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Greenhouse gas emissions from heavy-duty natural gas, hybrid, and conventional diesel on-road trucks during freight transport

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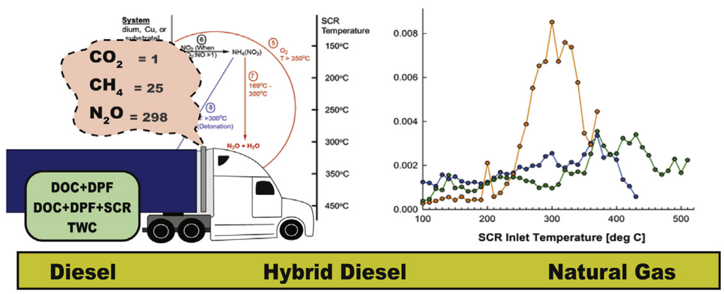
HIGHLIGHTS

- Carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) measured from trucks.
- Seven trucks were measured on-road including diesel, hybrid diesel, and natural gas.
- N_2O was ten times higher for diesel trucks with selective catalytic reduction.
- CO_2 equivalent (CO_2-eq) considers relative warming potentials of all three gases.
- Natural gas and hybrid diesel vehicles had lower CO_2-eq for selected routes only.

GRAPHICAL ABSTRACT



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ABSTRACT

Heavy-duty on-road vehicles account for 70% of all freight transport and 20% of transportation-sector greenhouse gas (GHG) emissions in the United States. This study measured three prevalent GHG emissions – carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) – from seven heavy-duty vehicles, fueled by diesel and compressed natural gas (CNG), and compliant to the MY 2007 or 2010 U.S. EPA emission standards, while operated over six routes used for freight movement in California. Total combined (tractor, trailer, and payload) weights were 68,000 ± 1000 lbs. for the seven vehicles. Using the International Panel on Climate Change (IPCC) radiative forcing values for a 100-year time horizon, N_2O emissions accounted for 2.6–8.3% of total tailpipe CO_2 equivalent emissions (CO_2-eq) for diesel vehicles equipped with Diesel Oxidation Catalyst, Diesel Particulate Filter, and Selective Catalytic Reduction system (DOC + DPF + SCR), and CH_4 emissions accounted for 1.4–5.9% of CO_2-eq emissions from the CNG-powered vehicle with a three-way catalyst (TWC). N_2O emissions from diesel vehicles equipped with SCR (0.17–0.30 g/mi) were an order of magnitude higher than diesel vehicles without SCR (0.013–0.023 g/mi) during highway operation. For the vehicles selected in this test program, we measured 11–22% lower CO_2-eq emissions from a hybrid compared to conventional diesel vehicles during transport over lower-speed routes of the freight transport system, but 20–27% higher CO_2-eq emissions during higher-speed routes. Similarly, a CNG vehicle emitted up to 15% lower CO_2-eq compared to conventional diesel vehicles over more neutral-grade highway routes, but emitted up to 12% greater CO_2-eq emissions over routes with higher engine loads.

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1. Introduction

Anthropogenic greenhouse gas (GHG) emissions have increased since the pre-industrial era, and in 2015, over 100 nations have agreed upon and adopted approaches (i.e. *The Paris Agreement*) to limit warming to less than 2 °C to prevent additional and irreversible economic, ecological, and infrastructure damage (IPCC, 2014; Meinshausen et al., 2009; United Nations 2015; Solomon et al., 2009; Tol; Walther et al., 2002). The United States is the second-highest GHG-emitting nation in the world, and in recent years emits about 15% of global GHG emissions (JRC, 2016). Several actions are already underway within the United States to reduce GHG emissions. In California, the Global Warming Solutions Act (AB 32) requires that California achieve 1990 emission levels by 2020 (CARB, 2014), Senate Bill (SB) 32 requires 40% below 1990 levels by 2030 (California, 2016), and Executive Order S-3-05 requires 80% below 1990 levels by 2050 (Schwarzenegger, 2005).

Mobile sources (including cars, trucks, off-road equipment, and others) currently account for 36% of California's GHG emissions (CARB, 2014). New passenger cars sold in the United States must emit ~5% lower GHG emissions for each model year (MY) between 2017 and 2025 (CARB, 2012). Another major source is heavy-duty (HD) on-road vehicles weighing greater than 14,000 lbs., where the federal Phase I and II GHG requirements are expected to reduce the sector's GHG emissions by nearly 40% for MY 2027 vehicles compared to a MY 2014 baseline (U.S. EPA, 2011; U.S. EPA, 2015a). The engines that power HD on-road vehicles have standards set for three GHGs: carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). Methane is a short-lived climate pollutant (average lifetime = 12.4 years), with global warming potential (GWP) equal to 25 times higher than CO_2 over a 100-year time horizon; N_2O is also a potent GHG with a longer lifetime (121 years) and a GWP of 298 over a 100-year time horizon (IPCC, 2013). N_2O is not only a heat-trapping pollutant, but also is the largest known remaining anthropogenic threat to the stratospheric ozone layer (Kanter et al., 2013). The agriculture sector is the predominant global control target for N_2O, but emissions reductions are also needed from mobile sources (Shcherbak et al., 2014). CO_2 emissions are regulated for MY 2014 and newer heavy-duty on-road engines depending on the application of the truck in which it will be operated (627–432 g CO_2 per brake-horsepower-hr [bhp-hr]), and CH_4 and N_2O standards (both 0.1 g/bhp-hr) apply to MY 2015 and newer engines (Table S1). The engine dynamometer Supplemental Emission Test (SET) assesses performance of engines sold in tractors over various steady-state torque and engine speeds. In addition to the engine standards, the Greenhouse Gas Emission Model (GEM) assesses compliance to applicable vehicle CO_2 or fuel economy standards (64–92 g CO_2/ton-mile of payload for tractors operating in freight applications, Table S2).

This paper summarizes the CO_2, N_2O, and CH_4 emissions from seven modern technology HD vehicles during real-world operation along major freight corridors in California. The on-road trucking industry hauls about 70% of all freight in the United States (U.S. EPA, 2015a), and therefore a sustainable freight system with low overall GHG emissions must ensure on-road trucks are achieving intended benchmarks. This assessment includes one diesel engine certified to the MY 2007 emission standard, one hybrid diesel vehicle with a MY 2011 engine, four MY 2013 or 2014 conventional diesel engines, and one MY 2013 compressed natural gas (CNG) engine. Total vehicle weight was 68,000 ± 1000 lbs. to correspond to median freight hauling loads (39,250 lbs. of payload = 19.63 tons). This study reports emissions from 96 total trips lasting 1–2 h each, all starting and beginning from a stop, for six route classifications (Figs. S1 and S2). Three of the six routes were defined by repeat trip over the same geographical locations (Local Drayage, Near-Dock Drayage, and Urban Arterial). For the other three routes (Hill Climb, Interstate, and Regional Highway), trips were performed in many locations throughout California, and classification was determined operationally.

