563.1 mg/mile was measured for a Tier 0 Light-Duty Truck vehicle (Vehicle # 1) that had an odometer reading of 284,117 miles. The Tier III PC vehicle also recorded low emission rates of NO_x (4.3 mg/mile) and CO (19 mg/mile) as compared with the Tier 0 Light-Duty Truck vehicle that had comparatively high NO_x (797 mg/mile) and CO (23,134 mg/mile) emission rates. The data also showed that, although the maximum NH₃ emission rate was 563.1 mg/mile, the 75th percentile was 128.5 mg/mile. Moreover, 83.7% of all the emission rates were within one standard deviation from the mean emission rate of 114.7 mg/mile. This indicates that most vehicles were linked with comparatively small NH₃ emission rates, and only a few vehicles recorded high emissions rates, as clearly shown in the right-skewed histogram plotted in Fig. 6. Most NH₃ emission rates appeared to be below 100 mg/mile.

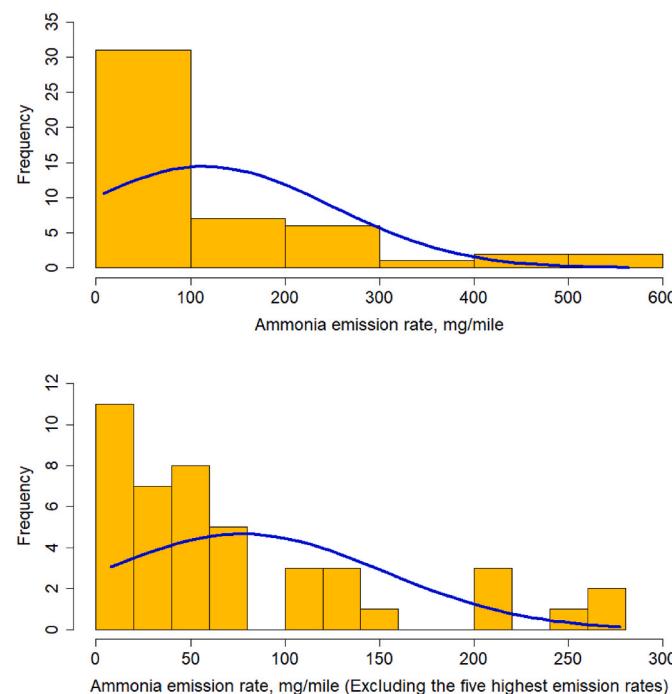


Fig. 6. Histogram of NH₃ averaged emission rates for the entire fleet of gasoline vehicles.

Although most NH₃ emission rates were less than 100 mg/mile, the histogram shown in Fig. 6 also shows that some elevated NH₃ averaged emission rates were also recorded. This is also supported by the boxplot shown in Fig. 5, which shows five extreme (very high) NH₃ emission rates. These extreme NH₃ emission rates were not within 1.5 times the interquartile range (75th percentile– 25th percentile) from the 75th percentile. The 25th and 75th percentiles were 22.9 and 128.5 mg/mile, respectively. The five extreme NH₃ emission rates of 326.7, 404.9, 415.1, 506.3 and 563.1 mg/mile were measured for Tier II (Vehicle #28), NLEV (Vehicle #10), NLEV (Vehicle # 8), Tier 11 (Vehicle #18) and Tier 0 (Vehicle #1) motor vehicles, respectively. Similar outcomes obtained by (Durbin et al., 2002), who also reported that the highest NH₃ emission rates were measured from Tier 0, Tier I and TLEV vehicles. Czerwinski et al. (2016), Durbin et al. (2004) and Huai et al. (2003) also reported that NH₃ emissions from vehicles with aged catalysts were higher than NH₃ from vehicles with newer catalysts.

