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Testing of unregulated emissions from heavy duty natural gas vehicles. Part 2

(Provning av oreglerade emissioner i tunga metangasdrivna fordon. Del 2)

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"Catalyzing energygas development for sustainable solutions"



Testing of unregulated emissions from heavy duty natural gas vehicles part 2, complement to 2013:289 (Provning av oreglerade emissioner i tunga metangasdrivna fordon del 2, komplement till 2013:289)

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Also included:

- Appendix B1. Engine oil: Specific gas engine requirements and suitable quality
- Appendix B2. Engine oil formulation and consumption: Influence on particle emissions A literature study with comments

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Malmö 2014

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Malmö, Sweden 2014

Martin Ragnar Chief Executive Officer

Summary

This report is a complement to SGC report 2013:289 "Testing of unregulated emissions from heavy duty natural gas vehicles". The main report presents the testing and test results from three heavy duty natural gas vehicles. This report includes the test results from an additional HD NGV tested in the same manner as the previous three ones. Details of the testing can be found in the main report.

The tested vehicle was a CNG fuelled garbage truck approved according to emission standard Euro VI. The vehicle is denoted "vehicle no 4" in this report.

The combustion principle of the engine was stoichiometric. The vehicle was equipped with waste gate turbo, EGR and a three-way catalytic converter. The fuel used in the test was CNG from the tank of the vehicle which was filled up on a commercial fuelling station.

The tests were performed with both cold and warm engine.

The emissions analyzed were the regulated exhaust components carbon monoxide (CO), total hydrocarbons (NMHC and CH₄), nitrogen oxides (NOx), ammonia (NH₃) and particulate matter (PM). Also carbon dioxide (CO₂) was measured.

In addition were unregulated emissions analyzed. The compounds analyzed were aldehydes, PAH, N₂O, particulate number (PN) and particle size distribution.

Hardly any NO₂ was detected, and the levels of NO were approximately 10% of the levels detected for the DDF vehicle in the cold start tests and even less in the warm start tests.

The vehicle emitted more N_2O during the cold start test compared to the DDF vehicle (vehicle 2). During the warm start tests were no emissions of N_2O detected.

The ammonia emissions detected were above the 10 ppm limit during the cold start tests and slightly below during the warm start test. The phenomenon is common in positive ignited vehicles equipped with a three-way-catalyst (TWC).

For all particle sizes, it can be concluded that cold start tests generated slightly more particulates than warm start tests and vehicle no 4 (EU VI) together with vehicle no 3 (lean-burn) generates the least total number of particles whereas the MK1 reference vehicle and the DDF vehicle generates the most, regardless of fuel mode.

Just as for the dedicated gas vehicles presented in the main report were the aldehyde emission levels of this vehicle low. Formaldehyde and acetaldehyde were dominating compounds. The levels of formaldehyde and acetaldehyde during the cold start tests showed a large variance and the influence of the start temperature is therefore unclear. However, the vehicle shows significantly higher levels of THC during the cold start tests and it can therefore, based on previous experience, be suspected that the cold start generates more formaldehydes compared to the warm start.

The amount of total PAH from vehicle no 4 is low compared to the diesel vehicles (DDF, diesel mode, and MK1 reference), and vehicle no 1. For these tests, the cold starts generate approximately 30 % more PAH than the warm start tests.

Both particulate and volatile PAHs from Vehicle no 4 are in line with the results from previous tested vehicles.

Sammanfattning

Denna rapport är ett komplement till SGC rapport 2013: 289 "Provning av oreglerade emissioner i tunga metangasdrivna fordon ". I huvudrapporten presenteras provning samt provresultat från 3 tunga metangasdrivna fordon. Denna rapport inkluderar provresultaten från ytterligare en HD NGV provad på samma sätt som de tidigare 3. Provuppgifter presenteras i detalj i huvudrapporten.

Det provade fordonet var en CNG driven sopbil godkänd enligt emissionsstandarden Euro VI. Fordonet betecknas "fordon nr 4" i denna rapport.