2. Methods

2.1. Test vehicles and trailer

Table 1 lists the vehicles, engines, certification information, and fuels for each vehicle in the study. Vehicles were selected to represent four emission technology groups: (1) one conventional diesel certified to the MY 2007 emission standards with DOC + DPF aftertreatment; (2) four conventional diesel engines (MY, 2013 through 2014) certified to the MY 2010 emission standards with a DOC + DPF + SCR aftertreatment system; (3) one hybrid diesel engine (MY, 2011) certified to a transition family emission limit (FEL) of 0.46 g NOx/bhp-hr using a DOC + DPF but no SCR; and (4), one stoichiometric compressed natural gas (CNG) engine (MY, 2013) certified to the MY 2010 emission standard using a TWC. All seven engines utilized exhaust gas recirculation (EGR) as part of the emission control system. All seven engines had certification PM emissions of at least three times below the current standard (0.01 g/bhp-hr), and CO + non-methane hydrocarbon (NMHC) emission of at least two times below current standards (15.5 g CO/bhp-hr and 0.14 g NMHC/bhp-hr) as reported elsewhere by Quiros et al. (2016). Odometer readings were below the regulatory useful life limit of 435,000 miles for Vehicles 1–5, 7, and 185,000 miles for Vehicle 6.

All tractors pulled the mobile laboratory (TEMS) affixed to a flatbed trailer along with an on-board power generator, and other emissions measurement equipment. Previous works have evaluated the TEMS system (Kappanna et al., 2013) and the sampling configuration used in the study (Quiros et al., 2016). The trailer was not equipped with any aerodynamic drag-reducing equipment, such as trailer skirts as required by U.S. EPA SmartWay program requirements, although the flatbed trailer used in this study may have exhibited some drag-reducing properties due to its low ground clearance. The TEMS is equipped with a constant volume sampler (CVS) set to approximately 1800 cubic feet per minute (CFM).

2.2. Instruments and data processing

Diluted CO_2 was measured using a MEXA 7200d (Horiba Ltd., Japan) laboratory-grade bench analyzer reporting other criteria gases used for regulatory compliance testing. Total molar flow rates for dilute CO_2 measurements were obtained from the total flow of the CVS.

N_2O and CH_4 emissions were measured in real-time using an MKS Instruments Inc. (San Jose, CA, USA) MultiGas 2030-HS High Speed Fourier-Transform Infrared (FTIR) gas analyzer. Minimum detectable concentrations (MDC) for N_2O and CH_4 were 0.25 and 0.5 ppm, respectively. N_2O and CH_4 were measured from the raw exhaust, and CO_2 was measured from the CVS in the mobile laboratory. Exhaust flow was measured using a pitot-tube high-speed electronic flow module (EFM-HS) manufactured by Sensors, Inc. (Saline MI, USA). The Engine control unit (ECU) data broadcast over the controller area network using the SAE J1939 protocol were recorded to measure engine torques, engine speed, temperatures at various locations of the engine and aftertreatment system, and other parameters. Raw emissions data were post-processed according to CFR guidelines for performing drift correction (1065.672), performing dry-to-wet conversion of analyzers operating downstream of a chiller (1065.659), and for performing dilution-air background correction (1065.667).

Table 1
Heavy-duty test engines, emissions information, and vehicle details. Vehicles 2, 4, 5, and 7 were all SCR-equipped and were equipped with an AMOX catalyst; however, Vehicles 4 and 5 were certified with a discrete physical module, and Vehicles 2 and 7 were equipped with the catalyst as part of the SCR system.

	Veh 1	Veh 2	Veh 3	Veh 4	Veh 5	Veh 6	Veh 7
Manufacturer	OEM 1	OEM 1	OEM 1	OEM 2	OEM 3	OEM 4	OEM 4
Engine MY	2007	2013	2013	2014	2014	2011	2013
Model	Diesel	Diesel	CNG	Diesel	Diesel	Hybrid Diesel	Diesel
Aftertreatment Configuration	DOC + DPF	DOC + DPF + SCR	TWC	DOC + DPF + SCR + AMOX	DOC + DPF + SCR + AMOX	DOC + DPF	DOC + DPF + SCR
Displacement [L]	15.0	15.0	11.9	14.8	12.8	7.6	12.4
Rated Power [hp]	550	450	400	505	405	260	475
GVWR [lbs.]	80,000	80,000	80,000	80,000	80,000	61,000	80,000
Odometer [mi]	393,174	123,471	11,142	110,680	40,420	34,260	186,389
SET CO_2 Cert [g/bhp-hr]	N/A	N/A	N/A	460	463	463	N/A

2.3. N_2O emissions variability analysis for diesel vehicles with SCR

Repeatability and reproducibility analyses were performed to evaluate the intra-vehicle and inter-vehicle variability of the N_2O emissions during on-road trips. Repeatability is the variability that results from repeated trips made with the same vehicle over the same route type (intra-vehicle variability). Reproducibility includes both intra-vehicle variability and inter-vehicle variability, the variability between groups (in this case, the four MY, 2013/2014 diesel vehicles with SCR). Note that repeatability and reproducibility are only valid between vehicles and repeat trips for the same route type. Using this approach, reproducibility and repeatability are reported separately for each of the six route types; however, only a subset of these parameters were calculated for the analysis performed in this paper. The intra-vehicle variability (s_r^2) and inter-vehicle variability (s_L^2) can be calculated using the following equations, which were adapted from the equations reported by Hu et al. (2014):

$$s_r^2 = \frac{\sum_{i=1}^{p}\sum_{j=1}^{n_i}\left(x_{ij} - \overline{x}_i\right)^2}{N - p} \tag{1}$$

$$s_L^2 = \frac{\frac{\sum_{i=1}^{p}(\overline{x}_i - \overline{x})^2}{p-1} - s_r^2}{\frac{\left(N^2 - \sum_{i=1}^{p} n_i^2\right)}{N(p-1)}} \tag{2}$$

Where, p is the total number of vehicles, N is the total number of trips (measurements), j is the j-th test from the test vehicle i (e.g., OEM 1–4), x is the emissions factor, $\overline{x_i}$ is the estimated mean N_2O emissions factor from i-th vehicle, and $\overline{x}$ is the estimated mean N_2O emission factor for all measurements.

$$N = \sum_{i=1}^{p} n_i \tag{3}$$

$$\overline{x_i} = \frac{1}{n_i}\sum_{j=1}^{n_i} x_{ij} \tag{4}$$

$$\overline{x} = \frac{1}{p}\sum_{l=1}^{p}\overline{x_i} \tag{5}$$

The sum of intra-vehicle and inter-vehicle variability equals total variability s_R^2 (reproducibility):

$$s_R^2 = s_r^2 + s_L^2 \tag{6}$$

3. Carbon dioxide emissions

3.1. Emissions per mile

Fig. 1 shows the MY 2013/2014 diesel engines, which were all certified with DOC, DPF, and SCR systems, generally had lower CO_2 emissions compared to the MY 2007 diesel and MY 2011 hybrid diesel engines, which were certified to higher NOx emission standards and were not equipped with SCR. Periodically, some diesel vehicles underwent active DPF regeneration to remove accumulated PM; this process increases lifetime CO_2 emission rates by up to a percent or two, and we excluded some trips to better intercompare emissions among routes and technologies (refer to Supporting Information (SI) Section 1). Calculated per payload-ton by scaling emissions to the actual payload of 19.6 tons, most route-average emissions were greater than the 76 g CO_2/ton-mi vehicle standard for mid-roof sleeper cabs under the GHG Phase 1 standards (Table S2). The actual CO_2 emission rates are generally numerically greater than the standard assessed using model-based certification approach (i.e. GEM), but simulated emissions are simulated over mostly steady state driving profiles with no road grade at either exactly 55 mi/hr or 65 mi/hr, and do not represent real-world driving conditions. Whereas real-world emission rates may exceed vehicle certification standards, the technologies operating on the road likely achieve GHG reductions, and may be compliant with the federal GHG vehicle standards.

