Ultrafine Particulate Emissions from Compressed Natural Gas and Clean Diesel Transit Buses

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Abstract

The New York State Department of Environmental Conservation (DEC) has joined with the New York City Metropolitan Transit Authority (MTA) and Environment Canada to evaluate emissions control technologies for transit buses. Particulate emissions were measured from buses powered by Detroit Diesel Series 50G Compressed Natural Gas (CNG) engines without diesel oxidation catalyst (DOC), Series 50 diesel engines with continuously regenerating diesel particulate filters (CRDPFs), and Series 50 diesel engines with DOC tested on a chassis dynamometer. Overall, the reduction in particulates was approximately equal for CNG and CRDPF buses, i.e., a 90% reduction from Series 50 diesel buses with DOC only, for all particle sizes. However, backfiring was observed for one of the CNG buses, characterized by sharp increases in ultrafine particle concentration, audible noises and changes in engine performance perceptible to the driver, and an increase in NOx emissions for the driving cycle. A retrofit campaign, which included the other two CNG buses in the study, was able to virtually eliminate this condition for on-road driving. While one of the retrofitted buses still exhibited small magnitude particulate increases (“micro-backfires”), these were detectable only by particulate instrumentation and did not significantly affect either the composite size distribution or the on-road performance. While particulate concentrations were relatively equivalent between CNG and CRDPF buses with the exception of backfiring, chemical analysis yielded significant other differences in the composition of their emissions indicating possible increased toxicity of CNG emissions. Specifically, levels of formaldehyde, benzene, toluene, ethylene, propylene and PAHs were all found to be elevated for the CNG buses compared to CRDPF buses.
## Experimental - Series 50G DDC Buses Tested

<table>
<thead>
<tr>
<th>NYCTA #</th>
<th>824</th>
<th>854</th>
<th>975</th>
<th>6019</th>
<th>6065</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Chassis</strong></td>
<td>New Flyer CLF 40</td>
<td>New Flyer CLF 40</td>
<td>New Flyer CLF 40</td>
<td>Orion V</td>
<td>Orion V</td>
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<tr>
<td><strong>Displacement</strong></td>
<td>8.5L</td>
<td>8.5L</td>
<td>8.5L</td>
<td>8.5L</td>
<td>8.5L</td>
</tr>
<tr>
<td><strong>Type</strong></td>
<td>4-Stroke</td>
<td>4-Stroke</td>
<td>4-Stroke</td>
<td>4-Stroke</td>
<td>4-Stroke</td>
</tr>
<tr>
<td><strong>Power (hp)</strong></td>
<td>275</td>
<td>275</td>
<td>275</td>
<td>275</td>
<td>275</td>
</tr>
<tr>
<td><strong>Configuration/No. Cylinders</strong></td>
<td>Inline 4 cylinder</td>
<td>Inline 4 cylinder</td>
<td>Inline 4 cylinder</td>
<td>Turbocharged Inline 4 cylinder</td>
<td>Turbocharged Inline 4 cylinder</td>
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<tr>
<td><strong>Catalytic After-treatment</strong></td>
<td>None</td>
<td>None</td>
<td>None</td>
<td>DOC/CRDPF</td>
<td>DOC/CRDPF</td>
</tr>
<tr>
<td><strong>Fuel</strong></td>
<td>CNG</td>
<td>CNG</td>
<td>CNG</td>
<td>30 ppm sulfur</td>
<td>30 ppm sulfur</td>
</tr>
<tr>
<td><strong>Test Cycles</strong></td>
<td>CBD, NYB</td>
<td>CBD, NYB</td>
<td>CBD, NYB</td>
<td>CBD, NYB</td>
<td>CBD, NYB</td>
</tr>
</tbody>
</table>

CBD – central business district driving cycle  
CNG – compressed natural gas  
CRDPF – continuously regenerating diesel particulate filter  
DOC – diesel oxidation catalyst  
NYB – New York bus driving cycle  
A minimum of 6 repetitions were performed for each driving cycle for each case.
# Experimental – Analytical Methods

