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Regulated, greenhouse gas, and particulate emissions from lean-burn and stoichiometric natural gas heavy-duty vehicles on different fuel compositions

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HIGHLIGHTS

- Natural gas composition impacts the emissions from lean-burn engines.
- Lower THC, CH$_4$, and NO$_x$ emissions for stoichiometric vs. lean-burn engines.
- NH$_3$ emissions produced important increases for the stoichiometric engines.
- Lubricant oil combustion was the main source for particle number formation.
- Higher carbonyl emissions for lean-burn vs. stoichiometric engines.

ARTICLE INFO

ABSTRACT

The gaseous and particulate matter (PM) emissions from three heavy-duty natural gas vehicles, including a lean-burn bus with an oxidation catalyst and two stoichiometric Class 8 trucks with three-way catalysts were evaluated. Testing was performed on a range of three to seven different test fuels with varying Wobbe and methane numbers. The lean-burn vehicle showed general trends of higher emissions of nitrogen oxides (NO$_x$) and non-methane hydrocarbons (NMHC), and lower emissions of total hydrocarbons (THC), methane (CH$_4$), and formaldehyde, and improved fuel economy for the fuels with low methane numbers. The stoichiometric trucks showed some trends toward lower THC, CH$_4$, and NO$_x$ emissions with the low methane number fuels, whereas some increases in NMHC, carbon monoxide (CO), and ammonia (NH$_3$) emissions were also observed. Results of the particle size distributions revealed bimodal size distribution profiles for all three vehicles, with a predominant nucleation mode close to 10 nm for the lean-burn bus and one of the stoichiometric trucks.

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1. Introduction

Recent forecasts for natural gas (NG) resources in the United States (US) suggest that NG will be abundant and low cost for many decades, giving reason to study the efficiencies and the environmental impact of the multiple paths for its use [1]. The US government is continually pushing the use of natural gas engines in order to reduce foreign oil dependence and achieve lower greenhouse gas (GHG) emissions. The most important GHGs are carbon dioxide (CO$_2$), methane (CH$_4$), and nitrous oxide (N$_2$O), with the transportation sector being the main contributor of the overall GHG emissions in the US [2]. Therefore, the introduction of natural gas as a potential alternative to conventional liquid fuels in the heavy-duty vehicle segment (vehicles with gross vehicle weight ratings ranging from 3.9 to 15 tons and over), which consumes a large amount of fuel, is a fast growing market. In California, the use of NG has been increasing for a number of years, due predominantly to expanded power and home heating needs. Currently, California supplies 85–90% of its needs with NG imported domestically from the Rockies, from southwest states, such as Texas, and from Canada [3]. As innovations in horizontal drilling and hydraulic fracturing are unlocking vast unconventional reserves of US domestic NG, however, the composition of imported domestic

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NG supplies could change. NG quality varies with geographic location and season as well as the degree it is processed. So far, the shale revolution is providing US domestic NG at extraordinarily low prices, which could change the economics for processing valuable natural gas liquids (NGLs), such as ethane, propane, butanes, pentanes and hexanes plus. This could lead to natural gas with high Wobbe numbers and lower methane numbers being injected into the pipeline, which could affect the combustion pathways that lead to the formation of carbon monoxide (CO), nitrogen oxides (NO_x), particulate matter (PM), and other harmful pollutants.

Natural gas vehicles (NGVs) have been implemented in a variety of applications as part of efforts to improve urban air quality, particularly within California [4–6]. Most NGVs utilize fuel cylinders containing NG that has been compressed at high pressure (~250 bars), reducing its volume by 99% compared to standard atmospheric conditions; this allows significantly greater driving range between fueling events [7]. Although NG use is widespread in California and the US due to electricity generation and domestic heating, a dedicated NG refueling infrastructure for heavy-duty NGVs is still lacking. However, the use of NG powered transit buses, school buses, and waste haulers in densely populated urban areas remains an attractive alternative to petroleum diesel, since travel distances are relatively short and a central refueling network already exists.

Two technologies have been widely being used for NG heavy-duty engines, namely lean-burn combustion and stoichiometric combustion. Older technology NGVs are equipped with lean-burn engines and oxidation catalysts to effectively control CO and formaldehyde emissions. Current heavy-duty NGVs are equipped with spark-ignited stoichiometric combustion engines, with water cooled exhaust gas recirculation (EGR) technology, and three-way catalysts (TWC) in order to meet the more stringent 2010 NO_x emission standards from the US Environmental Protection Agency (USEPA). Stoichiometric combustion engines with TWC are superior to lean-burn combustion engines with oxidation catalysts for reducing NO_x emissions [8,9]. However, stoichiometric engines with TWCs produce higher CO emissions than lean-burn engines [9]. PM emissions from both stoichiometric and lean-burn combustion NG engines are very low due to the almost homogeneous combustion of the air–gas mixture, and the absence of large hydrocarbon chains and aromatics in the fuel [10].

For NGVs, one issue that has been shown to be important with respect to emissions is the effect of changing the composition of the fuel. This is part of a broader range of issues which are classified under the term interchangeability, which is the ability to substitute one gaseous fuel for another in a combustion application without materially changing operational safety, efficiency, performance or materially increasing air pollutant emissions. Changes in the NG composition used in NGVs can affect the reliability, efficiency, and exhaust emissions. Previous studies conducted with small stationary source engines, heavy-duty engines/vehicles, and light-duty vehicles have shown that NG composition can have an impact on emissions [11–14]. Karavalakis et al. [15] showed higher NO_x emissions when they tested a 2002 lean-burn NG waste hauler on lower methane number/higher Wobbe number fuels. Hajbabaei and colleagues [16] reported NO_x and non-methane hydrocarbon (NMHC) emission increases for fuels with low methane contents when they tested two transit buses equipped with lean-burn NG engines. However, they did not find any fuel effect on NO_x emissions when they tested a bus with a stoichiometric combustion engine and a TWC. The effect of NG composition on exhaust emissions was also confirmed by Feist et al. [17] where they found NO_x and total hydrocarbon (THC) emissions increases with higher Wobbe number fuels under lean-burn engine combustion, while the stoichiometric engines showed no clear trends for NO_x and THC emissions with different fuels.

The present study builds on the work of Karavalakis et al. [15] and Hajbabaei et al. [16] discussed above. Specifically, these earlier studies showed that fuel composition can have important emissions impacts in lean-burn NG engines in refuse haulers and transit buses, while stoichiometric TWC-equipped NG engines in a transit bus and a refuse hauler did not show significant emissions differences for different NG fuels. This study aims at evaluating the impact of NG composition on the criteria emissions, carbonyl compounds, and particulate matter emissions from a legacy lean-burn NG engine school bus with an oxidation catalyst and two current technology stoichiometric combustion NG Class 8 trucks (vehicles with gross vehicle weight ratings ranging from 15 to 27 tons) equipped with TWCs. Given the emissions changes seen in the legacy lean-burn NG refuse hauler and transit bus, it is important to evaluate the potential emissions impacts of NG fuel composition for school buses that represent an important vehicle category that are commonly equipped with legacy lean-burn NG engines that has not been studied in terms of emissions impacts with changing NG composition. Similarly, while transit buses and refuse haulers with stoichiometric TWC-equipped NG engines have not shown strong fuel effects, it is important to also evaluate the impacts of NG fuel composition for class 8 trucks equipped with stoichiometric NG engines and TWCs, which were not included in the previous studies but represent an important segment of the heavy-duty NGV fleet that operate in urban port areas, such as the Ports of Los Angeles and Long Beach. Testing was conducted on a range of seven fuels with varying Wobbe numbers and methane numbers. Gaseous and particulate emission results are discussed in the context of changing fuel composition, along with the influences of the driving cycle and engine technology.