Motorns förbränningsprincip var sk stökiometrisk. Fordonet var utrustat med waste gate turbo, EGR och en trevägskatalysator. Bränslet som används vid provningen var CNG från fordonets egen tank som fylldes på en vanlig tankstation.

Proverna genomfördes med både kall och varm motor.

De emissioner som analyserade var de reglerade avgaskomponenterna kolmonoxid (CO), kolväten (NMHC och CH4), kväveoxider (NOx), ammoniak (NH3) och partiklar (PM). Även koldioxid (CO2) mättes.

Utöver detta analyserades oreglerade emissioner. De föreningar som analyserades var aldehyder, PAH, N2O, partikelantal (PN) och partikelstorleksfördelning.

Knappast någon NO2 detekterades, och nivåerna av NO var ungefär 10% av de nivåer som uppmättes för DDF fordonet i kallstartsprov och ännu mindre i varmstartsprovning.

Fordonet avger mer N2O vid kallstarttest jämfört med DDF fordonet (fordon nr 2). Under varmstartsproven detekterades inga utsläpp av N2O.

De ammoniakutsläpp som uppmättes låg över 10 ppm gränsen vid kallstartsproven och något under vid varmstartsprovning. Fenomenet är vanligt hos gnisttända fordon försedda med en trevägskatalysator.

Precis som för de dedikerade gasfordonen som presenteras i huvudrapporten var utsläppen av aldehyder från detta fordon låga. Formaldehyd och acetaldehyd vad de dominerade aldehyderna. Halterna av formaldehyd och acetaldehyd under kallstartsproven visade stor standardavvikelse och därmed är påverkan av starttemperatur oklar. Bilen släpper dock ut betydligt högre halter av THC under kallstartsproven och det kan därför, baserat på tidigare erfarenheter, misstänkas att kallstart genererar mer formaldehyd jämfört med varmstart.





Figur A. Total mängd aldehyder för samtliga fordon

För alla partikelstorlekar kan konstateras att kallstartprover genererar något fler partiklar än varmstartprover och fordon nr 4 (EU VI) tillsammans med fordon nr 3 (lean-burn) genererade den lägsta mängden partiklar medan MK1 referensfordon och DDF fordonet genererar flest, oavsett bränsle mode.



Figur B. Partikelstorleksfördelning för alla fordon vid varmprov



Figur C. Partikelantal för alla provade fordon

Mängden totala PAH från bil nr 4 är låg jämfört med dieselfordonen (DDF, diesel mode och MK1 referens), och bil nr 1. För dessa prov genererade kallstartproven ungefär 30% mer PAH än varmstartproven.

Både partikulära och flyktiga PAH från fordon nr 4 är i linje med resultaten från tidigare testade fordon.



Figur D. Total mängd PAH, alla fordon, varma och kalla prov

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1. Experimental

1.1 Test Vehicle

Data of the test vehicle is presented in table 1. The vehicle is referred to as "Vehicle no 4". Vehicle no 1-3 are presented in the main report. The European emission standards for gas engines are presented in table 2. Regulated emission components are presented in table 3.

	Vehicle nr 4
Year model:	2014
Mileage [km]:	9790
Combustion principle:	Stoichiometric spark ignited
Fuel:	CNG
After treatment system	Wastegate turbo, EGR, TWC
Engine power [kW]:	250
Emission standard	Euro VI

	СО	NMHC	CH₄	NOx	NH₃	РМ	PN#*	Test Cycle
	g/kWh	g/kWh	g/kWh	g/kWh	ррт	g/kWh	10 ¹² /kWh	
Euro III	5.45	0.78	1.6	5.0	-	0.16		ETC
Euro IV	4.0	0.55	1.1	3.5	-	0.03		ETC
Euro V	4.0	0.55	1.1	2.0	-	0.03		ETC
EEV	3.0	0.40	0.65	2.0	-	0.02		ETC
Euro VI	4.0	0.16	0.5	0.46	10	0.01	0.6	WHTC

 Table 2. European emission legislation for gas engines (engine test)