The Interstate Highway Route includes trips with 80% of the operation above 40 mi/hr on highways with no extended grade as shown in Fig. S2 (trip average and standard deviation (SD) = 51.2 ± 3.45 mi/hr). CO_2 emissions from the MY 2013/2014 diesel (1465 ± 189 g/mi), the MY 2007 diesel (1524 ± 239 g/mi), and the MY 2013 CNG (1317 ± 190 g/mi) vehicles were not statistically different. However, the hybrid diesel vehicle had 22% higher CO_2 emissions (1825 g/mi) than the average of the conventional diesel vehicles and 38% higher CO_2 emissions than the CNG vehicle. Although all trucks were Class 8 and capable of industrial freight transport, the engine of the hybrid drivetrain had substantially smaller engine displacement volume, peak power, and weight classification than the conventional diesel vehicles (Table 1) and can help explain elevated highway emissions. Figs. S3 and S4 show the MY 2011 diesel hybrid vehicle exhibited less fluctuations in vehicle power, and operated with an engine speed of around ~2000 rpm compared to ~1200 rpm for a MY 2013/2014 diesel vehicle during steady-state operation over the Interstate Highway Route. Upward of 22% CO_2 emissions dis-benefits were observed from this engine platform during higher-load operation, which were likely an artifact not of the hybrid technology but due to the engine platform that was sized. During highway operation, the contribution of power from the on-board battery diminishes as the state of charge is depleted, and a greater fraction of energy must come from the on-board combustion engine. The consideration of

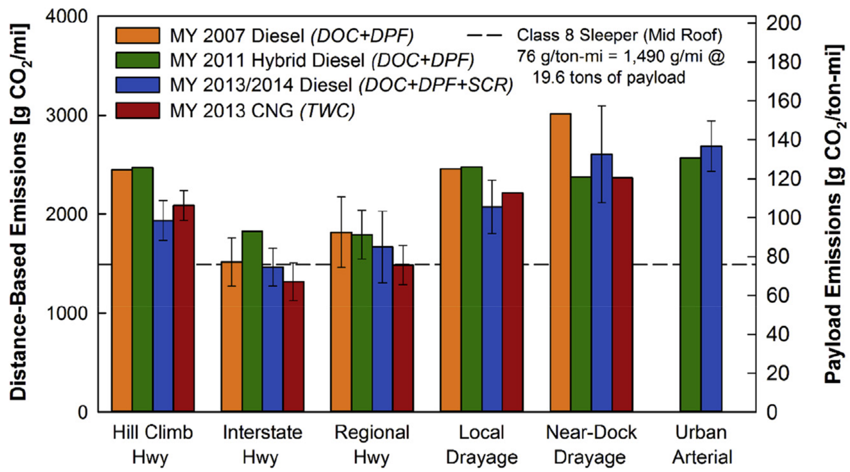

Fig. 1. Average distance and payload-specific CO_2 emissions for each vehicle group and route. Tabulated summary of the data is located in Table 2. Error bars indicate plus and minus one SD of average trip emissions, each trip lasting an average of 94 ± 35 min. The absence of error bars indicates two or fewer trips per route-vehicle combination. The figure represents a total of 96 trips and 148 h of on-road data among all seven vehicles. The dashed horizontal line indicates the MY 2014–2016 vehicle standard for a mid-roof class 8 sleeper cab tractors under the U.S. EPA Phase I GHG regulation. The terms diesel oxidation catalyst (DOC), diesel particulate filter (DPF), selective catalytic reduction (SCR), and three-way catalyst (TWC) refer to the aftertreatment options configured on each major technology group. The MY 2013/2014 diesel vehicle group (blue) includes the average of four total vehicles: two MY 2013 and two MY 2014 vehicles, one each from four major HD engine manufacturers. The MY 2007 diesel and CNG vehicle were not operated on the Urban Arterial Route, and therefore no data bars are presented here or in subsequent figures.

engine sizing and application-specific placement of heavy-duty hybrid vehicle technologies will need to be considered not only for the primary customer of a truck, but also the secondary market into which the truck enters when the truck is sold. Similar effects were found in a recent study of medium heavy-duty hybrid-diesel engines under a variety of duty cycles (Thorton et al., 2015). Nevertheless, most vehicles had the lowest CO_2 emissions over the Interstate Highway Route, and subsequent comparisons reference it as a baseline.

The Hill Climb Route was conducted over either two highway passes entering and leaving the South Coast Air Basin in California, I-5 (i.e. the "Grapevine") or I-15 (i.e. The "Cajon Pass"). All of these trips included downhill to uphill portions, a quasi-continuous incline ascending at least 800 m. The average of positive grades of this trip was 2.3% with an interquartile range (IQR, 25th to 75th percentiles) of 0.83–3.33%, and minimum and maximum road grades were −6.5 and 6.5%, respectively. Hill Climb Route CO_2 emissions were 32–62% higher than the Interstate Highway Route depending on vehicle group. The MY 2013/2014 diesel vehicles emitted 7.3% less CO_2 than the CNG vehicle during the Hill Climb Highway Route, and had the lowest CO_2 emissions of all vehicle groups (1936 g/mi). During all nine trips completed on the Hill Climb Route, the final elevation was greater than the initial elevation (net ascents ranged between 35 and 1 002 m), likely contributing to the increased distance-specific emission rates relative to "flat" Interstate Highway Driving. However, for the trip that ascended only 35 m (starting elevation of 305 m, maximum elevation of 1 252 m, final elevation of 340 m), the trip-average CO_2 emissions were still 17% higher (1792 g/mi) than the average of trips completed over the Interstate Highway Route for that same vehicle (Vehicle 5). This evidences that freight transport through highway passes that start and finish at the same elevations, can produce excess CO_2 per-mile compared to flat highway routes.

The Regional Highway Route included trips with less than 80% of operation above 40 mi/hr, where lower-speed operation resulted from urban congestion and more frequent highway interchanges (trip average and SD = 34.0 ± 6.45 mi/hr). The Regional Highway Route had CO_2 emissions 20% higher (MY, 2007 diesel), 1.8% lower (hybrid diesel), 14% higher (MY, 2013/2014 diesel), and 13% higher (2011 CNG) than the Interstate Highway Route. Although the hybrid diesel truck had slightly lower CO_2 emissions for the Regional compared to Interstate Highway Route, overall CO_2 emission factors were 7.5% higher than the MY 2013/2014 diesel group (1667 g/mi) and 21% higher emissions than the CNG vehicle (1483 g/mi). By virtue of the operational definition of the Regional Highway Route, a variety of traffic conditions were included in the route classification, and therefore test-to-test variability resulted in overlapping ranges of measurements across vehicle technologies (Fig. 1). Nevertheless, the average trip speeds ranged 30.6–36.1 mph (Table 2), and the CNG vehicle had the lowest CO_2 emissions over the Regional Highway Route by 11% compared to all other vehicle groups.