2. Experimental

2.1. Test vehicles and fuels

One school bus equipped with a 2005 lean-burn John Deere 8.1 L 6081H engine and an oxidation catalyst and two Class 8 trucks equipped with stoichiometric engines and cooled exhaust gas recirculation (EGR) systems and TWCs were employed in this study. The stoichiometric NGVs included a truck with a 2012 Cummins Westport ISL-G 8.9 L engine and a truck with a 2013 Cummins Westport ISX12-G engine. The test weights for the vehicles were 16.8 tons for the school bus and 28 tons for the two Class 8 trucks.

Seven fuels were employed for this study including three high methane number fuels and four high Wobbe number fuels. Fuels H1 and H2 represent historical baseline gases for Southern California and they are based on actual pipeline data. Fuel H1 was representative of Texas Pipeline gas and served as the baseline fuel, while Fuel H2 was representative of Rocky Mountain Pipeline gas. Fuel LM3 is representative of Peruvian liquefied natural gas (LNG) that was modified to meet a Wobbe number of 1385, which is a typical pipeline specification, and a methane number of 75. Fuel LM4 was representative of Untreated Middle East LNG with a high Wobbe number (above 1400). Fuel LM5 and Fuel LM6 were hypothetical fuels with compositions designed to evaluate whether two fuels with the same Wobbe number and methane number, but different compositions, would produce different exhaust emissions. Fuel H7 was a compressed natural gas (CNG) blend produced from a LNG fuel tank. Fuel H7 had almost no inert components because inerts were removed during the liquefaction process. The main properties of the test fuels are presented in Table 1. Note that not all fuels were tested on each vehicle; testing for the John Deere vehicle was conducted on all seven fuels, since previous studies showed strong fuel effects on tailpipe emissions

Table 1
Main properties of the test fuels.

Fuels #	Description	Methane	Ethane	Propane	I-butane	N_2	CO_2	MN	Wobbe #	HHV	H/C ratio
H1	Baseline, Texas Pipeline	96	1.8	0.4	0.15	0.7	0.95	99	1338	1021	3.94
H2	Baseline, Rocky Mountain Pipeline	94.5	3.5	0.6	0.3	0.35	0.75	95	1361	1046	3.89
LM3	Peruvian LNG	88.3	10.5	0	0	1.2	0	84	1385	1083	3.81
LM4	Middle East LNG-Untreated	89.3	6.8	2.6	1.3	0	0	80	1428	1136	3.73
LM5	High Ethane	83.65	10.75	2.7	0.2	2.7	0	75.3	1385	1115	3.71
LM6	High Propane	87.2	4.5	4.4	1.2	2.7	0	75.1	1385	1116	3.70
LM7	L-CNG fuel (school bus)	95.24	4.39	0.11	0.01	0.25	0	97	1352	1029	3.91
H7	L-CNG fuel (ISX12 G truck)	94.63	4.61	0.14	0.02	0.55	0	96	1347	1027	3.91

MN = Methane Number determined via CARB calculations; Wobbe # = HHV/square root of the specific gravity of the blend with respect to air; HHV = Higher Heating Value; H/C = ratio of hydrogen to carbon atoms in the hydrocarbon portion of the blend.

from older technology lean-burn natural gas engines [15]. The Cummins ISL-G was tested on H1, LM5, and LM6, whereas the Cummins ISX12-G was tested on H1, LM4, LM5, and H7. Fuel selection for the current technology stoichiometric vehicles was based on findings from previous studies, indicating that high Wobbe number/heavier hydrocarbon content fuels could alter the emissions profile in the tailpipe [16]. It should also be mentioned that each vehicle was filled from a different station for H7.

2.2. Driving cycle and measurement protocol

The school bus was exercised on all seven fuels over the Central Business District (CBD) cycle. Note that six tests were run on each fuel over a specially developed CBD cycle, which consisted of a single CBD cycle as a warm-up, followed by two iterations (i.e., a double) CBD cycle. The CBD cycle was repeated twice to provide a sufficient particle sample for analysis. More details for the CBD cycle are provided in the Supplementary Material. The Cummins ISL-G and Cummins ISX12-G trucks were exercised over the Near Dock duty cycle and the Local Haul duty cycle, respectively. Both cycles are segments of the drayage truck port cycle developed by TIAX in conjunction with the Ports of Long Beach and Los Angeles. Information about both test cycles is given in the Supplementary Material.

All tests were conducted at CE-CERT's Heavy-Duty Chassis Dynamometer facility. Emissions measurements were obtained using the CE-CERT Mobile Emissions Laboratory (MEL). The facility and sampling setup have been described in detail previously and are only discussed briefly here [18]. For all tests, emissions measurements of THC, NMHC, CH_4, CO, NO_x, CO_2, and PM, were measured using standard instruments, as shown in the Supplementary Material. Measurements of ammonia (NH_3) were also obtained on a real-time basis using a Unisearch Associates Inc. LasIR S Series tunable diode laser (TDL) near infrared absorption spectrometer. Measurements of nitrous oxide (N_2O) were made using a Fourier Transform Infrared (FTIR) instrument. Carbonyl emissions, PM mass, particle number, and particle size distributions were also measured. Detailed information on the method used to collect and analyze these compounds is provided in the Supplementary Material.

3. Results and discussion

The figures for each pollutant, presented in the following sections, show results for each vehicle/fuel/cycle combination based on the average of tests conducted on that particular test combination. The error bars on the figures are the standard deviation over all tests for test combination. The statistical analyses were conducted using a 2-tailed, 2 sample equal variance *t*-test. For the statistical analyses, results are considered to be statistically

significant for $p \leqslant 0.05$, or marginally statistically significant for $0.05 < p \leqslant 0.1$.

3.1. Regulated emissions

Emissions of NO_x are shown in Fig. 1a for the John Deere school bus and the Cummins trucks. For the John Deere bus, NO_x emission levels were clearly influenced by the fuel composition, with the low methane/higher Wobbe number fuels resulting in higher NO_x emissions compared to the higher methane number fuels. These increases ranged from 14.2% to 35.1%, at a statistically significant level. Comparing the Class 8 trucks, it was observed that the Cummins ISX12-G truck produced substantially lower NO_x emission levels than the Cummins ISL-G truck. This could be attributed to the smaller ISL-G engine requirement to work 'harder' than the ISX12-G engine in carrying a similar load. It was also observed that for both trucks, the low methane number fuels generally showed lower NO_x levels than the high methane number fuels. This is opposite to the trends for the lean-burn John Deere engine, where the low methane number fuels clearly produced higher NO_x emissions than the baseline fuels. For the Cummins ISL-G truck, NO_x emissions showed some trends toward lower emissions with the low methane/high Wobbe number fuels compared to H1. In particular, NO_x emissions showed statistically significant reductions of 36.7% for LM6 compared to H1. For the Cummins ISX12-G truck, NO_x emissions generally showed weak trends between fuels with the exception of LM5, which showed marginally statistically significant reductions of 19.3% and 20.2%, respectively, compared to H1 and H7.