The five elevated NH₃ averaged emission rates were all reported for old vehicles with aged catalysts. This supports the hypothesis that high-mileage old vehicles are responsible for higher emission rates of ammonia than newer vehicles. For instance, the vehicles with the first (Vehicle #1), third (Vehicle #8) and fourth (Vehicle #10) highest NH₃ emission rates had mileage readings greater than 250,000 miles, and the mileage readings from the vehicles with the second (vehicle #18) and fifth (Vehicle #28) highest NH₃ emission rates were 166,095 and 136,804 miles, respectively. These five vehicles with the highest NH₃ emissions also recorded high emission rates of NO_x and CO. In fact, some of these five vehicles violated the U.S. EPA emission standard for CO (3400 mg/mile for Tier 0, Tier 1 and NLEV vehicles and 4200 mg/mile for Tier II vehicles) and NO_x (1000, 200 and 70 mg/mile for Tier 0, NLEV and Tier II vehicles, respectively), as clearly shown in Table 1. Therefore, it is likely that these vehicles were running with aged TWC converters that no longer efficiently controlled ammonia precursors, including CO and NO_x. There was no way to determine if the TWC converters were replaced during the vehicle's lifetime. Original converters on motor vehicles are usually designed to last for the life of the vehicle only if they are properly used and well maintained. However, TWC converters are expected to be replaced due to reduced performance after approximately 100,000 miles of service for older vehicles (Kidd et al., 2006). Removing the five vehicles that recorded the extreme emission rates from the entire test fleet would reduce the mean NH₃ emission rate by 32.6% from 114.7 mg/mile to 77.4 mg/mile. This obviously suggests that NH₃ emissions from the on-road gasoline fleet are largely produced by a small fraction of very high emitting aged-catalyst vehicles. Hence, limiting the number of on-road old gasoline vehicles with high mileage readings would significantly lower the total NH₃ emissions from the Wasatch Front on-road gasoline motor vehicle fleet. Alternatively, the authorities can encourage drivers and vehicles owner to replace their TWC converters based on vehicles' manufacturers recommendations. The authorities could also carry out inspection and maintenance programs that aim to check the performance of vehicles TWC converters to reduce ammonia exhaust emissions from the on-road vehicles fleets.

4.3. Ammonia emission rates for vehicles of the same tier level

The NH₃ emission rates from gasoline vehicles of the same emission standard are shown in Fig. 7. The variability of NH₃ emission rates within each tier level, especially for Tier I and NLEV vehicles, could be due to unique vehicles characteristics and differences in ammonia precursor concentrations. The fact that NH₃ exhaust emission rates are not regulated as of yet could also explain the variable emission rates of NH₃ for vehicles with similar emissions standards (Durbin et al., 2002). The standards are arranged in the plot in order of increasing stringency for tailpipe emissions of CO and NO_x. Note that the emissions technology category might be confounded with the mileage factor, as old vehicles (Tier 0, Tier I and NLEV) generally have higher mileage than newer

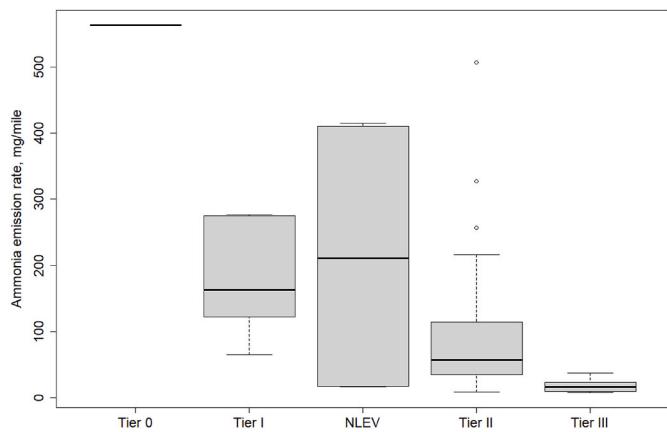


Fig. 7. NH₃ emission rates of each tier level vehicles.