<table>
<thead>
<tr>
<th>Analytes</th>
<th>Instrumentation/Analytical Method</th>
<th>Sample Collection</th>
</tr>
</thead>
<tbody>
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<td><strong>Regulated Emissions</strong></td>
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<td></td>
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<tr>
<td>Carbon Monoxide CO</td>
<td>Non-Dispersive Infrared Detection (NDIR)</td>
<td>Continuous Collection</td>
</tr>
<tr>
<td></td>
<td>Gas chromatography (GC)</td>
<td>Tedlar Bag</td>
</tr>
<tr>
<td>Carbon Dioxide CO2</td>
<td>Non-Dispersive Infrared Detection (NDIR)</td>
<td>Continuous Collection</td>
</tr>
<tr>
<td>Oxides of Nitrogen NOx</td>
<td>Heated Chemiluminescence Detection</td>
<td>Continuous Collection</td>
</tr>
<tr>
<td>Total Hydrocarbons THC</td>
<td>Heated Flame Ionization Detection (HFID)</td>
<td>Continuous Collection</td>
</tr>
<tr>
<td>Particulate Matter PM</td>
<td>Gravimetric Procedure</td>
<td>70 mm Pallflex T60A20 Filters</td>
</tr>
<tr>
<td><strong>Unregulated Emissions</strong></td>
<td></td>
<td></td>
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<tr>
<td>Soluble Organic Fraction SOF</td>
<td>Dichloromethane Soxhlet Extraction</td>
<td>47 mm Pallflex T60A20 Filters</td>
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<tr>
<td>Organic Carbon /Elemental Carbon EC/OC</td>
<td>Thermal/Optical Transmittance (TOT)</td>
<td>Quartz coated Filters</td>
</tr>
<tr>
<td>Particle Phase Sulfate</td>
<td>Ion Chromatography (IC)</td>
<td>Teflon membrane filters</td>
</tr>
<tr>
<td>Sulfur Dioxide SO2</td>
<td>Ion Chromatography (IC)</td>
<td>Potassium Carbonate Coated Filters</td>
</tr>
<tr>
<td>Carbonyl Compounds</td>
<td>High Performance Liquid Chromatography (HPLC)</td>
<td>2,4-DNPH coated- Silica Gel Cartridges</td>
</tr>
<tr>
<td>Volatile Organic Compounds VOC</td>
<td>Gas Chromatography - Flame Ionization Detection (GC-FID)</td>
<td>Tedlar Bag</td>
</tr>
<tr>
<td>Methane and Light Hydrocarbon Compounds</td>
<td>Gas Chromatography (GC)</td>
<td>Tedlar Bag</td>
</tr>
<tr>
<td>Polycyclic Aromatic Hydrocarbons (PAH) and Nitro-PAH</td>
<td>High resolution gas chromatography/ mass spectrometry (HRGC/MS)</td>
<td>Pallflex T60A20 Filter and Polyurethane Foam</td>
</tr>
<tr>
<td>Particulate Size/Number Distribution</td>
<td>Electric Low Pressure Impactor (ELPI)</td>
<td>Continuous Collection</td>
</tr>
<tr>
<td></td>
<td>Scanning Mobility Particle Sizer (SMPS)</td>
<td>Continuous Collection</td>
</tr>
</tbody>
</table>
Regulated Emissions - CBD Cycle

THC & CO: CNG > CRDPF
CNG Bus #824 NOx
The results from regulated emissions testing for total hydrocarbon (THC), CO, particulate matter (PM) and NOx, and calculated fuel economy (FE) and CO₂ for CNG and CRDPF buses are presented here. The most significant differences between CRDPF and CNG are the emissions for THC and CO, which are much greater for CNG than for CRDPF. Higher CO and THC or CH₄ emissions for CNG are consistent with the results of prior studies. NOx emissions from CNG Bus #824 were greater than those from the other two CNG buses and both the CRDPF buses. While prior studies have found a large variability in NOx emissions from CNG buses, the atypical results from CNG Bus #824 found in this study may be due to frequent backfires as discussed below.
SOx Emissions – CBD Cycle

- SO2: diesel > CNG, CRDPF
- CRDPF: SO2 → SO4

Emission rate (mg/mile)
A comparison of emissions of $\text{SO}_2/\text{SO}_4$ from CNG, diesel and CRDPF buses for the CBD driving cycle is shown here. Higher $\text{SO}_2/\text{SO}_4$ emissions are expected from the OEM diesel buses than from the CNG buses since 30 ppm S diesel fuel has a much higher sulfur concentration than CNG. Yet the CNG buses produced similar $\text{SO}_2$ emissions to the CRDPF buses (which also use 30 ppm S sulfur fuel), suggesting that lube oil may be a significant contributor since CNG fuel is low in sulfur.

In contrast to $\text{SO}_2$, emissions of PM phase $\text{SO}_4$ from CRDPF buses were much higher than those from CNG buses. Installation of a CRDPF on a diesel bus did not alter the total $\text{SO}_x$ emission, but only converted a portion of the $\text{SO}_2$ to PM phase $\text{SO}_4$, a process related to the release of sulfate by the CRDPF.
Carbonyl Emissions – CBD Cycle