The increases in NO_x emissions with LM3, LM4, LM5, and LM6 for the lean-burn engine fitted with the oxidation catalyst could be attributed to the presence of high molecular-weight hydrocarbons in these fuels. The addition of higher hydrocarbons (ethane and propane) can increase the adiabatic flame temperature or the adiabatic flame speeds of the fuels. Flame speed is dependent on both the composition of the fuel and the amount of dilution in the combustion mixture. High Wobbe number/low methane number fuels with increased concentrations of ethane and propane have caused increased flame speed and subsequently combustion temperature increases [17]. Previous studies have also shown that lean-burn engines run richer as methane number decreases [3,17]. This can lead to the oxidation of more fuel, higher combustion temperatures, and increased cylinder pressures. It is also possible that the higher hydrocarbons promote the formation of reactive radicals, which result in increased formation of prompt NO_x. The results reported here are also in agreement with previous studies that have reported higher NO_x emissions with low methane number fuels from lean-burn engines [15,16,19,20].

The lower NO_x emission levels for the Cummins trucks can be due to the TWC [8]. The newer stoichiometric engines also employed cooled EGR that introduces inert exhaust gases into the combustion cylinder, reducing cylinder combustion
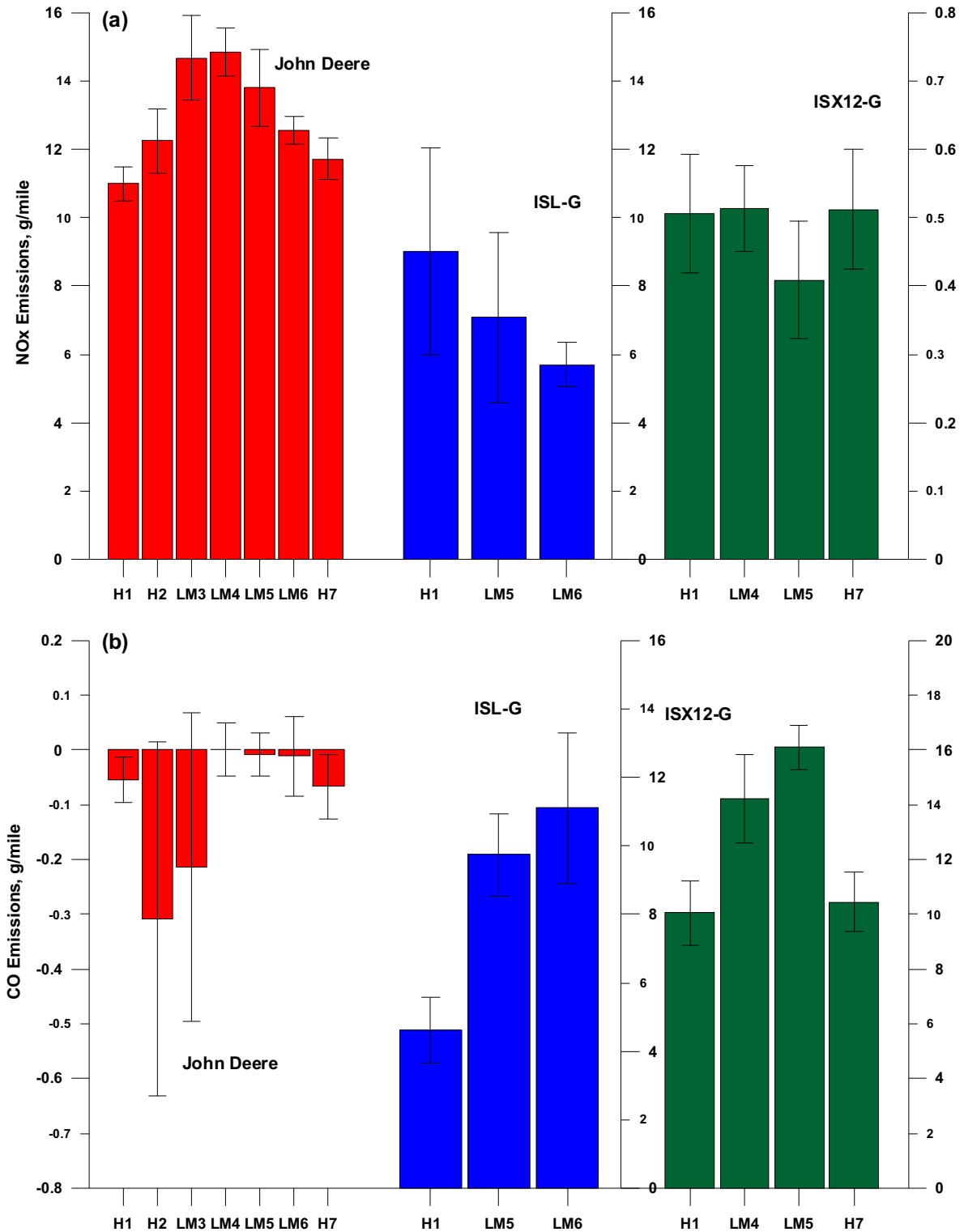
Fig. 1. NO$_x$ (a) and CO (b) emissions for the lean-burn John Deere bus and the stoichiometric Cummins trucks. The error bars represent one standard deviation of the average values.

temperatures and resulting in lower NO$_x$ emissions [9]. The slight decrease in NO$_x$ emissions observed for the low methane fuels may be due to slightly richer air/fuel (A/F) ratios for stoichiometric combustion. The resultant decrease in oxygen may also lead to increased effectiveness in the TWC's ability to further reduce NO$_x$ emissions. Stoichiometric engines generally exhibit tighter A/F ratio control, so any change in the A/F ratio should be slight with

minimal engine effects. However, along with decreases in NO$_x$ emissions from operation on low methane fuels, both Cummins trucks exhibited increased CO emissions as discussed in the following paragraphs, which is consistent with slightly richer combustion.

Fig. 1b presents the CO emissions for the John Deere school bus and the Cummins trucks as a function of the different fuels. The CO

emissions for the John Deere school bus were found at very low levels that were close to the measurement background, with no strong effects between the test fuels. The low CO levels were due to the high conversion efficiency of the oxidation catalyst as a result of the excess oxygen available in the exhaust of the lean-burn engine. CO emission results from the Cummins engines were significantly higher than those from the lean-burn John Deere engine. This can be attributed to the impact of richer operating conditions and the lower oxygen concentration for the stoichiometric combustion compared to lean-burn combustion [21]. Specifically, richer combustion will lead to both increased engine-out CO as well as a reduction in the efficiency of removing CO over the catalyst. Similar findings were also seen in previous studies [9,16]. The fuel effect in CO emissions was particularly noticeable for both Cummins trucks, with LM5 and LM6 producing statistically significant increases of 111.1% and 140.8%, respectively, compared to H1 for Cummins ISL-G. For the Cummins ISX12-G truck, CO emissions showed statistically significant increases of 41.5% and 60.3%, respectively, for LM4 and LM5 compared to H1.

For the John Deere school bus, reductions in THC emissions were seen for the fuels with lower methane numbers and higher hydrocarbon contents than the higher methane number fuels, as shown in Fig. 2a. THC emissions showed statistically significant reductions ranging from 8.2% to 17.4% for the low methane fuels and H7 compared to H1. Similar to H1, the low methane number fuels exhibited statistically significant reductions in THC emissions compared to H2. For the Cummins ISL-G truck, THC emissions showed statistically significant reductions of 20.5% and 15.7%, respectively, for LM5 and LM6 compared to H1. For the Cummins ISX12-G truck, THC emissions did not show strong differences between the test fuels, although some trends toward lower THC emissions for the low methane fuels were observed. It is worth noting that THC emissions were significantly lower for the stoichiometric Cummins engines compared to the John Deere lean-burn engine. This can be attributed to the differences in the engine technology, since the older technology lean-burn John Deere engine was fitted with an oxidation catalyst designed to meet an earlier certification standard, while the stoichiometric Cummins engines were fitted with TWC devices designed to meet a more recent, more stringent certification standard. All test vehicles showed trends of higher THC emissions for the fuels with higher methane contents, which is consistent with results previously reported by other authors [15–17]. This was probably due to the fact that the THC emissions from all engine/aftertreatment types were predominately methane, while the NMHC emissions were very low. Another factor that may have impacted THC levels was the increased flame speeds of the low methane number fuels that under higher combustion and exhaust temperatures resulted in more complete oxidation of the fuel in the catalyst.