vehicles (Tier II and Tier III). The mean NH₃ averaged emission rates as a function of vehicle emissions technology were as follows: 563.1 mg/mile for Tier 0 vehicles, 177.8 mg/mile for Tier I vehicles, 213.6 mg/mile for NLEV vehicles, 94.4 mg/mile for Tier II vehicles, and 18.9 mg/mile for Tier III vehicles. In general, NH₃ emission rates had a decreasing trend as the standards increased in stringency towards limiting ammonia precursor compounds. A similar general trend of decreasing ammonia emissions as the emissions standards increased in stringency was also observed by (Durbin et al., 2002; Livingston et al., 2009). Tier II and Tier III vehicles were linked with lower NH₃ emission rates than Tier 0 and Tier I vehicles. The NLEV vehicles, however, had higher NH₃ emission rates than the Tier I vehicles. This could be explained by the fact that the NLEV tier emission standard extended only to lighter motor vehicles and didn't include vehicles with a GVWR larger than 6000 pounds. Based on that, the 2002 MPV Chevrolet Tahoe (GVWR >6000 lbs.), which had the highest NH₃ factor among other NLEV vehicles, was subjected to less stringent emission standards than other NLEV vehicles. Also, the NLEV and Tier I vehicles have the same CO emissions standard. A similar trend was also observed by (Livingston et al., 2009), who showed that vehicles

made between 2001 and 2003 (California classification: Transitional Low-Emission Vehicle (TLEV)) unexpectedly had higher NH₃ emission rates than Tier I vehicles.

The outcomes of the ANOVA test (Supplementary 5) show that the differences in NH₃ emission rates among the vehicles of different tier levels are statistically significant at the 95% confidence level. The F-statistic value of 6.69 is larger than the upper 95% point of the F distribution with 4 and 44 degrees of freedom ($F_{0.95,4,44}$) of 2.594. This clearly indicates that there is more significant differences between the different groups (Tiers) than within the groups (Berthouex and Brown, 2002). The $\text{Pr}(>F)$ value of 2.7×10^{-4} also indicates that the differences in NH₃ emission rates among the vehicles of different tier levels are statistically significant at the 95% confidence level. This is because its significantly smaller than the tail area probability of the 95% confidence level of $\alpha = 0.05$. All this clearly suggests that gasoline vehicles of different tier standards have significantly different NH₃ emissions rates. The significant difference in NH₃ emissions rates among gasoline vehicles of different tier is likely due to differences in NH₃ precursors, including NO_x and CO, vehicle characteristics, odometer readings, and vehicle model years. Fig. 8 clearly shows that post-catalyst CO and NO_x exhaust emission rates and mileage readings for each tier level had the same general trend as that of NH₃ emission rates. Specifically, the Tier II and Tier III vehicles had lower CO and NO_x emission factors and mileage readings than Tier 0, Tier I, and NLEV vehicles. The odometer reading is a good proxy of catalyst age and vehicle model year (Durbin et al., 2004). showed that bench-aged catalysts produced 12% higher CO ammonia precursor emissions than as-received catalysts. Lower NO emissions (g/kg fuel) were also measured for newer vehicles in a study in Tulsa, Oklahoma and Denver, Colorado in 2005 by (Burgard et al., 2006). Higher NH₃ emission rates were measured for bench-aged catalysts than as-received catalysts by (Durbin et al., 2004; Huai et al., 2003). Hence, the significant difference in NH₃ emissions rates among gasoline vehicles of different tier levels is likely due to the fact that vehicle tier level is defined based on vehicles age (Model Year -MY), which seems to play an important role in NH₃ production rates from gasoline motor vehicles. Vehicles age/Model year is a good indication of the age of fitted TWC converters.

