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Authors

Hajbabaei, Maryam
Karavalakis, Georgios
Johnson, Kent C
et al.

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Impact of natural gas fuel composition on criteria, toxic, and particle emissions from transit buses equipped with lean burn and stoichiometric engines

Maryam Hajbabaei [a], Georgios Karavalakis [a], Kent C. Johnson [a], Linda Lee [b], Thomas D. Durbin [a,*]

[a] University of California, Riverside, Bourns College of Engineering, Center for Environmental Research and Technology (CE-CERT), 1084 Columbia Avenue, Riverside, CA 92507, USA
[b] California Air Resources Board (CARB), 1001 I Street, Sacramento, CA 95814, USA

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ABSTRACT

This study investigated the impacts of varying natural gas composition on the exhaust emissions from different technology transit buses. For this study, two CNG (compressed natural gas) buses equipped with lean burn combustion and OCs (oxidation catalysts), and one stoichiometric CNG bus equipped with a TWC (three-way catalyst) and EGR (exhaust gas recirculation) were tested on a chassis dynamometer over the CBD (Central Business District) cycle on six different gas blends each. The gases represented a range of compositions from gases with high levels of methane and correspondingly lower energy contents/WN (Wobbe number) to gases with higher levels of heavier hydrocarbons and correspondingly higher energy contents/WN. For the lean burn buses, gases with low methane contents exhibited higher NO_x (nitrogen oxides) (19%–53%) and NMHC (non-methane hydrocarbon) (39%–102%) emissions, but lower emissions of THC (total hydrocarbon) (9%–24%), CH_4 (methane) (23%–33%), and formaldehyde emissions (14%–45%). The stoichiometric engine bus with a TWC showed significantly reduced NO_x and THC emissions compared to the lean burn buses, but did show higher levels of CO (carbon monoxide) and NH_3 (ammonia). PM (particulate matter) mass emissions did not show any fuel effects, while PN (particle number) emissions exhibited some reductions for the higher WN gases.

1. Introduction

In an effort to improve air quality, reduce greenhouse gas emissions, and reduce dependency on fossil fuels, regulatory agencies have implemented a variety of legislative measures to increase the use of alternative fuels. NG (natural gas) is one of the most prominent alternative fuels with significantly larger reserves compared to crude oil, and also the potential for air quality benefits in vehicles [1]. In recent years, there have been substantial changes in the NG market due to the rapid development of horizontal drilling and hydraulic fracturing. Such advanced techniques have unlocked vast reserves of oil and gas trapped underneath sedimentary rocks or shale. The U.S. (United State) EIA (Energy Information Administration) anticipates U.S. NG production to continue to expand into the future, growing from levels of 23.5 quadrillion Btu in 2011 to a projected 33.9 quadrillion Btu in 2040, representing a sizable 44% increase [2]. Shale gas production, which already accounted for 23% of total U.S. natural gas production in 2010, is expected to be the primary driver of this expansion, with shale gas production going from 6.8 tcf (trillion cubic feet) in 2011 to 13.6 tcf in 2035 [3].

The quality of natural gas depends on both its source and the degree to which it is processed. Natural gas can be produced from oil fields (termed associated gas) or from gas fields (termed non-associated gas). Associated gas is typically higher in heavier hydrocarbons, which gives the gas a higher WN (Wobbe Number) and a lower MN (Methane Number). Associated gas is often processed using techniques such as refrigeration, lean oil absorption, and cryogenic extraction to recover valuable NGLs (natural gas liquids), such as ethane, propane, butanes, pentanes and hexanes plus, for other uses [4,5]. Traditional North American gas from Texas, for example, is often processed to recover feedstock for chemical plants. This process lowers the WN and increases the MN of the resulting NG

* Corresponding author. Tel.: +1 951 781 5794; fax: +1 951 781 579.
E-mail addresses: mhajb001@ucr.edu (M. Hajbabaei), durbin@cert.ucr.edu (T.D. Durbin).

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stream. As NG production continues to increase, it is likely that a wider range of NG compositions could be introduced into the marketplace, either due to different sources of production or perhaps a reduced emphasis on recovering NGLs from NG if the economics for these secondary products change. This could lead to NG with higher WNs and lower MNs being fed into the pipeline, which would likewise result in a pipeline gas with a higher WN and lower MN.

A number of studies have compared the emissions of NGVs with diesel powered HD (heavy-duty) vehicles over a wide range of engine and aftertreatment configurations [6–11]. For the pre-2008 lean burn technologies, NG engines show reductions in PM (particulate matter) relative to diesel engine, and also slight reductions in CO_2 (carbon dioxide) emissions [6,7,9,10,12,13]. Emissions comparisons between NG and diesel for CO (carbon monoxide) and HCs (hydrocarbons) showed different trends over a range of studies depending on the specific technology tested, the condition of the vehicles, if the HCs were measured as THC (total hydrocarbons) or NMHC (non-methane hydrocarbon), and other factors [6,7,9,10,14,15]. The lean burn NG engines produced prior to the introduction of the Cummins Westport ISL-G could achieve reductions in NO_x (nitrogen oxides) emissions relative to diesel engines without aftertreatment, but their NO_x emissions were sometimes more variable in practice [6,10,16]. The latest standards for NO_x emissions have necessitated that use of SCR (selective catalytic reduction) systems on diesel engines. This led to the implementation of stoichiometric combustion engines and TWC (three-way catalyst) aftertreatment systems as the primary technology being used with NG engines, as employed with the Cummins Westport ISL-G [17–19]. The low levels of carbon–carbon bonds in NG and the absence of aromatics compared to diesel fuel also reduces soot formation in NGVs [20]. NGVs have generally higher CH_4 (methane) emissions, which is a greenhouse gas. CH_4 is less of a concern in the photochemical smog cycle, however, since it is less reactive compared to other hydrocarbons.

With the growing expansion of natural gas production and the potential change for natural gas fuel composition from source to source, it is crucial to investigate the effect of natural gas fuel composition on the performance and operation of natural gas heavy-duty vehicles, especially since limited comprehensive studies have been conducted in this area. In an earlier chassis dynamometer study, Graboski et al. [21] tested five different NG compositions in a bus equipped with a heavy-duty Cummins B5.9G lean-burn engine at high altitude. They found that THC emissions increased with increasing levels of inert gases and NO_x emissions increased with increasing fuel heating value, while CO and PM emissions were unaffected by fuel gas composition due to their low values. In a recent study, Karavalakis et al. [22] tested a refuse hauler with a Cummins Westport lean-burn spark ignited engine and an OC over the William H. Martin Refuse Truck Cycle on seven different gases. They found that NO_x and NMHC emissions increased for gases with higher levels of heavier hydrocarbons/higher WN, while THC, and CH_4 emissions increased for gases with higher levels of CH_4. They also reported reductions in PM mass for gases with more heavier hydrocarbons and reductions in particle number emissions for some gases with more heavier hydrocarbons, but not for others. Feist et al. [8,23] also investigated the impact of several NG fuels for three different 1998–2006 HD lean burn NG engines with OCs (oxidation catalysts) and one 2008 HD stoichiometric NG engine with a TWC on an engine dynamometer. They observed that all lean-burn engines showed increased NO_x and HC emissions with higher WN fuels, while the stoichiometric engine showed no clear trends for NO_x or HC emissions with varying NG composition. They also found that PM and CO emissions did not show strong trends with MN or WN, and that low WN fuels resulted in increased fuel consumption. A number of other studies have also investigated the impact of NG composition on emissions, although most of these studies have focused on other applications, such as light-duty vehicles and engines, generators, and compressors [24–30].