Emissions of NMHC showed strong fuel trends for the John Deere bus, with the lower methane number fuels producing higher NMHC emissions than the higher methane number fuels (Fig. 2b). The NMHC emissions showed statistically significant increases ranging from 26.3% to 133.2% for H2 and the low methane fuels compared to H1. Fuels H2 and H7 both showed similar reductions in NMHC emissions compared to low methane number fuels. For the Cummins ISL-G truck, the only statistically significant increase was observed for LM5 relative to H1. For the Cummins ISX12-G truck, NMHC emissions showed stronger trends for the low methane fuels, with LM4 and LM5 showing marginally statistically significant and statistically significant increases of 253% and 219.8%, respectively, compared to H1. Overall, all three vehicles emitted very low levels of NMHC emissions compared to THC emissions, with the NMHC emissions for the stoichiometric trucks close to background levels. Previous studies have also shown that

NMHC emissions increased with low methane fuels for lean-burn engines [15–17]. THC emissions from natural gas engines are predominately unburned fuel, therefore, the non-methane hydrocarbon fraction of THC exhaust emission typically trends with the percentage of non-methane hydrocarbons in the test fuel.

3.2. Greenhouse gas emissions and fuel economy

CH_4 is a greenhouse gas with a global warming potential 25 times higher than CO_2 that is emitted directly from vehicles at the tailpipe as a result of fuel combustion. CH_4 emission levels have been detected as a problem in both lean-burn and stoichiometric natural gas engines with oxidation catalysts and TWCs, respectively. The emissions of CH_4 for the John Deere school bus and the Cummins trucks are shown in Fig. 3. For the John Deere school bus, CH_4 emissions showed clear reductions for the fuels with lower methane numbers and higher hydrocarbon contents, ranging from 9.1% to 26.5% compared to H1. Similar statistically significant reductions in CH_4 emissions were also seen for H2 and H7 when compared to low methane fuels. For the Cummins ISL-G truck, the CH_4 emissions showed statistically significant decreases of 25.6% and 12.4%, respectively, for LM5 and LM6 compared to H1. For the Cummins ISX12-G truck, the CH_4 emissions showed a statistically significant increase of 19.2% for H7 compared to H1, while statistically significant reductions in CH_4 emissions for H1, LM4, and LM5 compared to H7 were also observed. In general, the higher CH_4 emissions seen for the higher methane number fuels could be due to the fact that CH_4 is less reactive from a combustion standpoint than higher chain hydrocarbons, so it is more likely to go through the combustion process unburned and pass unreacted with oxygen across the catalyst [7,22]. Higher chain hydrocarbons usually dissociate via the breaking up of the C–C bonds rather than the C–H bonds, which usually have much higher bond dissociation energies.

Although limited nitrous oxide (N_2O) is produced in aftertreatment systems, it is included in recent greenhouse gas regulations, which count N_2O as CO_2 equivalents. This is because, according to the Fifth Assessment Report (AR5), N_2O has a lifetime of approximately 121 years in the atmosphere and a Global Warming Potential (GWP) of 265 based on a 100 year time horizon (265 times more powerful than CO_2 in terms of heat trapping effects) [23]. N_2O emissions can form over the surface of a TWC, but not over an oxidation catalyst. For the present vehicles, N_2O emissions are relatively low, with the newer TWC vehicles emitting at fairly low levels and the John Deere bus not having a mechanism to form N_2O emissions, as it is only equipped with an oxidation catalyst. For the John Deere bus, N_2O emissions trended higher for the low methane fuels compared to the high methane fuels, with the exception of LM3 (Fig. 4). Emissions of N_2O showed marginally statistically significant increases ranging from 38% to 79% for the low methane fuels compared to H1. Analogous to the John Deere bus, both trucks showed higher N_2O emissions for the low methane fuels. However, the differences in N_2O emissions between the test fuels were not statistically significant for either of the Cummins vehicles. The observed variability in N_2O emissions for all vehicle/fuel combinations was likely due to the very low N_2O emission levels.

Overall, our results showed that selectivity toward N_2O emissions was highly dependent on fuel composition. All vehicles showed a systematic increase in N_2O emissions with the low methane fuels, independent the engine technology and aftertreatment control. N_2O forms as an intermediate during the catalytic reduction of NO to molecular nitrogen (N_2). At high temperatures, NO is directly reduced to N_2; however, at lower temperatures, N_2O is an intermediate product [24–26]. Some of the reactions that promote the formation of N_2O include reactions with CO and hydrogen on the catalyst surface ($2NO + CO \rightarrow N_2O + CO_2$; $2NO + H_2 \rightarrow N_2O + H_2O$), which are more prevalent during rich combustion
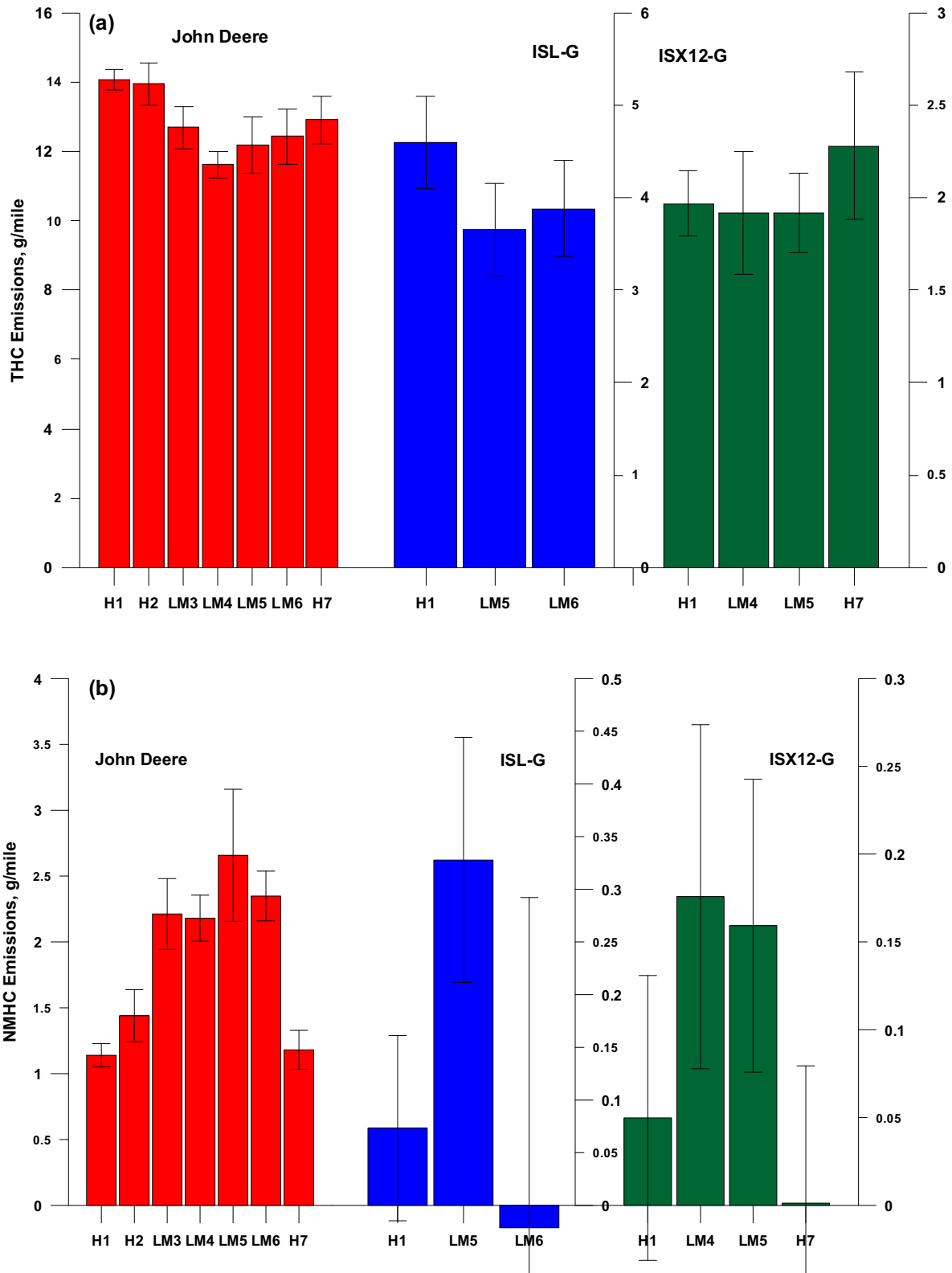
Fig. 2. THC (a) and NMHC (b) emissions for the lean-burn John Deere bus and the stoichiometric Cummins trucks. The error bars represent one standard deviation of the average values.