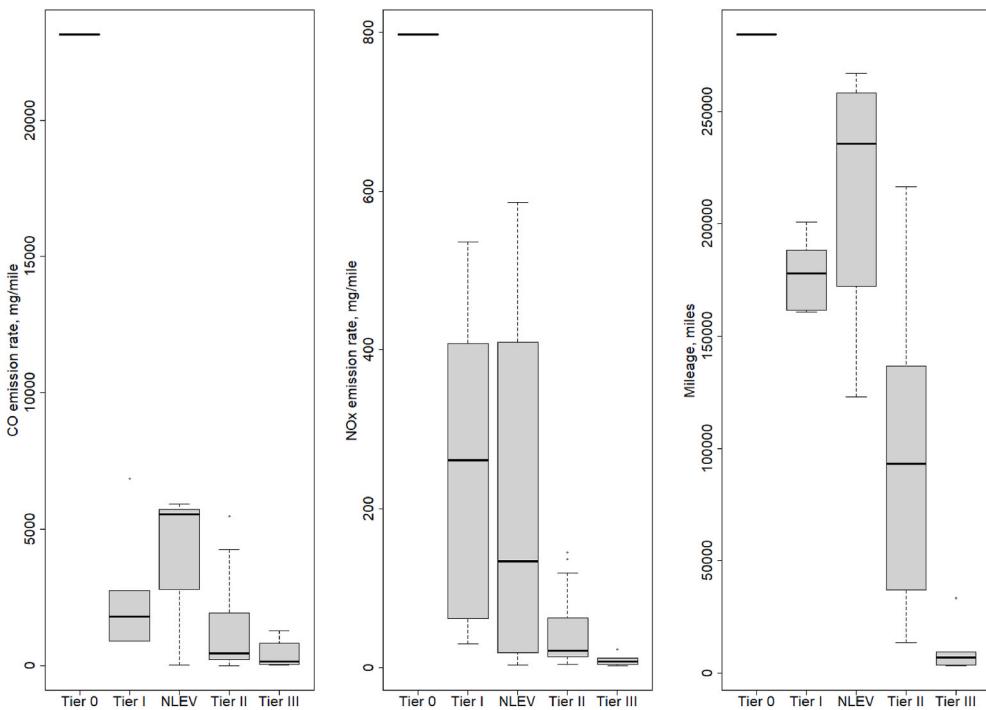


Fig. 8. Vehicles' Mileage and post-catalyst emission rates of ammonia precursors, CO and NO_x for each vehicle tier level.

4.4. Effect of vehicle's characteristics and post-catalyst exhaust gases

The effect of vehicle type and make factors on NH_3 emission rates was examined. The outcomes of the ANOVA tests showed that none of these factors had a statistically significant impact on NH_3 exhaust emission rates for gasoline motor vehicles at the 95% confidence level. This could be explained by the fact that, regardless of vehicle manufacturer, all were equipped with similar TWC converters that are the main cause of NH_3 exhaust emissions. Additionally, all national and international manufacturers follow the same emission regulations applicable in the United States. As for vehicle type, gasoline motor vehicles are classified into PC, MPV, and LDT primarily based on their weight, which appeared to have little impact on NH_3 exhaust emissions, as shown in Fig. 9(e). The Pearson's correlation coefficient (r) of 0.3 clearly indicates a small correlation with NH_3 emission rates. The Pearson's correlation coefficient measures the strength of the linear relationship between two variables. A correlation coefficient of $+0.7 = r \leq +1$ and $-1.0 = r \leq -0.7$ suggests a strong association between two variables, whereas a correlation coefficient of $+0.4 = r < +0.7$ and $-0.7 < r \leq -0.4$ suggests a moderate correlation. A correlation coefficient of $0 = r < +0.4$ and $-0.4 < r \leq 0$ suggests a weak correlation between tested variables (Dancey and Reidy, 2007; Ratner, 2009).

The data also shows that vehicle model year (Fig. 9(a)) and mileage (Fig. 9(b)) factors had a moderate correlation with NH_3 emission rates,

as the Pearson's correlation coefficient (r) indicates. The mileage factor had a stronger correlation with NH_3 emission rates than the model year factor, with a correlation coefficient value of $r = 0.6$. This clearly indicates that NH_3 emission rates increase with increased mileage for gasoline vehicles. These outcomes are consistent with (Bishop et al., 2010; Bishop and Stedman, 2015; Durbin et al., 2004), each of whom measured higher NH_3 emission rates from aged catalysts than from low-mileage catalysts. The negative moderate correlation ($r = -0.5$) between the vehicle model year factor and NH_3 emission rates also supports the idea that gasoline motor vehicles with long-used TWC converters emit NH_3 at a higher rate than vehicles with well-operating TWC converters. The model year and odometer readings are good indications of catalyst's age.