Limited information is available on the unregulated emissions from NGVs, including gaseous toxic pollutants and PAHs (polycyclic aromatic hydrocarbons). Kado et al. [13] found that the carbonyl emissions from CNG (compressed natural gas) buses were primarily formaldehyde. Formaldehyde emissions from these buses were much greater than those of diesel buses fitted with OCs, and CRTs (continuously regenerating traps). Ayala et al. [10] also found that formaldehyde emissions were reduced by OCs on CNG buses by over 95% over the CBD (Central Business District) cycle. Okamoto et al. [31] and Kado et al. [13] performed mutagenic tests on the exhaust from transit buses operating on CNG. They both reported lower mutagenic activity for CNG buses equipped with OCs, compared to buses without OCs. Kado et al. [13] also found that mutagenic activity using the TA98NR test strain decreased, indicating the possible presence of nitro-PAH in the PM emissions. Turrio-Baldassarri et al. [32] showed that a spark ignition heavy-duty urban bus NG engine with a TWC produced 20 times lower formaldehyde, more than 30 times lower PM emissions, and 50 times lower PAH emissions, compared to a diesel engine without aftertreatment.

PN (particle number) emissions and particle size distributions are also of importance for NGVs. Particle emissions from NGVs are smaller in size than those from diesel engines. This can be an issue since nano-sized particles have adverse human health effects. They are carcinogenic and can be transported easily to human organs, such as the lungs and brain [7,12,13]. Jayaratne et al. [33] tested particle emissions from four CNG and four diesel buses. They found that PN emissions were significantly lower for the CNG buses. They also reported that all the particles emitted from the CNG buses were in the nanoparticle size range and composed mostly of ash from lubricating oil. Similar results were reported by Holmen and Ayala [34] when they monitored the PN emissions from buses equipped with diesel engines with an OEM (Original Equipment Manufacturer) catalyzed muffler and with a DPF (Diesel Particulate Filter), and with a CNG engine without aftertreatment. They found that PN emissions in the accumulation mode were 10–100 times lower for the CNG engine compared to the diesel engine with the catalyzed muffler. Lanni et al. [35] tested two diesel buses with DPFs and three CNG buses without aftertreatment over the CBD and NYB (New York Bus) cycles and found the particle size distributions ranged from 10 to 30 nm, with an apparent shift towards smaller diameters for the CNG buses.

The objective of the present study is to evaluate the impact of NG composition on the exhaust emissions of heavy-duty NG vehicles [36]. This study focuses on transit buses, a category of heavy-duty vehicles that warrants attention for controlling NO_x and PM emissions due to the fact that they operate primarily in populated urban and suburban settings. For this study, three NG transit buses were tested on a range of six different test gases over the CBD cycle. In addition to the regulated emissions and fuel economy, NH_3 (ammonia), carbonyl compounds, and PN emissions were also evaluated. Information from this study on the impact of changing NG composition on emissions can be used for regulatory development, to ensure new NG compositions do not have an adverse impact on air quality, and to evaluate the viability of using a broader mixture of NG blends in transportation applications.

2. Experimental procedures

2.1. Test fuels

For this study six NG blends were used. Gases H1 and H2 are representative of historical Texas and Rocky Mountain Pipeline

Table 1
Main properties of the fuel gas blends.

Gas#	Description	Methane	Ethane	Propane	I-butane	N_2	CO_2	MN	Wobbe number	HHV	H/C ratio	MON
H1	Texas Pipeline	96	1.8	0.4	0.15	0.7	0.95	99	1339	1021	3.94	135.1
H2	Rocky Mountain Pipeline	94.5	3.5	0.6	0.3	0.35	0.75	95	1361	1046	3.89	131.2
LM3	Peruvian LNG	88.3	10.5	0	0	1.2	0	84	1385	1083	3.81	125.7
LM4	Middle East, LNG-Untreated	89.3	6.8	2.6	1.3	0	0	80	1428	1136	3.73	121
LM5	Associated High Ethane	83.65	10.75	2.7	0.2	2.7	0	75.3	1385	1115	3.71	119.9
LM6	Associated High Propane	87.2	4.5	4.4	1.2	2.7	0	75.1	1385	1116	3.70	119.3

Gas composition is reported on a Mole percent basis; MN = methane number determined via California Air Recourses Board (CARB) calculations [37]; Wobbe number = HHV/square root of the specific gravity of gas blends with respect to air; HHV = higher heating value; H/C = ratio of hydrogen to carbon atoms in the hydrocarbon portion of the gas blend; MON = motor octane number derived via mathematical relation, which was developed in Ref. [37].

Gases and serve as the baseline fuels. The four other test gases all have lower methane contents and MNs, and corresponding higher WNs and HHVs. These gases are labeled 'LM' and are denoted as low methane gases throughout this paper. Gas LM3 is representative of Peruvian LNG (Liquefied Natural Gas) that has been modified to meet a WN of 1385 and a MN of 75. Gas LM4 is representative of Untreated Middle East LNG with a high WN (above 1400). Gas LM5 is a high ethane gas with a WN of 1385 and a MN of 75. Gas LM6 is a high propane, high butane gas with a WN of 1385 and a MN of 75. Gases LM5 and LM6 are hypothetical gases designed to investigate whether two fuels with the same WN and MN, but different compositions, would produce different exhaust emissions. Gases with higher propane and butane than pipeline gas are found in the South Central Coast region oil and gas fields, while gases with high ethane are found in San Joaquin Valley oil and gas fields. Gases LM5 and LM6 are both at the extremes for WN and MN, so the typical local gas in the pipeline in these areas will have lower WNs and higher MNs. A wide range of scenarios were examined in this study to evaluate the viability of permitting the use of a broader mixture of NG blends in transportation applications. The test fuels properties are presented in Table 1.

2.2. Test vehicles

Three buses were used in this study, including a bus equipped with a 2009 stoichiometric spark ignited Cummins Westport ISL-G8.9 L engine with a TWC and a cooled EGR (exhaust gas recirculation) system, a bus equipped with a 2004 John Deere 8.1L 6081H lean burn engine, and a bus equipped with a 2003 8.3L Cummins Westport C-Gas Plus lean burn engine. Both the 2004 John Deere and 2003 C-Gas Plus lean burn vehicles were fitted with OCs for controlling THC and CO emissions. These buses were selected to represent two older technology lean burn engines with a high population in the in-use fleet, and the newest technology engine available at the time of the testing. It should be noted that the John Deere bus was tested on two separate occasions, once before and again after a mechanical issue was discovered. Specifically, the bus lost compression in one of its combustion cylinders during the initial round of testing. The retesting on the repaired vehicle was done approximately one year after the initial testing. The main technical specifications of the engines are listed in Table 2.