[26,27]. Hence, the increases in N_2O emissions for the low methane fuels were consistent with the corresponding increases seen for CO emissions for these fuels, especially for the stoichiometric Cummins trucks. Increases in CO would also give rise to higher hydro-

gen levels on the catalyst surface due to the water–gas shift reaction [26,27]. For the lean-burn engine, CO emissions were very low due to the presence of an oxidation catalyst and did not follow the same patterns as N_2O emissions. However, N_2O emissions cor-
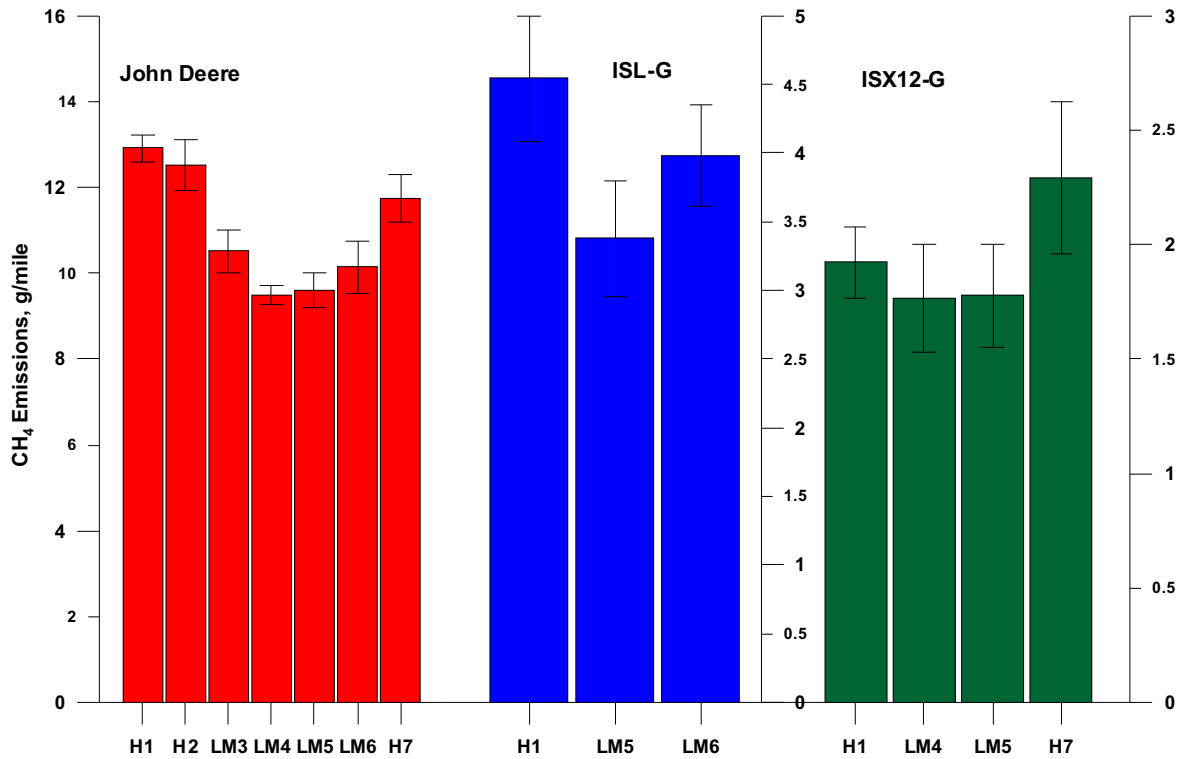
Fig. 3. CH_4 emissions for the lean-burn John Deere bus and the stoichiometric Cummins trucks. The error bars represent one standard deviation of the average values.
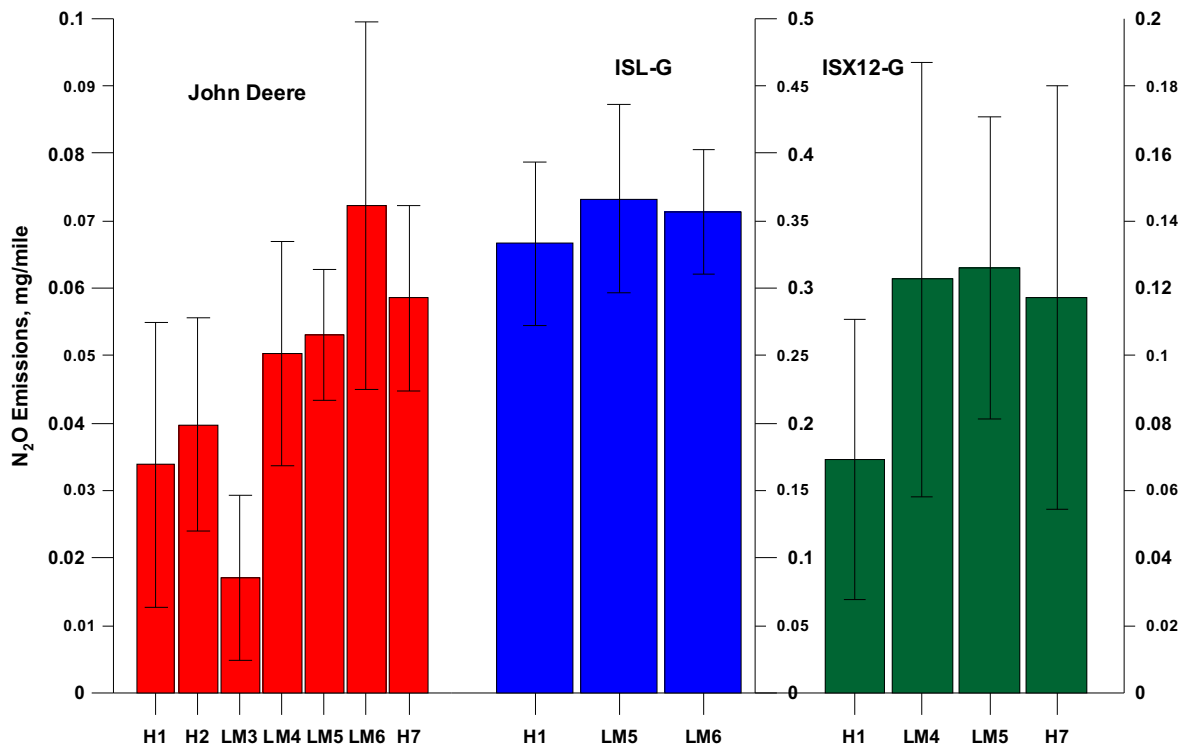

Fig. 4. N_2O emissions for the lean-burn John Deere bus and the stoichiometric Cummins trucks. The error bars represent one standard deviation of the average values.

roborate with NO_x emissions for this vehicle, suggesting that the low methane fuels produced more nitrogenous species under the present test conditions.