The correlation plots also show that vehicle engine displacement (Fig. 9(c)) factor had a moderate impact on NH_3 emission rates for gasoline motor vehicles ($r = 0.4$). The number of cylinders (Fig. 9(d)) factor also showed moderate correlation with ammonia emission rates with a correlation coefficient value of 0.4. The moderate correlation between NH_3 emission rates and both engine displacement and number of cylinders factors suggest that vehicles with comparatively high fuel combustion produce higher amounts of NH_3 exhaust due to higher concentrations of ammonia precursors. A moderate linear correlation was measured between engine size and post-catalyst CO (Fig. 9(h)) and NO_x (Fig. 9(i)) exhaust emissions rates with a Pearson's correlation

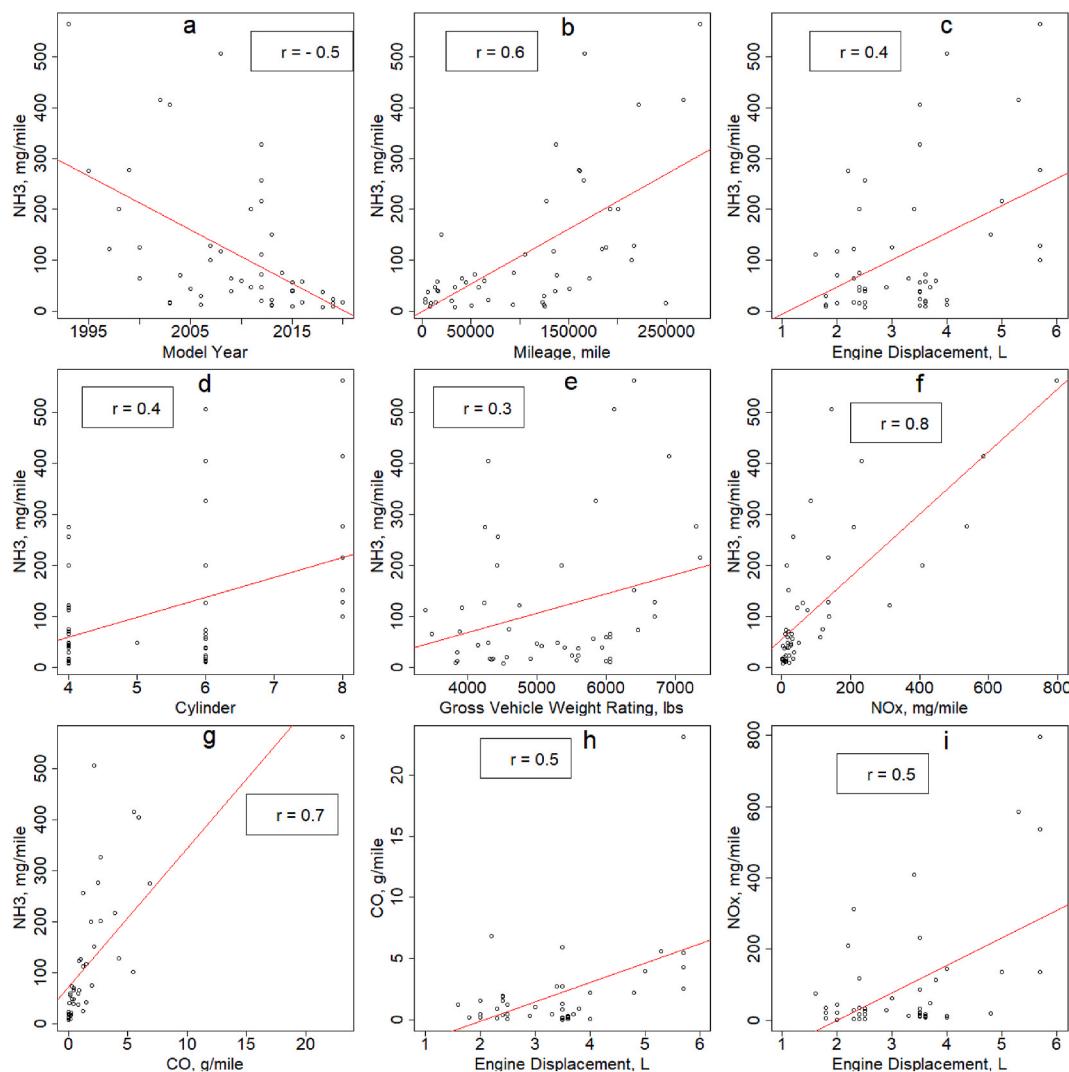


Fig. 9. Correlation between NH_3 emission rates and the vehicle characteristics and post-catalyst exhaust gases.

coefficient of 0.5. Engine displacement and number cylinders are good indications of the fuel volume combusted inside a vehicles' engine. Large engines with a large displacement volume usually produce larger volumes of total exhaust emissions than vehicle engines with smaller displacement. Similarly, the more cylinders in an engine, the more combustion occurs, and the more exhaust emissions production would occur. These larger volumes of exhaust emissions normally include NH₃ precursors that would result in higher NH₃ emissions. However, it should be noted that some vehicles with large engine displacement such as Tier II LDT trucks with GVWR above than 3750 pounds would record high ammonia emissions rates because they are certified to higher ammonia emissions standards.

The correlation plots presented in Fig. 9 also demonstrate that NH₃ emission rates had a strong correlation with post-catalyst CO exhaust emissions, as indicated by the correlation coefficient of $r = 0.7$. This is consistent with many previous studies, including (Czerwinski et al., 2016; Kean et al., 2009; Livingston et al., 2009). The scatter plots also show that a strong correlation was similarly measured between NH₃ and NO_x post-catalyst emissions ($r = 0.8$). The relationship between mean NH₃ emissions and NO_x emissions follows the trend reported in the literature (Bishop et al., 2010; Burgard et al., 2006). A correlation matrix (Supplementary 6) was also produced to visualize and