*For diesel engines; PN limit for positive ignition engines TBD

NMHC CH₄ Drive cycle CO NOx CO₂ PM PN (g/kWh) (g/km) (g/kWh) (g/km) (g/kWh) (g/km) (g/kWh) (g/km) (g/kWh) (g/km) (g/kWh) (g/km) (#/kWh) (#/100km) 0,25 0,53 0,006 0,007 2,2E+12 2,6E+12 1,44 1,66 0,07 0,08 0,29 0,46 734 850 WHVC cold 1 1,74 0,06 0,34 0,54 0,007 2,1E+12 2,4E+12 0.29 777 2,02 0,06 0,63 904 0,006 WHVC cold 2 2,0E+12 2,44 0,05 0,06 0,27 0,32 0,51 904 0,005 0,006 2,3E+12 2,84 0,43 777 WHVC cold 3 Average 2.4E+12 1.87 0.07 0.27 0.007 2.1E+12 2.17 0.06 0.32 0.48 0.56 763 886 0.006 WHVC cold 0,51 0,60 0,01 0,01 0,02 0,02 0,05 0,06 25 32 0,001 0,001 1,2E+11 1,3E+11 Stdev 1,7E+12 0,55 0,05 0,007 1,5E+12 0,64 0,05 0,01 0,01 0,14 0,16 710 824 0,006 WHVC warm 1 0,53 0.62 0,04 0,04 0,01 0,13 0,15 708 819 0,007 9,4E+11 1,1E+12 WHVC warm 2 0,01 0,006 Average 0,54 0,05 1,2E+12 1,4E+12 0,63 0,04 0,01 0,01 0,13 0,16 709 821 0,006 0,007 WHVC warm 3,7E+11 0,01 0,02 0.01 0.01 0.00 0.00 0.01 0.01 4 0,000 0,000 4,3E+11 1 Stdev

Table 3. Regulated emission results

1.3 Test Program

The vehicle has been tested during the WHVC test cycle. Both cold tests and warm tests have been performed. Regulated emissions have been measured as well as unregulated emissions.

The following have been measured:

- Regulated emissions and CO₂
- Particle number and size distribution (ELPI)
- Carbonyl compounds (Formaldehyde, Acetaldehyde, Acetone, Acrolein, Propionaldehyde, Crotonaldehyde, Methyletylketon, Butyraldehyde, Benzaldehyde)
- PAH analysis (chemical characterization, 20 different components);
- NH₃, N₂O, NO and NO₂ been measured with FTIR.

2. Results and discussion

Test results from vehicle no 1-3 are presented in the main report. Detailed test results (particulate size distribution, Aldehydes and PAH's) from vehicle no 4 are presented in the appendices of this report.

2.1 FTIR test results

The results from the FTIR measurements are presented in table 4.

Hardly any NO₂ was detected, and the levels of NO were approximately 10% of the levels detected for the DDF vehicle in the cold start tests and even less in the warm start tests.

The vehicle emitted about three times as much N_2O during the cold start test compared to the DDF vehicle (vehicle 2). During the warm start tests were almost no emissions of N_2O detected.

The ammonia emissions detected were above the Euro VI 10 ppm limit during the cold start tests and slightly below during the warm start test.

	N ₂ C	N	C	NC	2	NH ₃		
	(g/kWh)	(g/kWh) (g/km)		(g/km)	(g/kWh) (g/km)		ppm (average)	
WHVC cold 1	0,07	0,08	0,50	0,58	n.d	n.d	16,1	
WHVC cold 2	0,06	0,07	0,58	0,68	n.d	n.d	16,1	
WHVC cold 3	0,08	0,10	0,46	0,53	n.d	n.d	20,2	
Average	0,07	0,08	0,51	0,60	-	-	17,5	
STDEV	0,01	0,02	0,06	0,08	-	-	2,4	
WHVC warm 1	0,00	0,00	0,12	0,14	0,00	n.d	8,0	
WHVC warm 2	0,00	0,00	0,11	0,13	0,00	0,00	8,7	
Average	0,00	0,00	0,12	0,14	-	-	8,4	
STDEV	0,00	0,00	0,01	0,01	-	-	0,5	

Table 4



2.2 Particulate size distribution test results

The results are presented as diagrams in Figure 1-3 and numerical results are presented in Appendices. For all particle sizes, it can be concluded that cold start tests generated slightly more particulates than warm start tests.