CNG >> OEM diesel (formaldehyde)
CRDPF: below mdl
A comparison of carbonyl emissions from CNG, diesel and CRDPF buses for the CBD driving cycle is shown here. Formaldehyde, acetaldehyde and acetone are the dominant carbonyl species in the exhaust of diesel and CNG buses. The CNG buses produced more carbonyls, up to an order of magnitude greater than diesel buses. Further, greater than 96% of the total carbonyl emissions from CNG buses were formaldehyde, in contrast to 60-65% formaldehyde for the diesel buses. Emissions of other carbonyl species from CNG buses were similar to those from diesel buses, with the exception of formaldehyde. The difference in formaldehyde emission is the reason for the large difference of total carbonyl emissions between CNG and diesel buses. Due to the toxic nature of formaldehyde, this may be one of the reasons contributing to the observed higher potential toxicity from CNG buses reported by others. None of the CNG buses tested were equipped with an oxidation catalyst. For the CRDPF buses tested, the reduction of carbonyls by the oxidation catalyst in the CRDPF was so efficient that most carbonyls were below the method detection limit.
Speciated Hydrocarbon Emissions – CBD Cycle

CNG >> OEM diesel & CRDPF
Over 100 volatile hydrocarbon species were measured in this project, and a selection of volatile hydrocarbon emissions of interest from CNG, diesel and CRDPF buses during the CBD driving cycle are shown here. It can be seen that emissions of benzene, ethylene, propylene, and toluene from the CNG buses were much higher than those from diesel buses even without CRDPF but with DOC. Emissions of these compounds were further reduced when the diesel bus was retrofitted with CRDPF, although the reduction percentages varied from compound to compound. In addition to formaldehyde, these species may also contribute to the reported higher potential toxicity from CNG buses.
Polyaromatic Hydrocarbon (PAH) Emissions – CBD Cycle

CNG ~ diesel
- lube oil?
- thermal synthesis?
CRDPF lowest
Emissions of polyaromatic hydrocarbons (PAHs) identified and quantified in this study for CNG, diesel and CRDPF buses during the CBD driving cycle are shown here. Although CNG as a fuel contains much lower levels of PAHs than diesel fuel, similar PAH emissions were observed for CNG buses and diesel buses. This may indicate the presence of a pathway leading to elevated PAH levels in CNG bus exhaust, due to possible lube oil burning or thermal synthesis during CNG combustion (further analysis is warranted). Much lower PAH emissions were observed for the CRDPF buses, which may support recent observations showing unfavorable toxicity from CNG bus exhaust compared to CRDPF bus exhaust. Measurements of nitro-PAH (nPAH) emissions indicated slightly lower nPAH emissions from CNG buses than from diesel buses without CRDPF, and comparable nPAH emissions from CNG and CRDPF buses.
For one of the CNG buses tested (Bus #824), a sharp spike in PM number concentration during the CBD cycle was clearly observed both by the ELPI and the SMPS, as is shown by a time series of data collected during the CBD cycle. The ELPI results (left-hand, red axis) show a very sharp increase in particulates, most strongly in the smallest measured particle sizes, 30 and 60 nm. The SMPS results (right-hand, green axis) show a very sharp increase at the 10 nm size measured during the run. Detection by two independent instruments based on different fundamental principles indicates that this is an observed phenomenon rather than an instrumental artifact. This PM spike was also accompanied by an audible backfiring noise and a change in engine performance outwardly perceptible to the driver, as well as a large increase in NOx emissions as shown previously. This phenomenon was observed during multiple testing cycles for both the CBD and NYB driving cycles.

Many of NYCT’s buses in the fleet from which the CNG test buses were drawn had previously experienced backfiring to some degree. This behavior did not significantly affect their performance, could not be traced to a verifiable defect or mis-adjustment, and was generally accepted by bus operators as normal for these buses. The probable cause of the backfiring was an over-fueling event resulting in post-combustion ignition of excess fuel downstream of the combustion chamber, most likely caused by fuel governing components that are not working in an optimal way. After numerous reports of poor performance from the field, including incidents of backfiring, Detroit Diesel developed upgraded fuel system components which were installed on previously fielded engines as a retrofit campaign. At the time of testing, NYCT’s CNG bus fleet was undergoing this upgrade campaign, and anecdotal evidence after completion of the campaign on the entire fleet indicates that the product update virtually eliminated the previously experienced backfiring condition during on-road driving. Two of the CNG buses tested (#854 and #975) had been retrofit with the new components prior to the testing, while bus #824 had not been retrofit. When interpreting the results of this testing, the results from CNG bus #824 should be seen as typical of buses in service prior to the product upgrade and the results from CNG buses #854 and #975 as typical of buses in service after the upgrade. As such, the entire data set is representative of the range of results expected from buses in “real world” service.
Backfiring During CBD Cycle
ELPI Time Series - Backfire #2 Expanded Scale
CNG Bus #824 03/22/01 CBD #1 ott026

Number Concentration, \( dN/d\log D_p \) (#/cc)

Elapsed Time (s)