The Local Drayage Route was defined by trips over the same 22-mile route including urban surface street driving, accelerations onto highways, and congested highway driving (trip average and SD = 18.2 ± 3.79 mi/hr). Depending on vehicle group, CO_2 emissions were 36–68% higher compared to the Interstate Highway Route. Diesel vehicles with SCR (2071 g/mi) had 19–20% lower emissions than diesel vehicles without SCR (2457–2475 g/mi), and 7% lower emissions than the CNG vehicle (2213 g/mi). These data show that freight transport along urban congested highways can be associated with 60–70% higher CO_2 emissions per mile, and that for this duty cycle, the CNG and hybrid diesel drivetrains did not achieve any tailpipe CO_2 emissions reductions compared to diesel counterparts.

The Near-Dock Drayage (container pickup at ports, 8.2 ± 1.42 mi/hr) and Urban Arterial Routes ("last-mile" delivery, 17.5 ± 1.18 mi/hr) were also defined by the geographic route; characteristics are described in Figs. S1 and S2. For the Near Dock Route, per-mile CO_2 emissions increased by 30% (hybrid diesel) to 99% (MY, 2007 diesel), relative to the Interstate Highway Route. The hybrid diesel (2377 g/mi) and CNG vehicle (2369 g/mi) had 8.7 and 9.0% lower CO_2 emissions, respectively, than the newer MY 2013/2014 diesel vehicles (2604 g/mi). By virtue of the duty cycle of the Near Dock Route, per-mile CO_2 emissions were higher for all vehicle technologies compared to the Interstate Highway Route. However, CNG and diesel hybrid vehicles exhibited emissions reductions relative to conventional diesels over these short-haul and lower-speed

Table 2
Average trip emissions on a per-mile, brake-specific, and 100-yr global warming potential (GWP) basis, calculated using a GWP of 25 (CH_4) and 298 for (N_2O) according to the AR4 of the IPCC.

	Avg. Speed	Distance	Work/Distance	Vehicle Emission Rate [g/mi]				Engine Emission Rate [g/bhp-hr]		
	[mph]	[mi]	[bhp-hr/mi]	**CO_2**	**N_2O**	**CH_4**	**Total GHG (CO_2-eq)**	**CO_2**	**N_2O**	**CH_4**
MY 2007 Conventional Diesel (DOC + DPF) − Vehicle 1										
Hill Climb	53.2	74.1	4.3	2445	0.013	0.005	2449	572	0.003	0.001
Interstate	52.3	817	2.2	1516	0.016	0.005	1521	679	0.007	0.002
Regional	32.9	241	2.7	1815	0.023	0.010	1823	662	0.008	0.003
Local	19.6	22.0	3.7	2457	0.069	0.001	2477	666	0.019	0.000
Near-Dock	7.8	7.3	3.1	3015	0.129	0.061	3055	984	0.042	0.020
MY 2011 Hybrid Diesel (DOC + DPF) − Vehicle 6										
Hill Climb	32.2	57.1	3.8	2468	0.027	0.036	2477	654	0.007	0.009
Interstate	51.4	56.7	2.8	1825	0.013	0.014	1829	648	0.005	0.005
Regional	34.2	147	2.8	1792	0.018	0.027	1799	662	0.007	0.010
Local	14.3	21.9	3.7	2475	0.026	0.024	2483	670	0.007	0.007
Near-Dock	6.6	7.4	3.2	2377	0.022	0.033	2385	750	0.007	0.010
Urban	15.9	33.7	4.0	2567	0.040	0.032	2580	644	0.010	0.008
MY 2013/2014 Conventional Diesel (DOC + DPF + SCR) − Vehicles 2, 4, 5, & 7										
Hill Climb	44.6	426	4.3	1936	0.173	0.007	1988	451	0.040	0.002
Interstate	50.7	2 160	3.2	1465	0.172	0.005	1516	463	0.055	0.001
Regional	30.6	904	3.4	1667	0.294	0.010	1755	505	0.089	0.003
Local	16.8	78.0	4.5	2072	0.389	0.018	2220	538	0.111	0.005
Near-Dock	8.2	29.5	4.0	2604	0.221	0.033	2671	655	0.056	0.008
Urban	18.0	101	5.4	2687	0.821	0.020	2933	495	0.151	0.004
MY 2013 CNG (TWC) − Vehicle 3										
Hill Climb	40.0	94.5	4.8	2088	0.111	5.030	2246	431	0.023	1.039
Interstate	50.8	213.4	2.8	1317	0.100	2.885	1419	472	0.036	1.034
Regional	36.1	233.2	3.2	1484	0.033	2.387	1553	459	0.010	0.738
Local	15.6	21.9	3.5	2214	0.008	1.286	2248	628	0.002	0.365
Near-Dock	9.2	7.4	3.2	2369	0.383	6.240	2639	732	0.118	1.929

routes. This not only evidences the advantages of deploying these technologies into targeted segments in the freight transport to mitigate tailpipe CO_2 emissions, but short-haul low-speed routes are more amenable to be served by currently available vehicle range and refueling infrastructure (CARB, 2016b).

3.2. Emissions as a function of speed

Fig. 2 presents distance- and payload-specific emission rates (ER) as a function of trip averages (colored dots) and speed bins (gray boxplots) for (a) the MY 2007 diesel, (b) the MY 2013/2014 diesel, and (c) MY 2013 CNG vehicles. Each colored dot represents one trip, which started and ended from a stop, which is the approach used in the EMFAC model. The dashed line indicates trip-average emission rates predicted by the model according to the zero mile rate (ZMR) at 18.9 mi/hr, and the speed-correction factor (SCF) for MY 2013 and newer diesel vehicles. The comparison of the ZMR to vehicles in this study, which have accrued some mileage in the field, is relevant because EMFAC does not attribute any deterioration factor for CO_2 emission rates. In the case the test vehicles were beyond their useful life of 435,000 miles, or were improperly maintained, a direct comparison may not be valid as presented. The colored circles (this data) and dashed line (EMFAC trip-average prediction) are nearly identical at all trip-average speeds between 5 and 55 mi/hr. Larger discrepancies were observed for the MY 2007 diesel and MY 2013 CNG vehicles where up to 20% differences were observed for some trip-average speeds within the 5 to 55 mi/hr range. These differences may be due to vehicle selection, and the agreement between measured and modeled trip-average data demonstrates that the EMFAC approach, which included on chassis dynamometer data with no road grade consideration, generally achieves similar results as on-road measurement.