Figure 1. Particle size distribution of vehicle no 4, comparing the warm and the cold tests

Figure 2 and 3 shows that the dedicated gas vehicles generate significantly fewer particles than the DDF vehicle and the MK1 reference. For vehicle no 1, there are the smaller particle sizes (<0,1 μ m) causing the larger total particle number.



Figure 2. Particle size distribution of all tested vehicles during the warm tests

In Figure 3 it can be seen that vehicle no 4 (EU VI) together with vehicle no 3 (lean-burn) generates the least number of particles while the DDF vehicle and the MK1 reference vehicle generates the most, regardless of fuel mode.



Figure 3. Particle number of all tested vehicles

2.3 Measurement of aldehydes

Aldehyde emission results from the test vehicle are shown in Figure 4.

Just as for the dedicated gas vehicles presented in the main report were the aldehyde emission levels of this vehicle low. Formaldehyde and acetaldehyde were dominating compounds. The levels of formaldehyde and acetaldehyde during the cold start tests showed a large variance and the influence of the start temperature is therefore unclear. However, the vehicle shows significantly higher levels of THC during the cold start tests and it can therefore, based on previous experience, be suspected that the cold start generates more formaldehydes compared to the warm start.

Small portions of Propionaldehyde, Crotonaldehyde, Butyraldehyde, Benzaldehyde, Valeraldehyde and Hexaldehyde were also detected.







Figure 4. Aldehyde results, vehicle no 4



Figure 5. Total amount of aldehydes, all vehicles



2.4 PAH analysis

The PAH in the emissions can be derived from unburned residues of fuel and as a byproduct from the combustion. Gas vehicles generally have higher oil consumption in comparison to diesel vehicles and both the amount and the quality of the oil may influence the levels of PAH.

PAH emission results are presented in figure 6-8 and detailed results from vehicle no 4 are resented in appendices.

For vehicle no 4, samples from two cold start tests and two warm start tests were taken, but due to the small amount of sample material extracted from the filter and the PUF, the extracts from the two cold start tests and the two warm start tests, respectively, had to be merged in order to enable the analyze.

The amount of total PAH from vehicle no 4 are low compared to the diesel vehicles (DDF, diesel mode, and MK1 reference), and vehicle no 1. For these tests, the cold starts generate approximately 30 % more PAH than the warm start tests.



Figure 6. Total amount of PAH, all vehicles, warm and cold tests

Both particulate and volatile PAHs from Vehicle no 4 are in line with the results from previous tested vehicles.



Figure 7. Volatile PAH, all vehicles, warm and cold tests



Figure 8. Particulate PAH, all vehicles, warm and cold tests

3. Conclusions

Relatively high emissions of ammonia were detected and the Euro VI 10 ppm limit was exceeded during the cold start tests and only slightly below during the warm start test.

It has been widely documented that in a vehicle with a positive ignited engine and a three-way catalyst, ammonia may be formed as a secondary pollutant during the NOx reduction process over the three-way catalyst.

Ammonia is generated by reduction of NO. Under regular circumstances, NO is reduced to N_2 in the three-way catalyst. Under favorable conditions the reduction of NO does not stop at N_2 but continues to NH₃. The most clear "favorable condition" mentioned in literature is rich fuel mixture. Vehicle no 4 runs stoichiometric, but during the cold start there is a short period of a more rich A/F-ratio, which could explain the higher NH₃ values. There is also an effect of catalyst temperature which has been investigated in literature, but the results are not fully consequent [1, 2, 3].

Typically, conditions that favor formation of NH_3 also favor formation of N_2O . Just as for NH3, the emissions of N_2O are higher during the cold start tests.

The particle number for the Euro VI vehicle is lower in comparison to the other stoichiometrically operated vehicle (no 1), more in line with the lean burn vehicle no 3. When looking at the particle size distribution, the three dedicated NG vehicles are similar when looking at particle sizes larger than 0,1 μ m. For vehicle no 1, there are the smaller particle sizes (<0,1 μ m) causing the larger total particle number.