2.3. Test cycles and measurement protocol

Testing was performed over a specially developed CBD cycle. The driving pattern for the CBD cycle was developed as a general representation of transit bus operation in a downtown business district [6]. The cycle used in this study consisted of a single CBD cycle as a warm-up, followed by two iterations (i.e., a double) CBD cycle to provide a sufficient particle sample for analysis. The CBD cycle is characterized by an average speed of 20.23 km/h (13 mph), a maximum speed of 32.18 km/h (20 mph), an average acceleration of 0.89 m/s^2, and a maximum acceleration of 1.79 m/s^2. The driving distance for a single CBD cycle is 3.22 km, or 9.66 km for the full cycle, including the warm-up. A speed-time trace profile for the extended CBD can be found elsewhere [6]. Six tests were run on each vehicle/fuel combination for all vehicles, with a limited number of exceptions. The test matrix was randomized to allow some measure of the experimental reproducibility. Note that LM4 was not tested on the Cummins Westport C-Gas Plus bus.

Table 2
Technical specifications of the test engines.

Manufacturer	Cummins Westport	John Deere	Cummins Westport
Engine model	ISL-G	6081HF	C-Gas Plus
Model year	2009	2004	2003
Vehicle type	Bus	Bus	Bus
Engine family	9CEXH054 LBD	4JDXH08.1066	3CEXH0505CBK
Engine type	Stoichiometric Spark-ignited Turbocharged, EGR	Lean burn Spark-ignited Turbocharged	Lean burn Spark-ignited Turbocharged
Horsepower	280 HP	280 HP	280 HP
Number of cylinders	6	6	6
Bore and stroke	114 mm × 145 mm	116 mm × 129 mm	114 mm × 135 mm
Displacement	8.9 L	8.1 L	8.3 L
Compression ratio	12:1	16.5:1	10:1
Peak torque	900 ft-lbs. @ 1300 rpm	900 ft-lbs. @ 1500 rpm	850 ft-lbs. @ 1400 rpm
Aftertreatment	TWC	OC	OC
Certification level (g/bhp-hr)	NMHC: 0.13 NO_x: 0.10 CO: 1.2 PM: 0.009	NMHC + NO_x: 1.5 CO: 0.1 PM: 0.01	NMHC + NO_x: 1.7 CO: 2.0 PM: 0.01

2.4. Emission testing and analysis

The chassis dynamometer testing was conducted at the UCR (University of California, Riverside) CE-CERT's (Center for Environmental Research and Technology's) heavy-duty chassis dynamometer facility. The emissions measurements were obtained using CE-CERT's MEL (Mobile Emissions Laboratory) with a full dilution system [38,39]. For all tests, standard emissions measurements of THC, NMHC, CH_4, CO, NO_x, CO_2, and PM mass, were performed according to CFR (Code of Federal Regulations) Title 40 (40 CFR) 1065 requirements.

Total PN counts, particle size distributions, and PM mass were measured through a secondary dilution tunnel. Total PM mass determinations were collected using 47 mm Teflon® filters and measured with a 40 CFR Part 1065-compliant microbalance in a temperature and humidity controlled clean chamber. PN was measured using a TSI 3776 ultrafine-CPC (Condensation Particle Counter). This is a butanol-based CPC that has the ability to count particles down to 2.5 nm. This instrument can sample particles of about 300,000 per second, making the ultrafine CPC ideal for an accurate total PN measurement. Particle size distributions were measured using two different instruments due to the availability of different instruments at different times over the course of testing. A nano-SMPS (nano scanning mobility particle sizer) with 3085 TSI DMA (differential mobility analyzer) column was used for the 2009 Cummins Westport ISL-G8.9 bus and the John Deere bus tests. The size range of the nano-SMPS was 4–70 nm with a scan time of 118 s. For the C-Gas Plus bus testing, an EEPS (engine exhaust particle sizer) was used for both particle size distributions and PN measurement. The EEPS had a faster scan time of 1 s and a wider size range from 6 to 423 nm compared to the nano-SMPS. The faster scan time allows the EEPS to more accurately capture the size distributions under transient operating conditions. Measurements of NH_3 were obtained on a real-time basis using a Unisearch Associates Inc. LasIR S Series TDL (tunable diode laser) near infrared absorption spectrometer. The TDL system was used because it provides significant advantages for the measurement of exhaust NH_3 in sensitivity, response time, and the ability to measure in situ in raw exhaust [40].

Testing and analysis of carbonyl compounds were performed in accordance with protocols developed as part of the Auto/Oil Air Quality Improvement Research Program [41]. Samples for carbonyl analysis were collected through a heated line onto 2,4-DNPH (dinitrophenylhydrazine) coated silica cartridges (Waters Corp., Milford, MA). The samples were then stored in the refrigerator until subsequent analysis. Sampled cartridges were extracted using 5 mL of acetonitrile and analyzed with an Agilent 1200 series HPLC (high performance liquid chromatograph) equipped with a diode array detector using the HPLC sample injection and operating conditions as specified in the SAE [930142HP] protocol. Three carbonyl samples were typically collected for each vehicle/fuel combination.

3. Results and discussion

The figures for each pollutant show the results for each vehicle/fuel combination based on the average of the tests conducted on that particular test combination. The error bars on the figures are the standard deviation over all tests for each 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 $p \leq 0.05$, or marginally statistically significant for $0.05 < p \leq 0.1$ in this analysis. The John Deere results are shown separately for the initial and post-repair testing.

3.1. NO_x emissions

NO_x emission results are shown in Fig. 1. NO_x emission levels for the stoichiometric Cummins Westport ISL-G8.9 bus fitted with a TWC were significantly lower than those of the lean-burn John Deere and C-Gas Plus buses with OCs, noting that the emissions for the Cummins Westport ISL-G8.9 bus are multiplied by 50 in the figure. Similar results, showing that stoichiometric engines equipped with cooled EGR and TWC significantly reduce NO_x emissions, have been reported by other authors [14,15]. The effectiveness of the TWC in reducing NO_x emissions is a key to achieving the NO_x reductions seen for the ISL-G bus [17], coupled with the stoichiometric combustion needed to provide the conditions needed for the TWC to work optimally. In contrast, the OC does not provide catalytic reduction of NO_x. For the stoichiometric ISL-G8.9 Cummins bus, EGR also decreases NO_x emissions by introducing inert exhaust gas back into the combustion cylinder, which reduces the combustion temperature [19].

For the John Deere and C-Gas Plus buses, the NO_x emissions generally showed trends of higher NO_x emissions for the low methane gases. The C-Gas Plus bus showed statistically significant increases of 38%, 53%, and 32%, respectively, for LM3, LM5, and LM6 compared to H1. For the post-repair John Deere results, these increases were statistically significant for LM6 compared to H1 (+49%), while for the initial John Deere testing a statistically significant increase was found for LM4 fuel compared to H1 (+18.8%). The stoichiometric Cummins Westport ISL-G8.9 did not show significant differences between fuels for NO_x emissions.