CO_2 emissions for test vehicles as a function of the different fuels are shown in Table S1 (Supplementary Material). For the John Deere bus, the CO_2 emissions did not show strong trends between the test fuels over the CBD cycle, with the exception of H7 which showed a statistically significant decrease of 2.3% in CO_2 emissions compared to H1. Compared to H7, most fuels showed statistically significant increases in CO_2 emissions ranging from 2.3% to 3.5%,

with the exception of H2. Similar to the lean-burn engine, both stoichiometric engines did not show strong fuel effects in CO_2 emissions.

Table S1 (Supplementary Material) also presents the global warming potential (GWP) of each test vehicle/fuel combination. The GWP is expressed for each driving cycle in CO_2 equivalent (CO_2eq) units. For CH_4 and N_2O emissions, the GWP was calculated by converting these GHGs to CO_2eq assuming a 100 year GWP of 36 and 298 times GWP of CO_2, respectively. The GWP of the stoichiometric Cummins trucks was found to be higher than the lean-burn school bus. This result was attributable to the higher CO_2 emissions produced for the stoichiometric engines compared to the lean-burn engine, which significantly contributed to the total GWP of the exhaust emissions. Similar findings have been reported in previous studies [9]. CH_4 emissions for the stoichiometric vehicles did not show a large contribution to the total GWP of the exhaust emissions, whereas N_2O emissions for the stoichiometric vehicles showed a larger contribution to the total GWP of the exhaust emissions.

The volumetric fuel economy and the fuel economy calculated on a gasoline gallon equivalent (GGE) energy basis for all test vehicles are shown in Table S1 (Supplementary Material). For the John Deere, for the volumetric fuel economy, the low methane fuels with the higher heating values showed higher fuel economy compared to H1, H2, and H7. The fuel economy increases for LM3, LM4, LM5, and LM6 compared to H1 were all statistically significant ranging from 4% to 9.3% compared to H1. Fuel economy on a GGE energy basis did not show strong fuel effects with the exception of H7, which showed a statistically significant increase of 1.4% compared to H1. Volumetric fuel economy for the Cummins ISL-G truck showed statistically significant increases of 10.4% and 11.5%, respectively, for LM5 and LM6 compared to H1. Fuel economy on a GGE energy basis did not show any strong trends between the test fuels for the Cummins ISL-G truck over the Near Dock duty cycle (Table S1, Supplementary Material). For the Cummins ISX12-G truck, the volumetric fuel economy showed statistically significant increases of about 12% for both LM4 and LM5 and a marginally statistically significant increase of 4.2% for H7 compared to H1. Fuel economy on a GGE energy basis showed a marginally statistically significant increase of 2.7% for LM5 compared to H1. Overall, for both Cummins trucks, the low methane fuels with higher energy contents showed some trends of higher energy equivalent fuel economy compared to the H1.

3.3. Ammonia emissions

Emissions of NH_3 were found to be at substantially lower levels for the lean-burn John Deere vehicle compared to the stoichiometric engines, which ranged from 27.5 mg/mile to 51 mg/mile (Fig. 5). In general, NH_3 emissions for the John Deere bus showed a declining trend for all fuels compared to H1. Although emissions of NH_3 showed some statistically significant differences between the fuels, there were no consistent fuel effects since most of the differences were between the high methane number fuels. Emissions of NH_3 for both Cummins trucks were higher for the low methane fuels compared to H1 and H7. For the Cummins ISL-G truck, NH_3 emissions showed statistically significant increases of 36.7% and 17.4%, respectively for LM5 and LM6 compared to H1. For the Cummins ISX12-G truck, NH_3 emissions showed statistically significant increases of 28.9% and 35.1%, respectively, for LM4 and LM5 compared to H1. This could be due to the fact that the low methane number/higher flame speed fuels could produce higher exhaust temperatures and possibly slightly richer A/F ratios. Therefore, the conditions for the formation of hydrogen as a precursor and NH_3 as reaction product could be enhanced for the lower methane number fuels.

For TWC-equipped stoichiometric NG engines, the production of NH_3 takes place in the presence of hydrogen molecules, which in turn are produced during periods of rich air–fuel mixtures. Hydrogen could be either formed due to a water gas shift reaction involving CO and water or steam reforming reactions involving CH_4 and water in the exhaust [27–29]. It has been suggested that hydrogen produced in the water–gas shift reaction ($CO + H_2O \leftrightarrow CO_2 + H_2$) could be a major contributor to NH_3 formation through the overall reaction of $2NO + 2CO + 3H_2 \rightarrow 2NH_3 + 2CO_2$ [29]. It was expected that an increase in the proportion of CH_4 in the engine-out exhaust gas flux for the high methane number fuels would decrease the quantity of hydrogen available for NH_3 formation, due to the lower reactivity of CH_4 over the TWC. This could be a plausible explanation for the lower NH_3 emissions observed for the high methane fuels compared to the higher hydrocarbon content fuels. The presence of higher levels of CO also facilitates the formation of NH_3 in the exhaust of a TWC-equipped stoichiometric NG vehicle via the reaction described above. Under the present test conditions, the low methane fuels showed higher CO emissions resulting in higher NH_3 emissions. It should also be noted that NH_3 emissions from stoichiometric NG engines with TWC are generally higher compared to those found from current technology heavy-duty diesel vehicles equipped with selective catalytic reduction (SCR) [30].