Just as for all other tested vehicles the cold start generates slightly more particles compared to the warm start.

The aldehyde emission levels of this vehicle is low, just as for the other dedicated NG vehicles. Formaldehyde and acetaldehyde were dominating compounds. Due to large variances of the levels of aldehydes between the tests is it hard to draw a conclusion regarding influence of start temperature. However, higher levels of methane during the cold start test have for the other vehicles also resulted in higher levels of formaldehyde and it can be suspected that the same is valid also for this vehicle.

Low levels of PAH was detected. The volatile PAH's were in line with both the dedicated NG vehicles and the particulate PAH's were on the same level as the lean-burn vehicle (no 3).

4. Literature

[1] Heeb, N.V., Forss, A.M., Bruhlmann, S., Luscher, R., Saxer, C.J. and Hug, P., (2006). Threeway catalyst-induced formation of ammonia: velocity and acceleration dependent emission factors. Atmospheric Environment, Vol. 40, pp. 5986-5997

[2] DiGiulio C. D., Pihl J.A., Parks II J.E., Amiridis M.D., Toops T.J. (2014) Passive-ammonia selective catalytic reduction (SCR): Understanding NH3 formation over close-coupled three way catalysts (TWC). Catalysis Today, Vol. 231, pp 33-45

[3] <u>http://www.nettinc.com/information/emissions-faq/ammonia-hydrogen-sulfide-to-three-way-catalyst</u>



5. List of abbreviations

A/F Ratio	Air / Fuel Ratio
ACEA	European Automobile Manufacturers Association
API	American Petroleum Institute
BSFC	Brake Specific Fuel Consumption
BTX	"Benzene, Toluene, Xylene"
CBG	Compressed Bio Gas
CCV	Closed Crankcase Ventilation
CH ₄	Methane
CI	Compression Ignited
CNG	Compressed Natural Gas
CO	Carbon Monoxide
CO ₂	Carbon Dioxide
DDF	Diesel Dual Fuel
DI	Direct Injection
DPF	Diesel Particulate Filter
DPM	Diesel Particulate Matter
ECU	Electronic Control Unit
EEV	Enhanced Environmental friendly Vehicle
EGR	Exhaust Gas Recirculation
ELPI	Electrical Low Pressure Impactor
FAME	Fatty Acid Methyl Ester
FTIR	Fourier Transform InfraRed
GFV	Gas Fuelled Vehicles
GHG	Green House Gases
H/C	Hydrogen to Carbon ratio
HC, THC	Hydrocarbons, Total Hydrocarbons
HD	Heavy Duty
HPLC	High-performance liquid chromatography
HVO	Hydrotreated Vegetable Oil
IEA – AMF	International Energy Agency – Advanced Motor Fuels
IVL	Swedish Environmental Research Institute
LBG	Liquefied Bio Gas
LDV	Light Duty Vehicle
LNG	Liquefied Natural Gas
MK1	Environmental Class 1 (Miljöklass 1)
MPFI	Multi Port Fuel Injection
MS	Mass Spectrometer
NA	Naturally Aspirated
NEDC	New European Driving Cycle
NG	Natural Gas
NGV	Natural Gas Vehicle
NMHC	Non-Methane Hydrocarbons

NOx	Oxides of Nitrogen
OBD	On-Board Diagnostics
OEM	Original Equipment Manufacturer
OIGI	Oil Ignition Gas Injection
PAH	Poly cyclic Aromatic Hydrocarbons
PFI	Port Fuel Injected
PGM	Platinum Group Metal
PM	Particulate Matter
PN	Particulate Number
RON	Research Octane Number
(low) SAP	(low) Sulphated ash, Phosphorus and Sulphur
SCR	Selective Catalytic Reduction
SGC	Svenskt Gastekniskt Center
SI	Spark Ignited
TAN	Total Acid Number
TBN	Total Base Number
TCDD	2,3,7,8-tetraklordibenso-p-dioxin
TWC	Three Way Catalyst
UV	Ultra Violet
WHTC	World Harmonized Transient Cycle
WHVC	World Harmonized Vehicle Cycle