The increases in NO_x emissions with LM3, LM4, LM5, and LM6 gases for the lean burn engines could be attributed to the presence of higher molecular-weight hydrocarbons in these gases, which also leads to corresponding increases in WN. The addition of higher hydrocarbons (ethane and propane) can increase the adiabatic flame speed. As flame speed increases at constant ignition timing, peak pressure occurs earlier, at smaller cylinder volumes, and thus higher temperatures. Peak combustion temperatures are therefore higher due to the advanced location of the peak pressure and higher adiabatic flame temperature [8,23], which would result in higher NO_x emissions, as NO_x is generated predominantly through the strongly temperature-dependent thermal NO mechanism [30,42]. Previous studies have also shown that lean-burn engines run richer as MN is decreased [24]. 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.

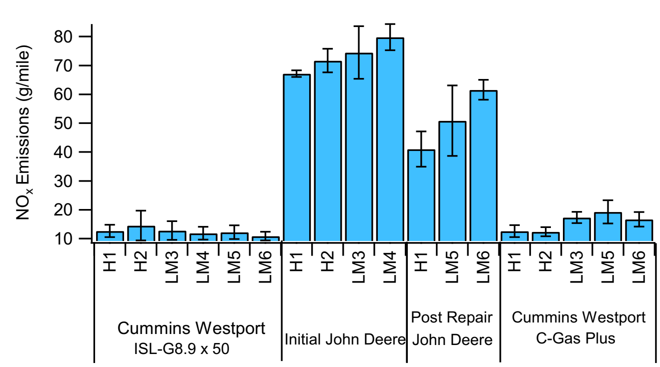

Fig. 1. Average NO_x emissions from NG buses over the CBD.

3.2. THC, NMHC, and CH_4 emissions

THC emissions results are shown in Fig. 2a. THC emissions were significantly lower for the Cummins Westport ISL-G8.9 bus than the older John Deere and C-Gas Plus buses, noting that the emissions for the Cummins Westport ISL-G8.9 bus are multiplied by 10 in the figure. This can be attributed to the differences in the engine and aftertreatment technologies, since the older engines are all lean-burn engines fitted with OCs designed to meet an earlier certification standard, and the ISL-G is a stoichiometric engine with a TWC that is designed to meet a more recent and more stringent certification standard. Most of THC emissions reductions are due to the greater conversion efficiency of the TWC, which is larger in size and has higher loadings of precious metals compared to the OCs [43]. Also, the conversion efficiency of CH_4, the predominate component of THC, can also be increased with different precious metals and under stoichiometric conditions [44]. Similar reductions have been seen in other studies [8,15,17]. Einewall et al. [17] found that catalyst efficiency was considerably higher for stoichiometric operation with a TWC compared to lean burn operation with an OC. Lean burn engines are typically characterized by cooler and slower combustion, which can lead to higher exhaust

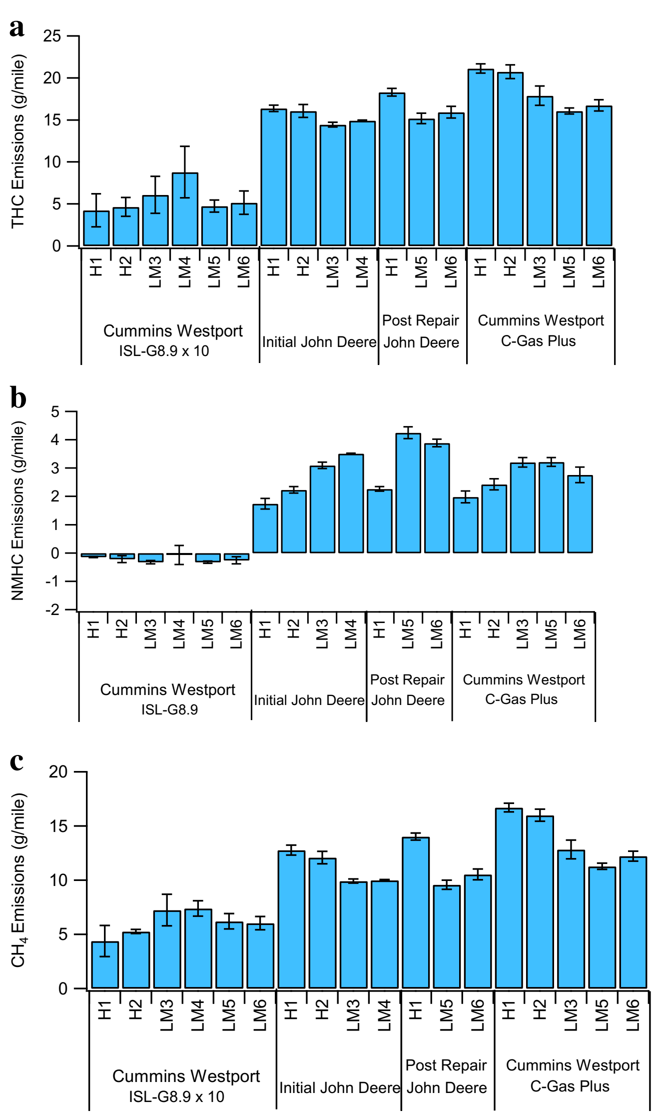

Fig. 2. (a–c). Average THC, NMHC, and CH_4 emissions from NG buses over the CBD.

temperatures. Wit et al. [45] observed higher exhaust temperature in stoichiometric engines which increased the temperature of TWC and improved CH_4 conversion efficiency compared to the learn burn engines. The cooler combustion temperatures for the lean burn engines and operation near the lean burn limit for HC formation, could also lead to higher levels of engine-out THC compared to stoichiometric engines.

The John Deere and C-Gas Plus buses showed trends of higher THC emissions for the gases with higher methane contents. For the C-Gas Plus bus, statistically significant reductions in THC emissions of 15%, 24%, and 21%, respectively, for LM3, LM5, and LM6 were found compared to H1. For the post-repair John Deere bus testing, LM5 and LM6 showed statistically significant reductions of 16.9% and 13.3%, respectively, in THC emissions compared to H1. For the initial testing on the John Deere bus, LM3 and LM4 showed statistically significant reductions of 11.8% and 8.8%, respectively, in THC emissions compared to H1. For the Cummins Westport ISL-G bus, THC emissions were very low, and did not show strong fuel trends. Although LM4 showed a slight increase in THC emissions compared to the baseline H1, the higher emission levels for LM4 are still on same the order as the background levels of the system, and as such appear to be simply an artifact of measuring at such low levels.

This trend of higher THC emissions for the gases with higher methane contents for the lean burn engines is consistent with results previously reported by other authors [6,8,22]. This is probably due to the fact that the THC emissions were predominately methane with lower levels of heavier hydrocarbons. CH_4 is also less reactive than higher hydrocarbons and a considerably more stable molecule, so it is more likely to go through the combustion process unburned and more difficult to oxidize with the catalyst [46]. The reductions in THC emissions for the low methane gases could also be due to more complete oxidation of the fuel as the adiabatic flame speeds and combustion temperatures increased, as discussed under the NO_x emissions section. The higher combustion temperatures could also lead to higher exhaust temperatures than the baseline gases, which could also result in higher conversion rates with the OC. A NO_x/THC tradeoff, possibly caused by changes in peak flame temperature or speed, was observed. THC emissions decreased with low MN fuels, while NO_x emissions increased with low MN and high WN fuels. These phenomena are in agreement with the results previously reported by Graboski et al. [21] and Karavalakis et al. [22].