explore the relationships among all the variables shown in Fig. 9. For instance, the plot showed that vehicle's model year was also correlated with CO and NO_x post-catalyst exhaust emissions with a Pearson's correlation coefficient of -0.6 and -0.7 , respectively. The mileage reading factor was strongly correlated with vehicle's model year ($r = -0.8$) and vehicle's technology ($r = -0.7$). The scatter plot matrix also illustrated that the engine displacement factor was strongly correlated with vehicle GVWR and number of cylinders factors with a correlation coefficient of 0.9. The measured cross-correlation among different variables may indicate that the impact of some variables is confounded with the impacts of other factors. The control for such confounding variables impacts would have resulted in extremely small vehicles sample sizes. The selection of specific vehicles with specific specifications may comprise the statistical robustness of obtained results (Livingston et al., 2009).

4.5. Comparisons with Wasatch Front emissions inventory

The vehicle test sample for this study had the same tier-level distribution as the on-road gasoline motor vehicle fleet along the Wasatch Front in the US State of Utah. The model years for the vehicles of the same tier level were not pre-defined. The research team randomly secured and tested motor vehicles until the total number of each Tier level was complete. As a result, NH₃ emission rates derived from this study are likely to be a good representation of NH₃ emissions for the Wasatch Front on-road gasoline motor vehicles fleet. According to the Office of Highway Policy Information (OHPI) in the U.S. Department of Transportation Federal Highway Administration (FHWA), motor vehicles in the State of Utah are usually driven approximately 13,884 miles per year. Assuming that this applies to on-road gasoline motor vehicles, the 1,826,584 on-road gasoline motor vehicles driven along the Wasatch Front drive for approximately 25.36×10^9 miles every year. This would yield total NH₃ emissions of 2909 metric tons per year from the entire gasoline-powered fleet. Ammonia emissions from diesel motor vehicles would contribute smaller amounts of ammonia emissions due to their small number and the fact that they emit comparatively smaller rates of ammonia than gasoline motor vehicles because they do not have TWC converters.