3.4. Carbonyl emissions

For all vehicles, low molecular weight aldehydes, such as formaldehyde followed by acetaldehyde, were the predominant compounds in the tailpipe, as shown in Table S2 (Supplementary Material). Our results are consistent with previous studies showing that formaldehyde and acetaldehyde were the most abundant aldehydes in NGV exhaust [31–33]. For all three vehicles, formaldehyde dominates the exhaust as a result from the high methane content of the fuels, since formaldehyde is an intermediate step in the oxidation of methane [34]. This was as expected since the fuel molecule dominates the THC emissions, with lower-carbon-numbered hydrocarbons making-up virtually all of the rest. The lean-burn vehicle resulted in substantially higher carbonyl emissions than the stoichiometric trucks. For the John Deere bus, formaldehyde emissions did not show strong fuel trends over the CBD cycle, with the exception of some differences for LM3 and LM4. Formaldehyde emissions followed the trends of the THC emissions as opposed to the trends of the NMHC emissions, with the lower methane fuels generally producing higher formaldehyde emissions. Formaldehyde is produced by oxidation of ethane. Acetaldehyde emissions exhibited clearer trends, with the lower methane number/higher Wobbe number fuels producing higher concentrations of acetaldehyde emissions, since acetaldehyde is also produced from the oxidation of ethane. Overall, acetaldehyde emissions showed increases ranging from 29.3% to 50.6% that were statistically significant or marginally statistically significant. Crotonaldehyde emissions showed strong increases with the low methane number fuels compared to H1 and H2, at a statistically significant level. Carbonyl emissions for the ISL-G truck were found to be at much lower levels than those for the ISX12-G truck. For the ISL-G truck, only formaldehyde and acetaldehyde were detected in the tailpipe, with heavier carbonyl compounds being below the detection limits of the method. For the ISL-G truck, there were no statistically significant differences between the test fuels for either the formaldehyde or the acetaldehyde emissions over the Near Dock duty cycle. For the ISX12-G truck, formaldehyde emissions did not show strong fuel trends, whereas acetaldehyde emissions showed statistically significant decreases of 61.6% and 67.8%, respectively, for LM5 compared to H1 and H7.
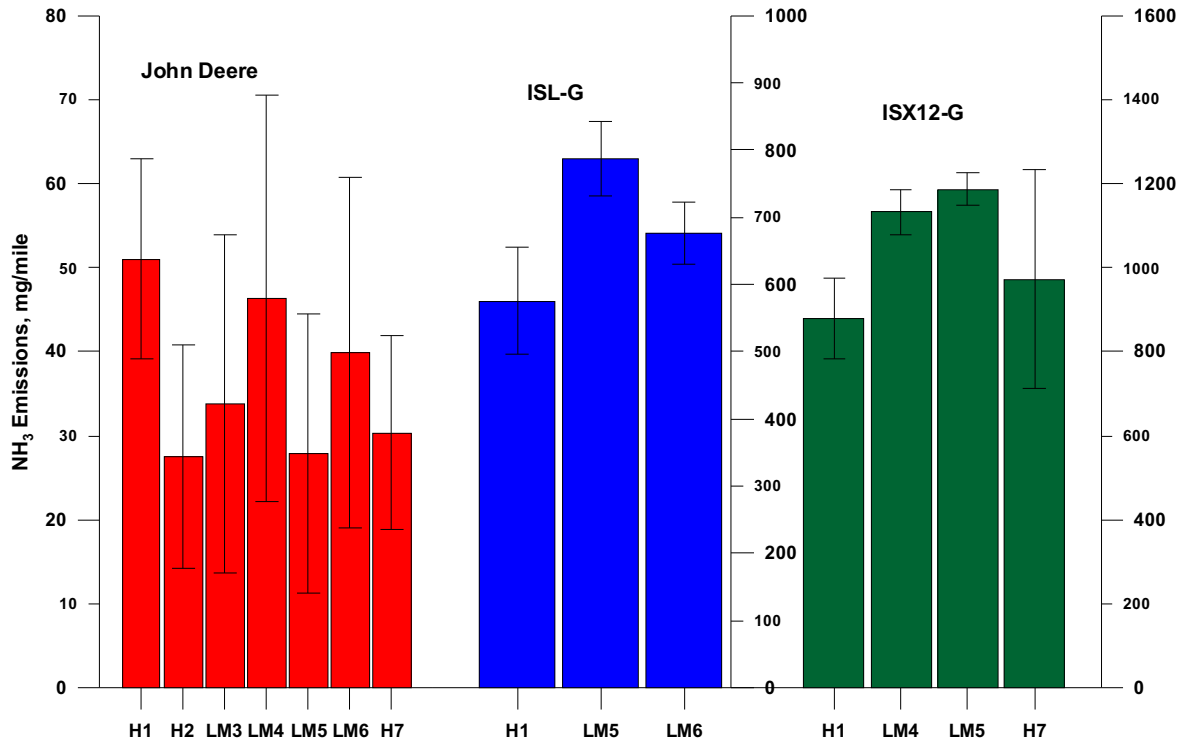
Fig. 5. NH$_3$ emissions for the lean-burn John Deere bus and the stoichiometric Cummins trucks. The error bars represent one standard deviation of the average values.

3.5. Particulate emissions

PM mass emissions are shown in Table 2 for all test vehicles. The results indicated that PM mass emissions were low for the John Deere bus on an absolute level, and in some cases close to the background levels. PM emissions for the stoichiometric engines were also found in low levels, with the Cummins ISL-G truck producing significantly lower PM mass emissions than the Cummins ISX12-G truck. Although some differences were seen between different fuels for different vehicles, overall the results did not show any consistent fuel trends for PM mass for the test vehicles. NG is primarily comprised of methane, which is the lowest molecular weight hydrocarbon, does not have carbon–carbon molecular bonds, and has a simpler structure compared to diesel and gasoline

fuels [35] Additionally, the lack of carbon bonding results in a substantially lower probability of benzene ring formation and hence lowers PM emissions [10]. The main source of PM in NG engines is considered to be the entry of engine lubricating oil into the combustion chamber [35]. Previous studies have shown that lubricant-oil-based additives and wear metals were a major fraction of the PM mass from NG buses [31,36,37].

Table 2 shows the total particle number (PN) emissions for the test vehicles. For the John Deere bus, the low methane number and higher flame speed fuels trended lower in PN emissions when compared to the higher methane number fuels. However, most of the differences between the fuels cannot be considered as statistically significant, with some exceptions seen for some fuels. Both stoichiometric trucks produced significantly lower PN emissions than the lean-burn vehicle, suggesting higher oil consumption for the lean-burn engine. For the Cummins ISL-G truck, PN emissions did not show any statistically significant differences between the test fuels. For the Cummins ISX12-G truck, PN emissions were higher than those of the ISL-G truck, with LM4 showing a statistically significant increase in PN emissions of 52.7% compared to H1.

Fig. 6(a–c) illustrates the average particle size distributions for the John Deere bus (a), Cummins ISL-G truck (b), and Cummins ISX12-G truck (c). The John Deere bus showed higher concentrations of nucleation and accumulation mode particles than the stoichiometric trucks. The nucleation mode particles appeared to dominate the particle size distribution profile for this vehicle over the CBD cycle. The particle size distributions for most fuels showed particle concentrations in the accumulation mode between 2×10^6 and 4×10^6 particles/cm^3 for particle diameters from 140 to 145 nm size range, which was much lower than those in the nucleation mode. It should be noted that there were no clear fuel trends in particle size distributions for the John Deere bus.

The Cummins ISL-G truck showed a decidedly bimodal particle size distribution. However, the accumulation mode is the prevalent mode of particle formation for this engine as opposed to the John

Table 2
PM mass and total particle number emissions for the lean-burn John Deere bus and the stoichiometric Cummins trucks.