All the NG buses emitted substantially lower levels of NMHC emissions compared to THC emissions, as shown in Fig. 2b, with the NMHC emissions for the stoichiometric Cummins Westport bus being at the background levels. This is consistent with expectations and indicates that the THC emissions from these vehicles are predominantly methane with little NMHC emissions. The very low NMHC emissions for the stoichiometric engine with a TWC are in agreement with other studies showing very low NMHC emissions for such engines [14,15,47]. The significantly lower levels of NMHC emissions from the stoichiometric bus engine were predominately due to the higher conversion efficiency for the TWC compared to the OC.

The lean burn buses all showed trends of higher NMHC emissions for the gases containing higher levels of NMHCs (i.e., ethane, propane, and butane, as shown in Table 1). Previous studies have also shown that NMHC emissions increased with decreasing methane number of the fuel gases [8,22]. THC emissions from natural gas engines are predominately unburned fuel, therefore, the non-methane hydrocarbon fraction of THC exhaust emissions typically trends with the percentage of NMHC in the test fuel. The C-Gas Plus bus showed statistically significant increases in NMHC emissions for H2, LM3, LM5, and LM6 of 22%, 62%, 62%, and 39%,

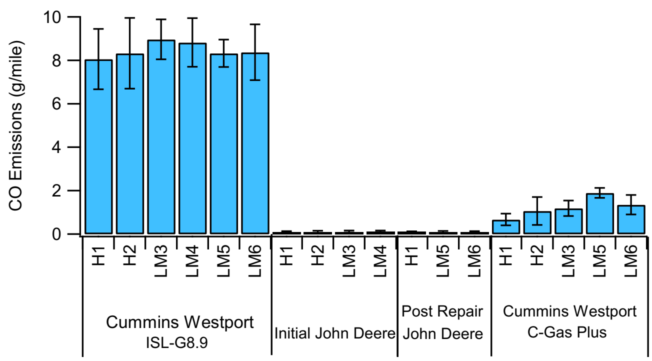

Fig. 3. Average CO emissions from NG buses over the CBD.

respectively, compared to H1. For the post-repair John Deere testing, LM5 and LM6 had statistically significant increases in NMHC emissions of 88% and 71%, respectively, compared to the H1. For the initial John Deere bus testing, the LM3 and LM4 gases showed statistically significant NMHC emissions increases of 78% and 102%, respectively, compared to H1, and of 39% and 57%, respectively, compared to H2.

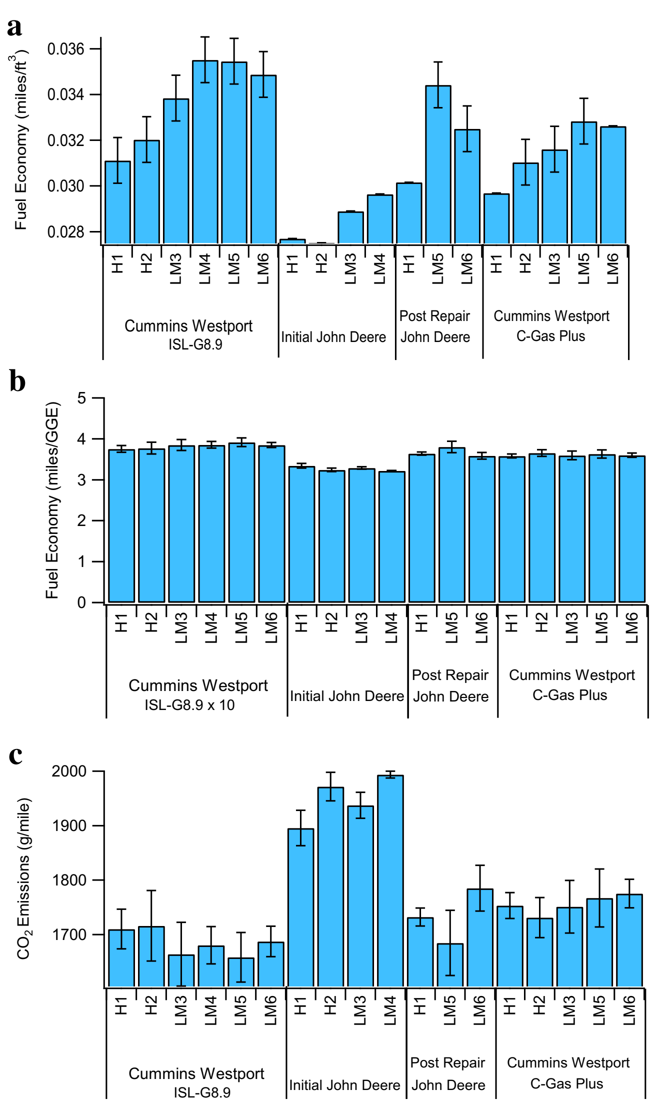

Fig. 4. (a–c). Average volumetric (a) and carbon balanced (b) fuel economy, and CO_2 emissions from NG buses over the CBD.

Methane is the major hydrocarbon in NG. It is non-reactive and does not participate in photochemical smog generating reactions, and is unregulated in the U.S. Fig. 2c shows the CH_4 emissions over the CBD cycle. The results showed that CH_4 emissions for the stoichiometric Cummins Westport ISL-G bus were about 95% lower than for the lean burn John Deere and C-Gas Plus buses, noting that the CH_4 emissions for the ISL-G are multiplied by 10 in the figure. The lower CH_4 emissions for the stoichiometric engine bus with the TWC, were primarily due to the larger size and higher precious metal loadings for the TWC. The different precious metals and stoichiometric combustion for the TWC can also promote the CH_4 conversion, resulting in lower CH_4 emissions [44].

The lean burn buses all showed a trend of higher CH_4 emissions for gases with higher methane contents, including H1 and H2. The C-Gas Plus bus showed the highest CH_4 emissions for H1 and H2, with reductions in CH_4 emissions of 4.3%, 23%, 33%, and 27%, respectively, for H2, LM3, LM5, and LM6 compared to H1, with most of the reductions being statistically significant. For the post-repair John Deere bus testing, H1 showed the highest CH_4 emissions, with statistically significant reductions in CH_4 emissions of 32% and 25%, respectively, for LM5 and LM6 compared to H1. For the initial John Deere test, H1 and H2 produced higher CH_4 emissions than those of LM3 and LM4. The stoichiometric Cummins Westport ISL-G showed slightly higher CH_4 emissions for gases LM3 and LM4, but similar to THC, the differences in CH_4 between gases are comparable to the background levels of the system, and hence, are probably an artifact of measuring at such low levels rather than real fuel effects.