The gray boxplots in Fig. 2 illustrate that median real-time emission rates generally match with the best-fit line derived from trip-average speeds. In Fig. 2a–b, the 40- and 45-mi/hr bins exhibited larger differences, which may have resulted from lower sample sizes and more transient operation within these speed bins where vehicles accelerated onto highways or out of congested highway regions. Additionally, bin-average values (gray X's) were typically greater than the medians, indicating a skewed-left distribution resulting from brief periods of higher CO_2 emission rates within each speed bin. Both the real-time (boxplots) and trip-average (colored circles) data indicate lower CO_2 emissions per mile are observed for trips with higher average or real-time speeds up to 65 mi/hr.

For emission rates above 40 mi/hr, these data do not suggest that limiting vehicle speed will result in increased CO_2 emissions. Rather, these data suggest that the probability of increased CO_2 emissions per mile is higher when selecting a segment or trip with a lesser compared to greater average speed from real-world driving activities. CO_2 emissions bins for higher speeds are likely associated with more steady state compared to transient operation, which is more likely at lower speeds. All else equal, increased road load at higher speeds is expected due to non-linear increases in aerodynamic drag forces. Overall, data suggest that the probability of decreased CO_2 emissions per mile is higher (maximum, 55 mi/hr) for trips with higher compared to lower average speeds (minimum, 7 mi/hr).

4. Nitrous oxide emissions

Fig. 3 shows the route-average N_2O emissions from diesel vehicles with a DOC + DPF but without SCR (the MY, 2007 diesel and hybrid diesel) were lower (0.015–0.075 g/mi) than the CNG vehicle (0.008–0.38 g/mi). Even for diesel vehicles without SCR, emission rates of N_2O were typically greater than the detection limit of 0.25 ppm. The highest N_2O emissions were generally from the diesel vehicles with DOC + DPF + SCR (MY, 2013/2014, 0.17–0.82 g/mi), except for the Near-Dock Drayage Route where CNG vehicle had the highest N_2O emissions. The Near-Dock Drayage Route had

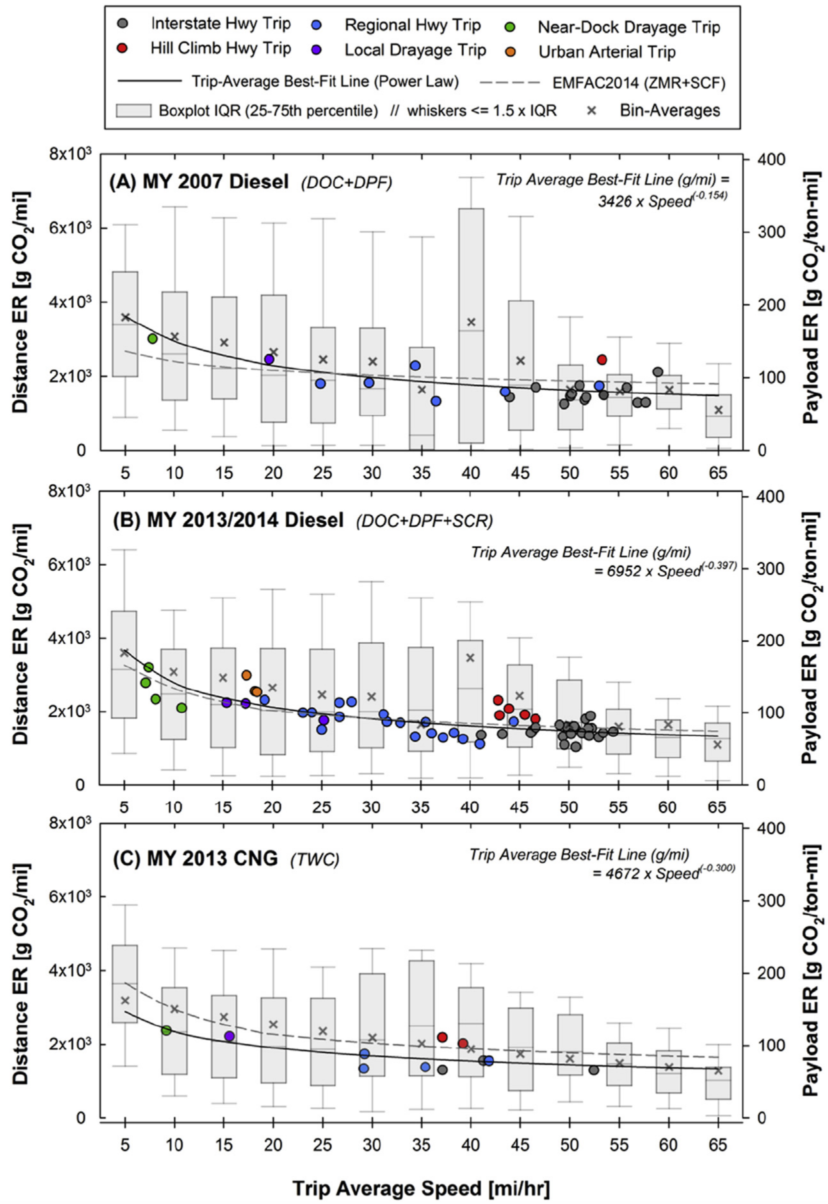

Fig. 2. Distance-specific CO_2 emission rates (ER) by trip (colored dots) and by speed bins (gray boxplots) for vehicles with the (A) the MY 2007 diesel, (B) MY 2013/2014 diesel, and (C) the MY 2013 CNG engines. Boxplots represent 5-mi/hr bins for speeds greater than 2.5 mi/hr, data are shown by the Interquartile Range (IQR, 25th to 75th percentile of observations), whiskers 1.5 times the IQR, and gray X's represent bin averages. Black represents the best-fit lines according to power-law fit annotations. Dashed gray lines indicate the EMFAC2014 prediction for the vehicle technology, where zero mile rates (ZMR) and speed correction factors (SCF) are considered. Payload specific ERs were calculated based on 39,250 lbs. payload (19.63 tons).

the lowest trip-average speeds, and also had the highest NOx emissions for all vehicle technologies, which is due to lower load as discussed elsewhere (Quiros et al., 2016). Although these engines were certified before the MY 2015 N_2O standard of 0.1 g bhp-hr, the average brake-specific emission rate (0.035 g/bhp-hr) was below the engine certification standard. Table 2 shows some route-technology combinations resulted in exceedances of the standard, and the next sections further describe emissions trends and some formation mechanisms. Some previous studies have demonstrated N_2O reductions by lowering fuel sulfur from 330 to 30 ppm in gasoline (Huai et al., 2004); however, no substantial additional N_2O reductions are expected from further reducing fuel sulfur content as all diesel vehicles were already operating on ULSD (5–10 ppm total sulfur).

4.1. N_2O emissions from diesel vehicles with DOC + DPF but without SCR

Low oxygen (O_2) content, especially in stoichiometric-combustion engines, can favor N_2O formation up to 25% of the NOx emission rates, and formation rates has been positively correlated with carbon monoxide (CO) emissions (Becker et al., 1999). However, much lower N_2O emissions were observed in the diesel engines, which have excess O_2 in the exhaust; when equipped with a DOC + DPF but no SCR, average N_2O/NOx emission emissions percentages were 0.23%.