- 110 nm
- 180 nm
- 270 nm
- 420 nm
- 680 nm
- 1100 nm
- 1670 nm
- 2600 nm
- 4200 nm
- 6800 nm
While backfiring by CNG Bus #824 most strongly affected the smallest particle sizes measured, it also affected all particle sizes. The magnitude of the increase in PM due to the backfire decreased with increasing particle size. This figure shows an expanded view of the ELPI data for the same backfire event at 500 s shown previously, with the magnitude of the PM increase decreasing for particle sizes from 110 to 6800 nm.
Backfire events for CNG Bus #824 measured by the ELPI and SMPS were always also outwardly detectible and were observed solely during decelerations. This figure shows a time series of the ELPI data at 30 nm from 313 s to 515 s elapsed time during a typical CBD cycle when three backfires occurred (black circles, left-hand axis). The corresponding changes in engine speed are shown as blue circles (right-hand axis). The red lines indicate the initiation of measured backfire events, which were observed to occur solely during decelerations, never during accelerations.
Backfiring During CBD Cycle?

CNG Bus #854 - ELPI and SMPS Time Series

CNG Bus #854 05/02/01 CBD #7

ELPI Data, Number (#/cc)

SMPS Data, 10 nm only, dN/dlogD

Elapsed Time (s)
Although it had undergone the retrofit, CNG bus #854 demonstrated characteristically similar sharp increases in PM observed by both the ELPI and SMPS, but with a much lesser magnitude than those exhibited by CNG bus #824. These may be due to “micro-backfires” occurring due to a similar mechanism to those experienced by CNG bus #824, but to a much smaller degree. Unlike the backfires experienced by CNG bus #824, those shown here for bus #854 are of ~10^3 lesser magnitude, were not accompanied by a corresponding increase in NOx emissions, and produced no apparent audible or other outward evidence that backfiring was occurring. While the retrofit did not eliminate such “micro-backfires” for CNG bus #854, it clearly reduced their magnitude so that they did not significantly impact either the on-road driving performance or the overall composite size distribution (see below).
ELPI Composite Size Distribution – CBD Cycle

- Avg OEM diesel (6019, 6065)
- Avg CRDPF (6019, 6065)
- Avg CNG (854, 975) without backfires
- CNG (824) with backfires
- CNG (824) backfires removed
The data shown here is the composite size distribution measured by the ELPI over the 10 minute CBD cycle for each of the cases. Overall, results were comparable for CRDPF and CNG for all particle sizes, i.e., a 90% reduction from OEM diesel buses.

However, some differences can be seen in the region from 30 to 180 nm. In this range, PM emissions from CNG buses #854 and 975 were somewhat lower than those from the CRDPF buses. Also, the effect of backfiring for CNG bus #824 can be seen in the higher PM emissions in the 30 and 60 nm size bins, as exhibited in the examples of ELPI and SMPS data shown previously. If the backfires are removed from the readings for CNG bus #824, the resultant distribution becomes comparable to that exhibited by CNG buses #854 and 975, which did not exhibit significant backfiring due to the retrofit.

Similar results to those found for the CBD cycle were also found for the NYB cycle. Although CNG buses #854 and #975 still demonstrated some backfires for the NYB cycle even after the retrofit, they were of roughly the same magnitude as the “micro-backfires” discussed above and similarly did not significantly impact on-road driving performance.
Conclusions

- **Regulated Emissions:**
  - CNG >> CRDPF for THC and CO

- **Unregulated Emissions:**
  - **SOx**
    - OEM Diesel > CNG, CRDPF
    - Conversion of SO2 to particulate phase SO4 in the CRDPF
  - **Carbonyl**
    - CNG >> OEM Diesel due to formaldehyde, CRDPF below detection limit
  - **Speciated HC**
    - CNG >> OEM Diesel and CRDPF (benzene, toluene, ethylene, propylene)
  - **PAHs**
    - CNG ~ OEM Diesel, CRDPF lowest
    - Source of CNG PAHs unclear (lube oil? thermal synthesis?)
    - No conversion of PAHs to nitro-PAHs in the CRDPF
  - **May contribute to possible increased toxicity of CNG emissions observed by others.**

- **Particulates:**
  - Overall CNG equal to CRDPF, i.e., 90% reduction in PM from OEM Diesel
  - **Backfiring** observed for CNG Bus #824:
    - Sharp particulate increase observed by both ELPI and SMPS
    - Audible noise and change in performance detectable to driver
    - Increase in NOx emissions
    - Significantly affects 30 – 180 nm size range, although all sizes affected
    - Occurred only on decelerations
  - **Retrofit campaign eliminated** backfiring during on-road driving
  - Small magnitude “micro-backfiring” exhibited by retrofitted CNG bus #854 but did not significantly affect composite particle size distribution or on-road driving performance.
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