3.3. *CO emissions*

CO emissions are shown in Fig. 3. It is evident that CO emissions for the stoichiometric Cummins Westport ISL-G8.9 vehicle were significantly higher than those emitted for the lean burn John Deere bus and for the lean burn C-Gas Plus bus. This can be attributed to the richer operating conditions of the stoichiometric combustion compared to lean burn combustion. Thus, less oxygen is available to oxidize CO to CO_2 during combustion or over the catalyst for the stoichiometric engine compared to the lean burn engine. This observation is consistent with the results of previous studies showing higher CO emissions for the stoichiometric Cummins Westport ISL-G engine compared to lean burn engines [15,47]. Although higher CO emissions were seen for the stoichiometric engine, the emissions are still relatively low compared to the certification limits. If a conversion factor of 4 bhp-hr/mile [15] is applied, the CO emissions levels are on the order of 2 g/bhp-hr, which is well below the certification standard of 15.5 g/bhp-hr [48]. Both initial and post repair John Deere testing showed very low CO emissions. For the Cummins Westport ISL-G and John Deere buses, no statistically significant differences in CO emissions between fuels were found. The C-Gas Plus bus showed some increases in CO emissions of 78%, 185% and 103%, respectively, for the low methane LM3, LM5 and LM6 gases compared to H1 that were statistically significant. Higher MN fuels also have higher octane ratings, which could contribute to more efficient combustion, thereby reducing CO emissions. The CO emissions for H2 were comparable to those of LM3 and LM6, however.

3.4. *Fuel economy and CO_2 emissions*

Fuel economy was determined using the EPA (Environmental Protection Agency) carbon balance method. Fuel economy is plotted on a volumetric basis in Fig. 4a. This is the most important metric for the NG consumer since fuel is purchased on a volumetric basis. For all the buses, the low methane gases with the higher

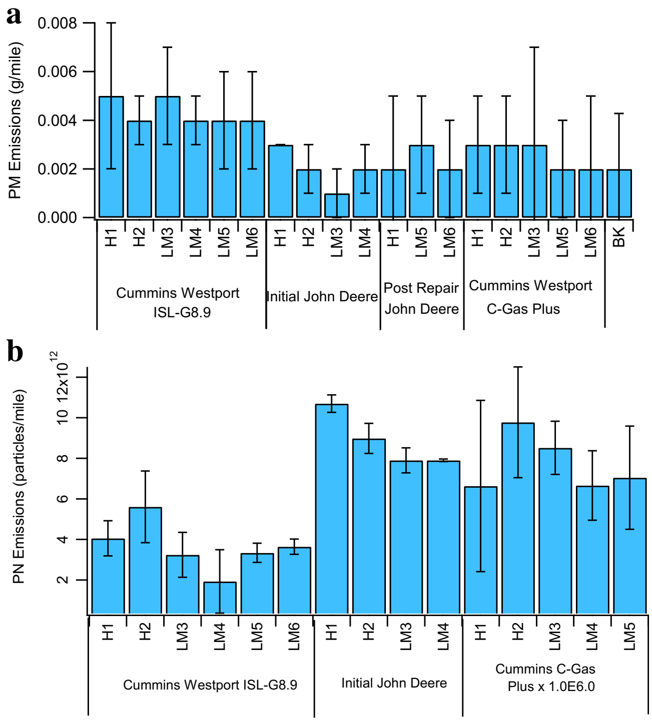

Fig. 5. (a–b). Average PM mass (a) and particle number (b) emissions from NG buses over the CBD.

heating values, i.e., LM3, LM4, LM5, and LM6, showed slightly higher fuel economy on a volumetric basis compared to H1 and H2. Fuel economy can also be examined on an energy equivalent basis, as shown in Fig. 4b. On this basis, the energy differences between the fuels are normalized so that the differences in fuels are more related to efficiency differences. Overall, the three buses showed comparable fuel economy results between fuels on an energy equivalent basis. The C-Gas Plus bus did not show any fuel effects, with the exception of LM5 and LM6 showing a marginally statistically significant 1.7–2.4% increase compared to H1. The energy equivalent fuel economy differences for the post-repair John Deere were only statistically significant for LM5, but were not statistically significant for LM6. Interestingly, the stoichiometric Cummins Westport ISL-G bus fuel economy results generally showed a trend of higher energy equivalent fuel economy for LM3, LM4, LM5, and LM6, which are the low methane gases with higher energy contents. The initial testing results for energy equivalent fuel economy on the John Deere, on the other hand, showed decreases in fuel economy for the low methane gases with higher energy contents (LM3 and LM4) which was only statistically significant for LM4, but this could be related to the mechanical failure.

CO_2 emissions from the three buses were comparable, as shown in Fig. 4c. The initial testing on the John Deere bus showed slightly higher CO_2 emissions, which could be related to its mechanical issues. The Cummins Westport ISL-G8.9, post-repair John Deere, and C-Gas Plus buses did not show strong trends in CO_2 emissions between the fuels. The initial testing of the John Deere bus showed slight, but statistically significant, increases in CO_2 emissions for H2 and LM4 compared to H1 and LM3. These differences could be related to the mechanical issue, however.

3.5. PM mass, particle number and particle size distributions

The results presented in Fig. 5a, indicated that total PM mass emissions were low for all three buses on an absolute level, and are