Appendix A1, Particle size distribution results

Table 5. Vehicle no 4

	Vehicle no 4, cold tests, average													
Particle aerodynamic diameter [µm]		0,03	0,063	0,109	0,173	0,267	0,407	0,655	1,021	1,655	2,52	4,085	6,56	tot
	[#/test]	7,35E+13	4,62E+13	4,93E+13	6,76E+13	2,26E+13	1,02E+13	3,49E+12	1,40E+12	5,91E+11	3,55E+11	1,16E+11	1,48E+11	9,23E+13
	[#/km]	3,70E+12	2,32E+12	2,48E+12	3,40E+12	1,14E+12	5,13E+11	1,76E+11	7,08E+10	2,98E+10	1,78E+10	5,86E+09	7,43E+09	4,64E+12
	[#/kWh]	3,19E+12	2,00E+12	2,14E+12	2,93E+12	9,82E+11	4,41E+11	1,51E+11	6,10E+10	2,56E+10	1,54E+10	5,05E+09	6,37E+09	4,00E+12

Vehicle no 4, warm tests, average														
Particle aerodynamic diameter [µm]		0,03	0,063	0,109	0,173	0,267	0,407	0,655	1,021	1,655	2,52	4,085	6,56	tot
	[#/test]	4,41E+12	1,19E+13	2,25E+13	3,77E+13	1,21E+13	4,61E+12	1,33E+12	4,76E+11	1,74E+11	1,00E+11	4,67E+10	3,08E+10	2,37E+13
	[#/km]	2,21E+11	5,99E+11	1,13E+12	1,89E+12	6,08E+11	2,31E+11	6,66E+10	2,39E+10	8,73E+09	5,04E+09	2,34E+09	1,55E+09	1,19E+12
	[#/kWh]	1,91E+11	5,17E+11	9,71E+11	1,63E+12	5,25E+11	1,99E+11	5,74E+10	2,06E+10	7,52E+09	4,34E+09	2,02E+09	1,33E+09	1,03E+12

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Appendix A2, Aldehyde emission results

	WHVC cold 1	WHVC cold 2	WHVC cold average	Stand- ard devia- tion	WHVC warm 1	WHVC warm 2	WHVC warm average	Stand- ard devia- tion
	(mg/km)	(mg/km)	(mg/km)	(mg/km)	(mg/km)	(mg/km)	(mg/km)	(mg/km)
Substance								
Formaldehyde	2,327	3,762	3,045	1,015	2,660	2,479	2,569	0,128
Acetaldehyde	1,330	2,083	1,706	0,532	2,083	2,034	2,058	0,035
Acetone	n.d.	n.d.			n.d.	n.d.		
Acrolein	n.d.	n.d.			n.d.	n.d.		
Propionaldehyde	0,269	0,396	0,333	0,090	0,496	0,395	0,446	0,072
Crotonaldehyde	0,104	0,134	0,119	0,021	0,149	0,188	0,169	0,028
Butyraldehyde	0,224	0,291	0,257	0,047	0,353	0,367	0,360	0,010
Benzaldehyde	0,066	0,073	0,070	0,006	0,103	0,128	0,116	0,018
Isovaleraldehyde	n.d.	n.d.			n.d.	n.d.		
Valeraldehyde	0,086	0,121	0,103	0,025	0,144	0,184	0,164	0,028
o-Tolualdehyde	n.d.	n.d.			n.d.	n.d.		
p-Tolualdehyde	n.d.	n.d.			n.d.	n.d.		
Hexaldehyde	0,159	0,223	0,191	0,045	0,330	0,260	0,295	0,049
Dimetyl-Benzalde- hyde	n.d.	n.d.			n.d.	n.d.		