Previous studies including (Baum et al., 2001; Burgard et al., 2006; Durbin et al., 2002; Emmenegger et al., 2004; Farren et al., 2020; Karlsson, 2004) have reported that ammonia emission rates from the transport system seem to be greater than what emission inventories indicate. That assertion is also supported by the results of this study, which estimate the Wasatch Front gasoline motor vehicle fleet emits nearly 8.0 tons of NH₃ every day, three and half times the estimates from

the 2014 national emission inventory, which estimated that the Wasatch Front gasoline motor vehicles fleet emits 2.3 tons of NH₃ into the atmosphere every day. Compared with other sources, the Wasatch Front gasoline motor vehicle fleet would emit the second highest amount of NH₃ after agricultural area sources. The 2014 national emission inventory showed that agricultural sources constitute the largest fraction of NH₃ emissions at approximately 18.6 tons per day (Baasandorj et al., 2018). This is more than double the contribution of the Wasatch Front gasoline motor vehicle fleet at 8.0 tons of NH₃ every day.

5. Conclusion

Ammonia exhaust emission rates from 47 light-duty gasoline motor vehicles were quantified using a portable ECM miniPEMS over on-road Real Driving Emissions (RDE) tests. The RDE tests were conducted on a predefined testing route designed using the local road network in the City of Logan, Utah. A portable Applus Autologic 5-Gas Portable Vehicle Gas Analyzer (model 310-0220) was also used to measure concentrations of post-catalyst carbon monoxide. Both instruments were carried onboard the tested vehicles during the test, while their sensors were mounted in the tested vehicle's engine exhaust. The vehicle test sample of 47 light-duty gasoline motor vehicles were chosen to represent the same tier-level distribution as the on-road gasoline vehicle fleet along the Wasatch Front located in the U.S. State of Utah. Unlike early studies that expected NH₃ exhaust emissions from motor vehicles to remain insignificant, even if all gasoline motor vehicles were equipped with TWC converters, the outcomes of this study show that ammonia emissions originating from exhaust might be comparable to NH₃ emissions from natural sources, especially in big cities and urban areas where motor vehicles are predominant.

A total of 145 on-road RDE tests were carried out to estimate the NH₃ emission rate of the on-road gasoline vehicle fleet along the Wasatch Front in the U.S. State of Utah. The outcomes showed that the tested vehicles had an average ammonia emission rate of 114.7 mg/mile ± 135.3 (StD). The study also showed that NH₃ emission rates for the tested motor vehicles varied significantly according to the different vehicle characteristics. Older motor vehicles (Tier 0, Tier 1 and NLEV) with high odometer readings recorded significantly higher emission rates of NH₃ than newer motor vehicles (Tier II and Tier 3) with low odometer readings. Ammonia emissions from Tier 0 (1981–1993), Tier I (1994–2000), NLEV (2001–2003), Tier II (2004–2016) and Tier III (2017–2025) vehicles had average emission rates of 563.1, 177.8, 213.6, 94.4 and 18.9 mg/mile, respectively. The outcomes also showed that the NH₃ emission rates of gasoline motor vehicles had a strong positive correlation with post-catalyst CO and NO_x exhaust concentrations, with correlation coefficients of 0.7 and 0.8, respectively. This suggests that more ammonia exhaust emissions would be expected when the post-catalyst CO and NO_x exhaust concentrations increase. The correlation analysis also revealed that NH₃ emission rates had a moderate (negative) correlation with the vehicles' model year ($r = -0.5$), and a moderate (positive) correlation with vehicles' mileage. This suggests that gasoline motor vehicles would cause higher emission rates of ammonia as they get older. Lastly, NH₃ emission rates were moderately correlated with engine displacement and number of cylinders and poorly correlated with the vehicle's GVWR. Based on these correlations, it would be increasing NH₃ emissions from LD gasoline motor vehicles with earlier model years, higher odometer reading, larger engine displacement and greater number of cylinders.

Data availability

The raw data and the code used to analyze the data and create all the plots in this article are open access and are made available at (Abualqumboz et al., 2022).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.apr.2022.101449>.

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