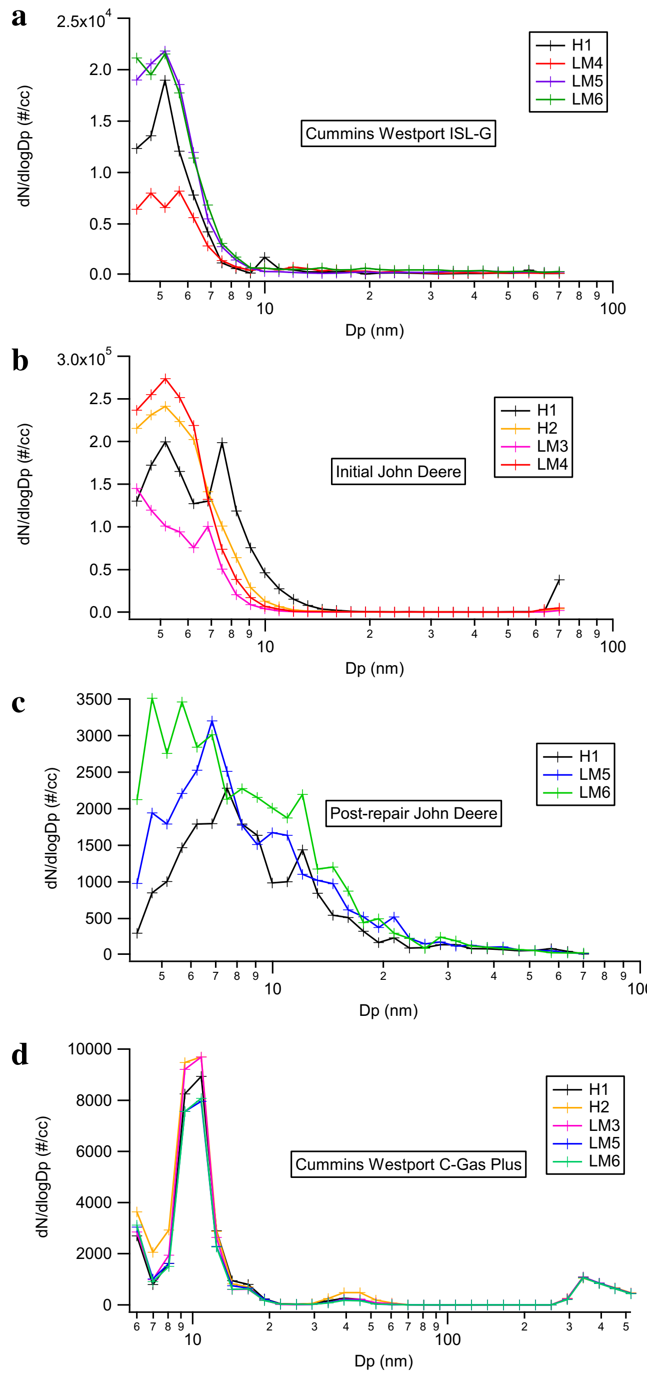

Fig. 6. (a–d). Average particle size distributions from NG buses over the CBD.

around the tunnel background levels. Although some differences were seen between fuels, these differences were all within the range of the tunnel background levels. The very low levels of PM mass emissions can be attributed to the fact that natural gas is primarily comprised of CH_4, which is the lowest molecular weight HC and has a simpler structure compared to diesel or gasoline fuels [49]. NG has a reduced tendency to form localized areas of rich combustion and generates unburned and partially oxidized hydrocarbons with lower molecular sizes in the exhaust, resulting in very low PM mass emission levels. Thus, the main source of PM in natural gas engines is considered to be the entry of engine lubricating oil into the combustion chamber [49]. It is worth noting that the stoichiometric bus produced somewhat higher PM emissions than the lean burn buses. This finding is not consistent with the results reported by Yoon et al. [15], but is in agreement with the results from Feist et al. [8,23] and Nylund et al. [47]. Under the present test conditions, it is possible that the OC was also more effective in removing and oxidizing volatile and semi-volatile hydrocarbons that are usually adsorbed onto carbon particles, than the TWC aftertreatment. The observed results also could indicate higher lubrication oil consumption for the bus with the stoichiometric fueling than the lean burn buses.

PN counts are presented in Fig. 5b for all cases except for the post-repair John Deere bus testing. PN counts were not measured for the post repair vehicle because of issues with the data acquisition system for the CPC. For the C-Gas Plus testing, the EEPS was used for the PN measurements. The C-Gas Plus PN measurements with EEPS showed somewhat greater variability than the other vehicles. For the initial John Deere bus testing, all test gases exhibited a statistically significant reduction in PN emissions compared to the baseline H1, with LM3 and LM4 showing the largest reductions. For the C-Gas Plus bus, H2 and LM3 showed PN emissions that were higher than H1, but these differences were not statistically significant. The greater variability for the C-Gas Plus bus PN measurements with the EEPS may also have made it more difficult to identify statistical trends, however. For the Cummins Westport ISL-G bus, some PN differences were seen between different fuels, but these differences were not statistically significant. The observed trends of lower PN emissions with the lower MN/higher WN and higher flame speed gases for the initial John Deere bus testing was somewhat unexpected since the presence of ethane and propane can enhance PM precursor formation. This trend could be due to higher temperature or more efficient combustion. Although the reduction of volatile and semi-volatile organics that are components of particles is not the primary function of the OC, the higher temperature combustion could lead to higher exhaust temperatures that could result in higher conversion efficiencies over the OC [45].

Measurements of the particle size distributions performed over the CBD cycle are displayed in Fig. 6(a–d). Particle size distributions for all buses/fuel combinations exhibited a consistent unimodal in nature nucleation mode, with peak particle diameters at around 4–10.8 nm. The findings of this study are in strong agreement with previous studies reporting that the majority of particles from CNG buses were in the nucleation mode [12,35,50]. The very low PM mass for the CNG blends indicates that the level of agglomeration to form larger carbonaceous particles and gas phase adsorption and condensation was relatively limited. Due to the absence of these larger particles, nucleation is the prevalent mode of particle formation. It should be noted that although the measurements with the nano-SMPS provide a good overall perspective of the particle sizes for the initial pre-repair John Deere bus testing and for the Cummins Westport ISL-G, the comparisons between fuels and the actual quantification of the PN concentrations are complicated by the relatively long scan time for the nano-SMPS instrument, which means this instrument samples only a small segment of its size range at any given time.

The C-Gas Plus bus produced unimodal distributions with a peak particle concentration at a diameter of 10.8 nm and with number concentrations ranging from 8000 to 9700 particles/cm^3. For the C-Gas Plus bus, the formation of a second nucleation mode at 30–50 nm size range was also observed. The Cummins Westport ISL-G bus produced unimodal distributions with a peak concentration at a diameter of 5.5 nm and with PN concentrations ranging from ~8000 to 22,000 particles/cm^3. For the initial John Deere testing, particle distributions exhibited a peak concentration at a diameter of 5.5 nm and PN concentrations ranged from ~90,000 to 270,000 particles/cm^3, while for the post repair John Deere bus particle sizes peaked at 8 nm and PN concentrations significantly decreased, and ranged from ~2200 to 2300 particles/cm^3. Under the present test conditions, consistent trends between fuels were not seen in the particle size distributions. Although there are differences in the fuels for different vehicles, they are not consistent between the different vehicles. For example, LM4 produced the highest level of nucleation particles for the initial John Deere bus, but the lowest for the Cummins Westport ISL-G bus. LM3 showed the highest emissions for the C-Gas Plus bus, but the lowest for the initial John Deere bus. Also, LM5 and LM6 showed the highest particle concentrations on the Cummins Westport ISL-G bus and the post-repair John Deere, but the lowest particle concentrations for the C-Gas Plus bus.

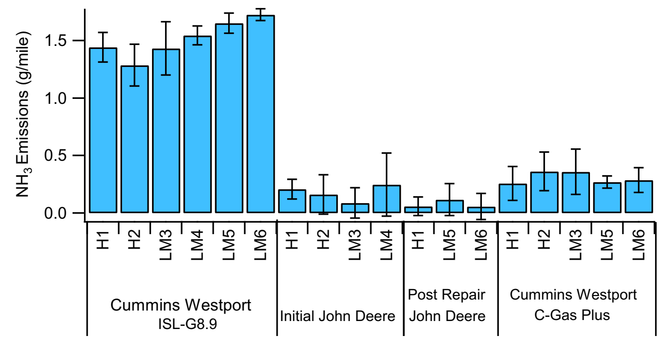

Fig. 7. Average NH_3 emissions from NG buses over the CBD.