Fig. 4 illustrates the formation mechanisms for N_2O for each technology group. For lean-combustion diesel engines, the redox catalyst in the DOC and catalyzed DPF is made of platinum (Pt) group metals (PGMs) – typically either Pt or palladium (Pd) – on

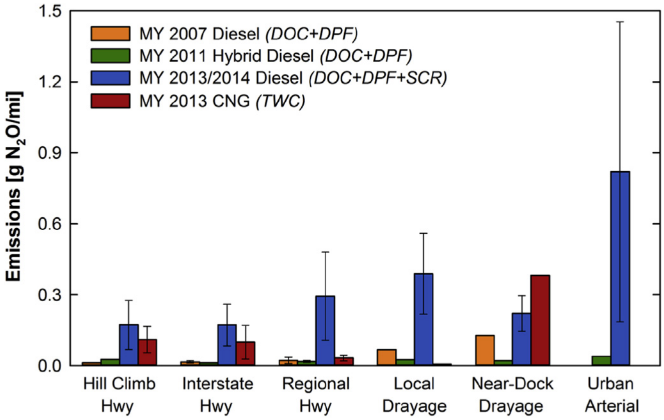

Fig. 3. Average distance-specific N_2O emissions for each vehicle group and route. Error bars indicate plus and minus one SD of average trip emissions. The absence of error bars indicates fewer than three trips per route-vehicle combination. Abbreviations and scope of data in this figure are identical to statistics provided for Fig. 1.

various wash-coat formulations. For lean-combustion diesel engines, the excess O_2 converts NO into mostly NO_2, especially when redox catalyst temperatures are >250 °C (Fig. 4, reaction 1); however for lower-temperature combustion (<250 °C), NO can be converted to either N_2 or N_2O in the presence of hydrocarbons (HC, Fig. 4, reactions 2–4). When N_2O formation is favored (reactions 2–3), its emissions rates are heavily dependent on the composition of hydrocarbons, exhaust temperature (fastest reactions occur at 150–250 °C), and catalyst formulations (generally proprietary). The measured N_2O emissions in our study likely occurred over the DOC and/or DPF as the product of hydrocarbon reduction of NOx (Graham et al., 2008; Kamasamudram et al., 2012; Wang et al., 2010).

Real-time emissions were a decreasing function of vehicle speed (Fig. S5a) and increased sharply for road grades above 5% (Fig. S5b). Trip-average emissions also decreased at increased average speeds (Fig. S5); and, results generally agree with the broad ranges of emission rates reported by U.S. EPA MOVES2014 model (0.0048–0.083 g/mi (U.S. EPA, 2015b)). Note that Figs. S5a–d present time-resolved data, and especially for road grade, operation during the high-grade bins likely occur for short periods of times. Sustained high-speed, high-grade, and high-load operation that results in changes in aftertreatment temperature may suggestion different effects. For instance, one previous study showed higher emission rates from retrofit trucks (0.121–0.189 g/mi), as well as 3–4 times higher N_2O emission rates during active DPF regeneration (Graham et al., 2008); however, in our work no substantial increase in N_2O emission rates were measured during the 5 of 29 trips that included active regeneration events (e.g. Fig. S7). Hydrocarbon concentrations in the DOC for MY 2007 and newer diesel engines were likely lower than for older engines, therefore favoring oxidation of NO into NO_2 (reaction 1) or N_2 (reaction 4) rather than N_2O (reaction 3).

4.2. N_2O emissions from diesel vehicles with SCR

Relative to diesel vehicles without SCR, the four diesel vehicles with DOC + DPF + SCR emitted 2.9–21 times higher route-average N_2O emissions (Fig. 3 and Table 2), and had the highest N_2O emissions of all vehicles for nearly all routes. Additionally, the SCR-equipped vehicles exhibited greater variability among repeat tests than other vehicles; coefficients of variation (CVs) increased from 23% for the diesel vehicles with a DOC + DPF but no SCR to 51% for those with DOC + DPF + SCR over the Interstate Highway Route,

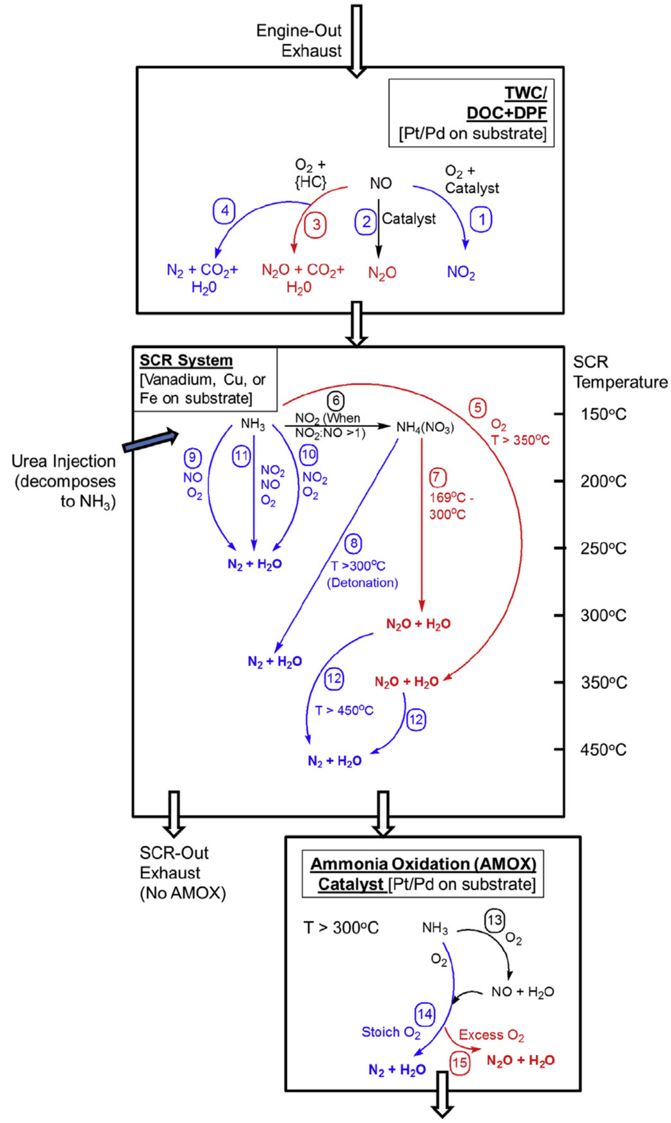

Fig. 4. Formation mechanisms for N_2O for through an oxidation catalyst and a catalytic SCR system (Hallstrom et al., 2013; Huai et al., 2004; Kamasamudram et al., 2012; Koebel et al., 2002; Koike et al., 1999; Madia et al., 2002; Willi et al., 1996). N_2O formation across an oxidation catalyst can be applied to diesel oxidation catalyst (DOC) or for a three-way catalyst on a CNG vehicle. Blue lines indicate pathways leading to reduction of NOx to inert N_2 gas; red lines indicate pathways leading to N_2O formation. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

where the CVs for CO_2 emissions remained relatively similar (16 and 13%, respectively). Using Equations (1)–(6), we calculated inter-vehicle and intra-vehicle variability of emissions from repeat trips made on Interstate Highway (n = 24) and Regional Highway (n = 17) routes. The analysis showed that intra-vehicle variability accounted for 21 and 26% of total variability (due to traffic conditions or other environmental variables) for Interstate and Regional Highway routes respectively, and that inter-vehicle variability (due to differences in engine models or OEMs) accounted for 79 and 74% of total variability.