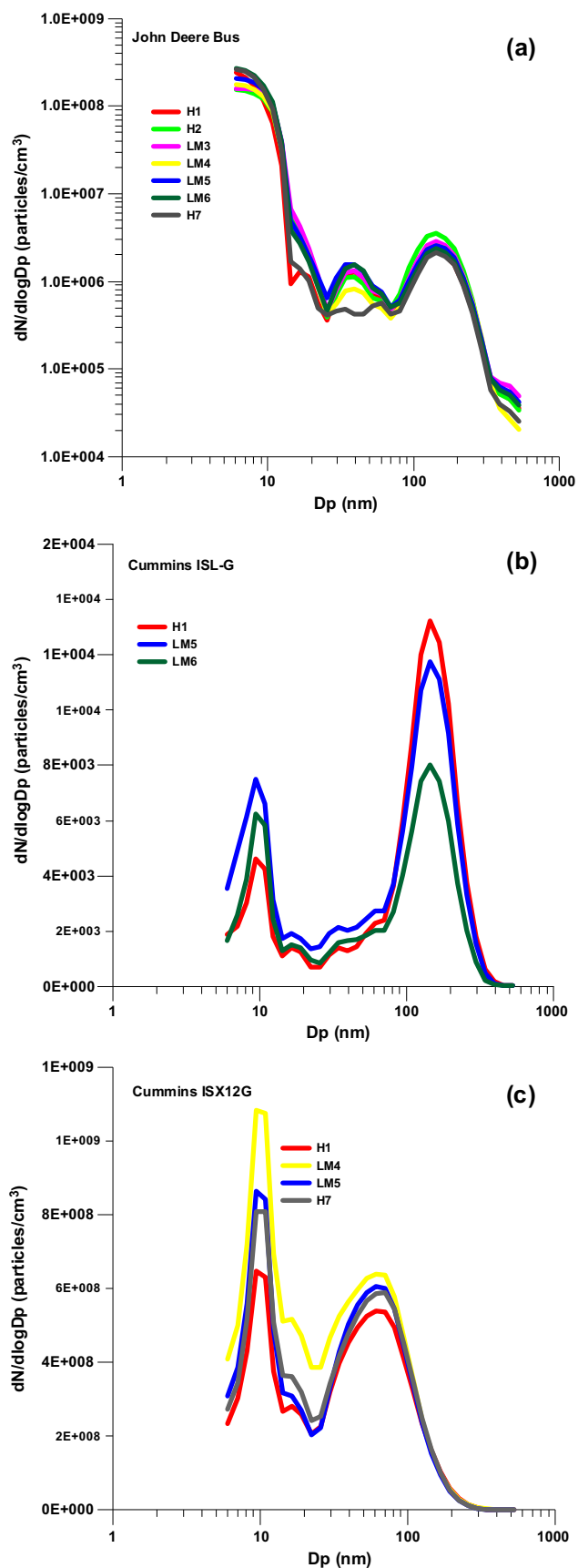| Fuel	John Deere	
	PM Mass (mg/mile)	Particle Number (particles/mile)
H1	0.0030 ± 0.0022	1.618E+14 ± 7.700E+12
H2	0.0059 ± 0.0029	1.461E+14 ± 2.136E+13
LM3	0.0096 ± 0.0021	1.479E+14 ± 2.696E+13
LM4	0.0039 ± 0.0031	1.462E+14 ± 1.635E+13
LM5	0.0013 ± 0.0013	1.542E+14 ± 2.065E+13
LM6	0.0029 ± 0.0024	1.771E+14 ± 2.183E+13
H7	0.0037 ± 0.0031	1.697E+14 ± 1.171E+13
Cummins ISL-G		
H1	0.0029 ± 0.0009	1.92E+12 ± 3.8E+11
LM5	0.0044 ± 0.0017	2.82E+12 ± 1.14E+12
LM6	0.0027 ± 0.0019	1.87E+12 ± 3.01E+11
Cummins ISX12-G		
H1	0.0177 ± 0.0084	6.08E+13 ± 1.72E+13
LM4	0.0255 ± 0.0054	9.29E+13 ± 1.55E+13
LM5	0.0195 ± 0.0033	7.33E+13 ± 8.7E+12
H7	0.0183 ± 0.0038	7.12E+13 ± 1.67E+13

Fig. 6. Average particle size distributions for the lean-burn John Deere bus (a), the stoichiometric Cummins ISL-G truck (b), and the stoichiometric Cummins ISX12-G truck (c).

Deere bus engine. The particle size distributions for the three test fuels showed particle concentrations in the nucleation mode from 5×10^3 to 8×10^3 for particle diameters centered from 9 to 10 nm in size. The highest concentrations in nucleation mode particles were seen for the low methane fuels, i.e., LM5 and LM6, compared to H1. For the accumulation mode particles, particle diameters ranging from 140 to 143 nm in size range with particle concentrations from 8×10^3 to 1×10^4. The high methane number H1 produced higher concentrations of accumulation mode particles followed by LM5 and LM6. Similar to the previous vehicles, the ISX12-G truck showed a decidedly bimodal particle size distribution. Unlike the particle size distribution profile for the ISL-G truck, but more similar to the school bus, the ISX12-G truck produced higher concentrations of nucleation mode particles compared to the accumulation mode particles. The particle size distributions for the test fuels showed particle concentrations in the nucleation mode from 6×10^8 to 1×10^9 for particle diameters centered from 9 to 10 nm in size. The particle concentrations in the nucleation mode for the ISX12-G truck were considerably higher when compared to the other vehicles tested. For the accumulation mode particles, particle concentrations were from 5×10^8 to 6×10^8 for particle diameters from 60 to 70 nm in size. Compared to ISL-G truck, the ISX12-G truck produced higher accumulation mode particle counts with smaller diameters. For the ISX12-G truck, a fuel effect was noticeable with the low methane/higher flame speed fuels showing higher accumulation and nucleation mode particle concentrations than H1 and H7.

In general, it is reasonable to theorize that the observed particle size distributions could be attributed to in-cylinder combustion of lubricant oil, which contributed sulfates nucleating with water to form sulfuric acid particles in the 10 nm peak size. Similar observations were reported by Thiruvengadam et al. [36] when they tested two 2007 NG buses fitted with Cummins ISLG280 engines and TWCs. The entry of lubricant oil into the combustion chamber is dependent on engine load. Typically low-load operations, such as those applied during the CBD for the John Deere bus, result in insufficient sealing of the piston rings, which can contribute to the combustion of lubricant oil [36]. It is also reasonable to assume that the low-load operation of CBD resulted in lower accumulation mode particles or soot emissions and increased the probability of the formation of inorganic nucleation mode particles. Lower accumulation particles were also observed for the Cummins ISL-G truck when operated over the Near Dock duty cycle compared to the Cummins ISX12-G truck. This result could be due to the third phase of the Local Haul duty cycle (long high-speed transient phase) where the vehicle was subjected to lower speed and load conditions compared to the third phase of the Near Dock duty cycle (short high-speed transient phase). The origin of nucleation mode particles observed under the present test conditions was mainly due to lubricant oil from the NG engine. This phenomenon has been explained by Khalek et al. [38] showing that lubricant oil additives do undergo volatilization when passing through the combustion chamber and a fraction of them renucleate to form nanoparticles.

4. Conclusions

This study revealed that the use of new stoichiometric combustion NG engines with TWCs in urban areas are capable of reducing THC, NMHC, CH_4, and NO_x emissions compared to older lean-burn engines with oxidation catalysts. Consistent with other studies in the literature, NH_3 emissions formed de novo in TWCs showed large increases with the stoichiometric vehicles. The increases in NH_3 emissions with current technology heavy-duty NGVs could have important environmental implications, since NH_3 is involved

in the formation of secondary aerosols and is also considered as a toxic pollutant. Stoichiometric combustion showed beneficial results in particle number emissions compared to lean combustion. The study does reveal that NG composition plays a significant role in the formation of most harmful emissions from older technology engines, while current engine platforms are not strongly affected by altering fuel composition. This was the case for the lean-burn engine with the oxidation catalyst, which showed higher NO_x and NMHC emissions and improved fuel economy on a volumetric basis, but lower THC and CH_4 emissions on low methane number fuels.

Acknowledgements

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.fuel.2016.02.034.

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