3.6. NH_3 emissions

Fig. 7 shows the ammonia emissions for the three buses over the CBD cycle. The results revealed that the stoichiometric Cummins Westport ISL-G bus produced substantially higher NH_3 emissions compared to the lean burn John Deere and Cummins C-Gas Plus buses. It has been documented that NH_3 is a secondary pollutant formed during the NO_x reduction process over the TWC, with its formation to be dependent to the presence of both NO (nitrogen oxide) and H_2 (hydrogen) in the exhaust stream [18,51]. For TWC equipped stoichiometric natural gas 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 [18]. 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 [52,53]. The NH_3 emissions for the John Deere bus (for both initial and post-repair tests) were very low by comparison with the stoichiometric ISL-G bus. The NH_3 emissions for the C-Gas Plus bus were higher than those for the John Deere bus, but were still much lower than those for the stoichiometric ISL-G bus.

In general, no consistent fuel effects were observed for the buses, and most of the emissions differences compared to H1 were not statistically significant. A slight trend towards higher NH_3 emissions was seen for the stoichiometric fueling bus for some of the lower MN/higher WN/higher flame speed gases. Since the higher WN gases can produce higher exhaust temperatures and possibly slightly richer air-fuel ratios, the conditions for the formation of hydrogen as a precursor and NH_3 as reaction product could be enhanced for the higher WN gases.

3.7. Carbonyl emissions

The emission levels for formaldehyde and acetaldehyde are shown in Fig. 8(a–b). Formaldehyde and acetaldehyde emissions were the most prominent measured aldehydes in the tailpipe, with

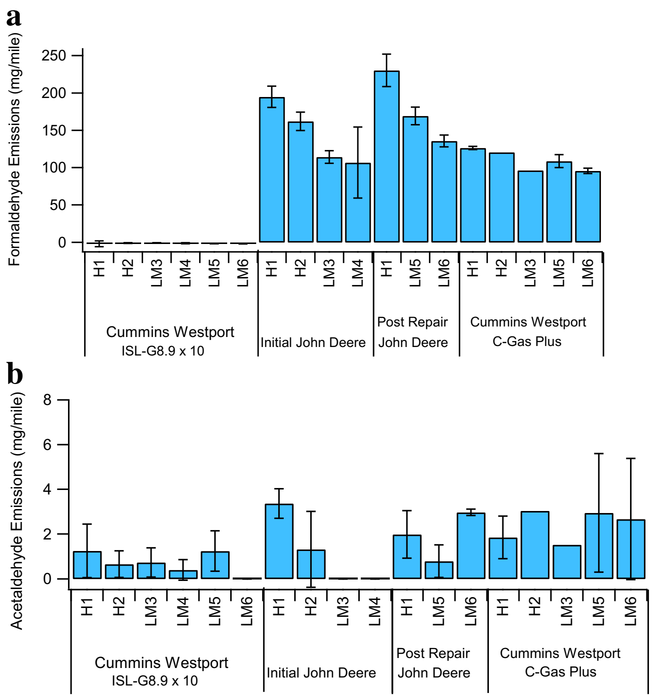

Fig. 8. (a–b). Average formaldehyde (a) and acetaldehyde (b) emissions from NG buses over the CBD.

formaldehyde being the dominant compound. Note that formaldehyde and acetaldehyde are the lowest molecular weight aldehydes, having one and two carbons, respectively. Our results are in agreement with previous studies showing that the most abundant aldehyde emissions from CNG vehicles come from the lowest molecular weight compounds [13,32,54,55]. The magnitudes of formaldehyde and acetaldehyde emissions were at the measurement limits for the stoichiometric Cummins Westport ISL-G bus, and did not show any fuel trends. It appeared that the TWC was effective in reducing both aldehydes to close to background levels. This result is consistent with previous studies documented that stoichiometric fueled NG vehicles fitted with TWC produce lower formaldehyde emissions than lean-burn NG vehicles [14,47].

For both the initial and post-repair John Deere bus tests, H1 and H2 showed the highest formaldehyde emissions compared to the other gases. For the post-repair John Deere testing, statistically significant reductions in formaldehyde emissions of 27% for LM5 and 41% for LM6 compared to H1 were found. For the initial John Deere testing, statistically significant reductions in formaldehyde of 16.9% for H2, 41% for LM3, and 45% for LM4 compared to H1 were found. For the John Deere bus, the formaldehyde results follow the same trends as the THC emissions, with gases with higher methane contents producing higher levels of formaldehyde. The same trend of higher formaldehyde emissions with the high methane gases was seen for the C-Gas Plus bus, although the trend was not as strong as for the John Deere. For the C-Gas Plus bus, H1 and H2 showed the highest formaldehyde emissions. Statistically significant reductions in formaldehyde emissions of 14% for LM5 and 24% for LM6 were found compared to H1 gas. For the acetaldehyde emissions, the buses did not show consistent fuel trends. However, for the initial John Deere bus testing, a statistically significant reduction of acetaldehyde emissions was seen for LM3 and LM4 compared to H1. H2 showed a marginally statistically significant reduction in acetaldehyde emissions compared to H1.

The higher formaldehyde emissions for the gases with higher methane contents are consistent with previous studies, since formaldehyde is an intermediate step in the oxidation of methane under high temperature conditions and across the catalyst [54]. The reductions in formaldehyde emissions for the low methane content gases may also be attributed to their higher adiabatic flame speeds, and ultimately to higher combustion temperature increases, which resulted in more complete oxidation of the fuel hydrocarbon fractions.

4. Conclusion

As the production of NG throughout the U.S. expands, there is potential for a wider range of natural gas compositions to be used in NGVs. It is important to evaluate whether changing compositions of NG will have adverse impacts on regional and global air quality. In this study, six blends of natural gas with different fuel compositions were tested. The gases represent a range of compositions from gases with high levels of methane and correspondingly lower energy contents and WNs to gases with higher levels of heavier hydrocarbons and correspondingly higher energy contents and WNs. Emissions testing was performed on three transit buses, a bus with a 2009 stoichiometric combustion, spark-ignited engine with cooled EGR and a TWC, and two buses with older 2002 and 2004 lean burn engines, fitted with OC over the CBD driving cycle.

The results showed that fuel composition influenced the formation of exhaust emissions from the older lean burn buses. Gases with low methane contents showed higher NO_x and NMHC emissions and improved fuel economy on a volumetric basis, but lower emissions of THC, CH_4, and formaldehyde emissions. Although trends were found between gases with higher vs. lower methane contents, other trends between gases were not as strong. For example, gases LM5 and LM6, which have varying contents of ethane and propane and butane, have similar emissions. The trends for the other emissions were not as consistent. The newest technology bus with the stoichiometric combustion engine and the TWC did not show any specific fuel effects.

The results show that NG fuel composition can have an impact on emissions for older technology heavy-duty vehicles even for gases within pipeline specifications, albeit at the extreme ranges of what might be found in the pipeline. This suggests that control of the NG specifications is still needed for older technology heavy-duty NGVs. It appears that newer technology heavy-duty natural gas engines can run on a wider range of NG fuels with varying composition without impacting emissions. Further study of the impact of NG composition for post-2007 engine is also planned for other applications, such as refuse trucks. Further studies also should be performed related to newer technology stoichiometric fueled NG engines and their associated NH_3 emissions to better understand the NH_3 formation mechanism and its possible contribution to secondary PM formation.

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