Fig. 4 shows the most desirable pathway for NOx reduction is a redox reaction of ammonia (NH_3), molecular oxygen (O_2), and an equimolar ratio of NO_2 and NO (NO: NO_2 = 1) across the SCR catalyst (typically Cu/Fe-zeolite or other base metal; Fig. 4, reaction 11) (Willi et al., 1996). N_2O is an undesired side product that forms primarily via direct O_2 oxidation of NH_3 (Fig. 4, reaction 5) or via reactions involving ammonium nitrate (AN) (Fig. 4, reaction 6) (Hallstrom et al., 2013; Kamasamudram et al., 2012; Madia et al., 2002; Willi et al., 1996). SCR catalyst formulation affects N_2O emissions via direct NH_3 oxidation, and Cu-Zeolite formulations show favorable N_2O formation, whereas Vanadium or Iron-based catalysts show less favorable N_2O formation (Kamasamudram et al., 2012). N_2O formation within an SCR system via AN reactions occurs primarily between exhaust temperatures of 200 and 300 °C and NO_2:NO > 1 (Hallstrom et al., 2013; Kamasamudram et al., 2012). At exhaust temperatures >300 °C, AN begins to decomposed/detonate and N_2O production via AN pathways become less important as shown in Fig. 4, reaction 8 (Kamasamudram et al., 2012; Koebel et al., 2002). N_2O formation via NH_3 direct oxidation becomes increasingly important between 350 and 500 °C, but as mentioned previously this is highly dependent on catalyst formulation (Hallstrom et al., 2013; Kamasamudram et al., 2012).

Fig. 5 presents average N_2O emission rates as a function of exhaust temperature, where N_2O formation rates peak between 350 and 425 °C for Vehicles 2 and 7. N_2O begins to thermally decompose at exhaust temperatures >400 °C, with near 100% decomposition at 600 °C (Huai et al., 2004; Kamasamudram et al., 2012; Koike et al., 1999), which creates a competition of N_2O formation/decomposition between 400 and 500 °C. When SCR temperatures approach and exceed 500–550 °C, such as during active DPF regeneration events as shown in Fig. S7 for Vehicle 7 (OEM 4), N_2O undergoes significant thermal decomposition, and therefore additional N_2O produced likely degraded soon thereafter.

Either downstream or at the terminus of the SCR system, SCR-equipped vehicles were equipped with a PGM rather than base-metal catalyst to serve as an ammonia oxidation catalyst (AMOX) to oxidize unreacted NH_3 leaving the SCR to prevent “ammonia slip”. Whereas all of the four engine platforms were equipped with an AMOX catalyst, the catalyst was designed as a separate physical module for two of the engine platforms, where the other two engine platforms had zone-coating at the downstream end of the SCR system. We report in Fig. 5 and Fig. S7 that discrete AMOX modules favor oxidation of NH_3 into N_2O at temperatures between 250 and 300 °C rather than 350–425 °C (Havenith and Verbeek, 1997; Kamasamudram et al., 2012), and in Fig. 6 that they favor 1.7 to 5.4 times greater N_2O emission rates depending on route.

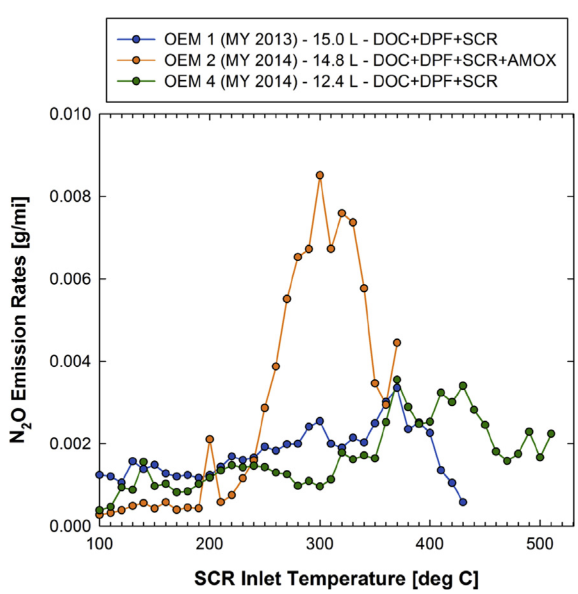

Fig. 5. Average temperature as a function of SCR Inlet Temperature for Vehicle 2 (OEM 1), Vehicle 4 (OEM 2), and Vehicle 7 (OEM 4). Average temperature was binned each 10 °C interval. For the shown temperature bins, there were a total of 64,236, 70,561, and 32,561 one-second observations for OEMs 1, 2, and 4, respectively.

Although SCR configuration and chemistry are strong predictors of formation rates as discussed above, Newtonian parameters are widely used by emission models. Vehicle speed and road-grade did not affect N_2O emissions as dramatically compared to diesel vehicles without SCR as shown in Figs. S5c and S5d. Additional parameters, such as kinetic intensity (KI) and positive kinetic energy (PKE) were calculated for each trip using methods described elsewhere (Milkins and Watson, 1983; O'Keefe et al., 2007; Watson, 1995), and also using micro-trips over various fixed lengths between 1 and 10 min; however no notable trends were identified. One challenge is that some trips have average speeds but driving behavior that result in unique chemistry situations across the SCR catalysts. Greatest variability for the diesel vehicles with SCR was measured during the Urban Arterial Routes (CV = 77%), which consisted of frequent fluctuations between 0 and 50 km/hr as shown in Fig. S2f. Design differences between the vehicles resulted in either greater formation, or lesser formation during these types of driving activities. Nevertheless, Fig. S6 presents N_2O emission rates as a function of trip-average speed using a logarithmic model. Continued tracking of new technology and field-aged vehicles with deterioration of emission control systems will help characterize in-use N_2O emissions trends; the current GHG engine standard of 0.1 g/bhp-hr will apply for all engine MYs as more stringent CO_2 are required.

4.3. N_2O emissions from compressed natural gas vehicles with a TWC

Increased N_2O emissions downstream of a TWC compared to engine-out levels have been documented over the past decades with gasoline vehicles (Becker et al., 1999; Berges et al., 1993). Work that is more recent has identified greater N_2O emissions when aftertreatment temperatures are <300 °C, which typically occurs briefly after cold-start (Huai et al., 2004; Kamasamudram et al., 2012). It is likely for this reason that the lowest-speed Near Dock Drayage Route was associated with the greatest N_2O emission rates per mile relative to the higher speed routes that achieve greater exhaust temperatures. N_2O formation across the TWC typically proceeds by NOx reduction via hydrocarbons in a manner similar to those reactions that can occur across a DOC (Fig. 4, reactions 2–4) (Kamasamudram et al., 2012). Route-average emission rates for this

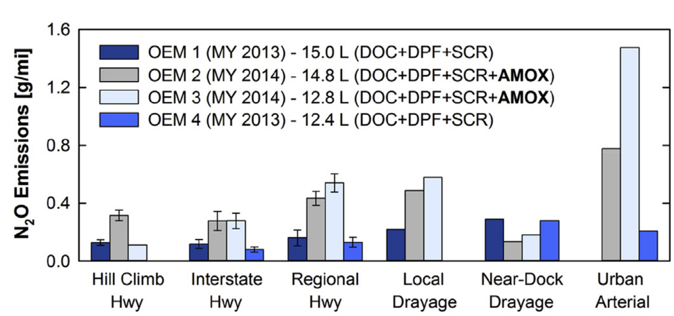

Fig. 6. Average distance-specific N_2O emissions for each original equipment manufacturer (OEM) for MY 2013/2014 diesel vehicles with DOC + DPF + SCR. AMOX refers to ammonia oxidation catalyst, which are certified as discrete separate modules for OEMs 2 and 3, but are zone-coated at the terminus of the SCR for OEMs 1 and 4. The figure shows the same amount of data for the MY 2013/2014 diesel vehicles shown in Fig. 3. Error bars indicate plus and minus one standard deviation (SD) of average trip emissions; the absence of error bars indicates fewer than three trips per route-vehicle combination. The figure represents a total of 56 trips among the four vehicles. Abbreviations and acronyms indicate aftertreatment configurations, which are described in the caption of Fig. 1.

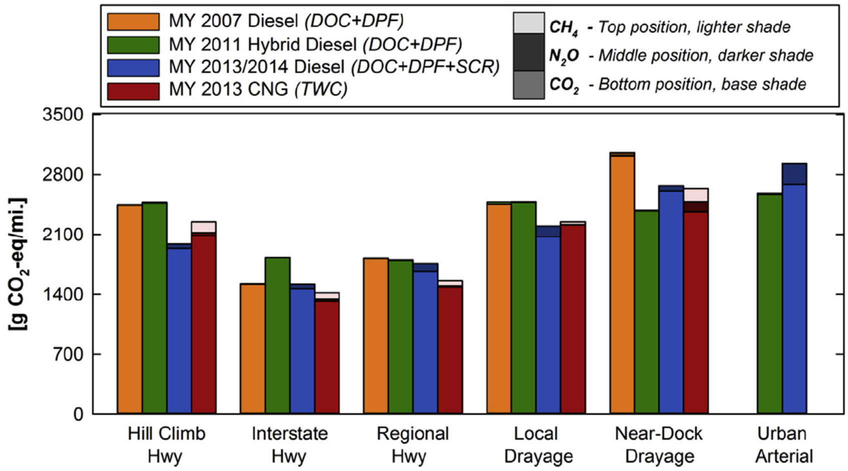

Fig. 7. CO_2 equivalent (CO2-e) emissions on a per-mile basis considering the 100-yr global warming potential of CO_2, N_2O, and CH_4 and from each technology group and route. All three species are reported for each stacked bar; actual emission rates can be found in Table 2. Abbreviations and scope of data in this figure are identical to statistics provided of Fig. 1.

study ranged between 0.008 and 0.38 g/mi, and the CNG vehicle emissions are generally less than SCR-equipped diesel vehicles. Our observed stoichiometric CNG emission rates are comparable to both other on-road testing of in-use transit buses (which had emissions between 0.01 and 0.12 g/mi) (Wang et al., 2015), and to on-road emissions data from lean-burn CNG buses (with emissions between 0.05 and 0.14 g/mi) (Graham et al., 2008). However, emissions rates are lower than reported by U.S. EPA Motor Vehicle Emissions Simulator (MOVES2014) for stoichiometric CNG engines (0.18–1.68 g/mi (U.S. EPA, 2015b)).

5. Methane emissions

Tailpipe CH_4 emissions from the CNG vehicle ranged 1.3 and 6.2 g/mi (Table 2) with highest emission rates measured during the Near Dock Drayage and Hill Climb Highway Routes. On a brake-specific basis, all route-average emissions from the CNG vehicle were 3.6–19 times higher than the MY 2015 standard of 0.1 g/bhp-hr (Fig. S8). Note that these estimates do not include assessment of any upstream CH_4 emissions from on-board, refueling, or distribution infrastructure leakages, which may represent an important and substantial fraction, and therefore should be considered as part of evaluating non-tailpipe GHG emissions from alternative fuel vehicles (Alvarez et al., 2012). Average raw exhaust concentrations of CH_4 from diesel engines measured during the study were typically below 1 ppm, which is less than remote ambient background levels measured in the South Coast Air Basin (Hsu et al., 2010).

6. CO_2 equivalent emissions

The tailpipe CO_2 equivalent (CO_2-eq) emissions were calculated using the 100-yr global warming potential (GWP) values used by the Fourth Assessment Report (AR4) of the IPCC(Forster et al., 2007): 1, 25, and 298 for CO_2, CH_4, and N_2O, respectively, and are presented in Fig. 7 and Table 2. The GWP values from AR4 were selected for consistency with more broad GHG assessments and total inventories developed by CARB and U.S. EPA (CARB, 2016a; U.S. EPA, 2017). Overall, the majority of tailpipe GHG emissions in this work originate from CO_2 (i.e. fuel combustion, technology-route averages range 89.8–99.8%). Fig. S8 and Fig. 7 illustrate diesel engines emitted negligible fractions of CH_4 (<0.05% of total CO_2-eq emissions) but N_2O emissions increased to 2.6–8.3% of route average CO_2-eq emissions when equipped with SCR. The CNG vehicle was the only vehicle tested with significant CH_4 emissions (route-average contributions were 1.4–5.9% of CO_2-eq), and had N_2O emissions rates between 0.1 and 4.3% of total CO_2-eq.

The MY 2013/2014 diesel vehicles CO_2-eq emissions were within 1% of the MY 2007 diesel vehicle over the Interstate Highway Route, and exhibited 11% lower CO_2-eq emissions over the Local Drayage Route, 13% lower emissions over the Near-Dock Drayage Route, and 19% lower emissions over the Hill Climb Highway Route. Despite increased N_2O formation over the SCR and AMOX catalysts, CO_2 reductions resulted in net overall CO_2-eq emissions reductions.

The hybrid diesel vehicle had 11–22% lower CO_2-eq emissions over the Near Dock Route compared to all other vehicle groups, but had 20–29% higher CO_2-eq emissions compared to all other vehicles over higher speed driving during the Interstate Highway Route, and generally higher CO_2-eq emissions for the Hill Climb Highway, Regional Highway, and Local Drayage Routes (−1.3–25% depending in route-vehicle comparison).

The CNG vehicle had 3–15% lower route-average CO_2-eq emissions compared to the average all diesel vehicles (hybrid and conventional). Additionally, the CNG vehicle had 13% lower CO_2-eq emissions over the Regional Highway Route, but 12% higher CO_2-eq emissions over the Hill Climb Highway Route compared to diesel vehicles equipped with SCR. This work corroborates previous findings that 10–20% net GHG tailpipe emissions reductions can be achieved by CNG vehicles compared to conventional diesel technologies (Graham et al., 2008), but demonstrates that the emissions benefits are heavily route dependent.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.atmosenv.2017.08.066.

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