Table 6. Vehicle no 4, distance specific aldehyde results

	WHVC cold 1	WHVC cold 2	WHVC cold average	Standard deviation	WHVC warm 1	WHVC warm 2	WHVC warm average	Standard deviation
	(mg/kWh)	(mg/kWh)	(mg/kWh)	(mg/kWh)	(mg/kWh)	(mg/kWh)	(mg/kWh)	(mg/kWh)
Substance								
Formaldehyde	2,012	3,231	2,621	0,862	2,291	2,144	2,218	0,104
Acetaldehyde	1,150	1,788	1,469	0,452	1,794	1,759	1,777	0,025
Acetone	n.d.	n.d.			n.d.	n.d.		
Acrolein	n.d.	n.d.			n.d.	n.d.		
Propionaldehyde	0,233	0,340	0,287	0,076	0,427	0,341	0,384	0,061
Crotonaldehyde	0,090	0,115	0,102	0,018	0,129	0,163	0,146	0,024
Butyraldehyde	0,193	0,250	0,221	0,040	0,304	0,318	0,311	0,010
Benzaldehyde	0,057	0,063	0,060	0,005	0,089	0,111	0,100	0,016
Isovaleraldehyde	n.d.	n.d.			n.d.	n.d.		
Valeraldehyde	0,074	0,104	0,089	0,021	0,124	0,159	0,141	0,025
o-Tolualdehyde	n.d.	n.d.			n.d.	n.d.		
p-Tolualdehyde	n.d.	n.d.			n.d.	n.d.		
Hexaldehyde	0,138	0,192	0,165	0,038	0,284	0,225	0,255	0,042
Dimetyl-Benzalde- hyde	n.d.	n.d.			n.d.	n.d.		

Table 7. Vehicle no 4, break specific aldehyde results



Appendix A3, PAH emission results

Table 8 Particulate PAH emissions, vehicle no 4

	WHVC cold	WHVC warm
	ng/km	ng/km
Phenanthrene	44,9	18,0
Anthracene	13,0	9,3
3-Methylphenanthrene	0,6	3,0
2-Methylphenanthrene	11,4	4,3
2-Methylanthracene	15,2	2,7
9-Methylphenanthrene	8,8	2,3
1-Methylphenanthrene	14,4	5,2
4H-cyclopenta(def)phenanthrene	2,7	1,0
9-Methylanthracene	0,0	0,0
2-PhenyInaphthalene	57,1	32,6
3,6-Dimethylphenanthrene	4,4	3,6
3,9-Dimethylphenanthrene	16,1	5,0
Fluoranthene	87,5	26,2
Pyrene	85,0	24,7
9,10-Dimethylanthracene	0,0	0,0
1-Methylfluoranthene	7,8	2,8
Benz(a)fluorene	2,6	0,8
Benz(b)fluorene	3,2	1,8
2-Methylpyrene	4,4	1,7
4-Methylpyrene	4,4	2,7
1-Methylpyrene	2,8	2,3
Benzo(ghi)fluoranthene	14,0	6,0
Benzo(c)phenanthrene	3,0	2,7
Benzo(b)naphto(1,2-d)thiop	0,8	0,3
Cyclopenta(cd)pyrene	12,4	14,9
Benz(a)anthracene	12,4	11,2
Chrysene	28,1	14,2
3-Methylchrysene	2,5	0,8
2-Methylchrysene	1,3	0,4
6-Methylchrysene	0,7	0,6
1-Methylchrysene	1,4	0,5
Benzo(b)fluoranthene	17,4	8,3
Benzo(k)fluoranthene	4,0	2,8
Benzo(e)pyrene	15,0	1,1
Benzo(a)pyrene	5,4	5,7
Perylene	0,8	0,0
Indeno(1,2,3-cd)fluoranthe	1,1	0,9
Indeno(1,2,3-cd)pyrene	13,6	6,9
Dibenz(a,h)anthracene	1,6	0,4
Picene	0,9	1,3
Benzo(ghi)perylene	0,0	0,0
Dibenzo(a,l)pyrene	4,2	0,8
Dibenzo(a,e)pyrene	1,6	0,0
Coronene	11,9	2,7
Dibenzo(a,i)pyrene	0,0	0,0
Dibenzo(a,h)pyrene	0,0	0,0
Total Particulate PAH	540,5	232,8



Table 9 Volatile PAH emissions, vehicle no1

	WHVC cold	WHVC warm
	ng/km	ng/km
Phenanthrene	288,0	245,0
Anthracene	60,2	58,2
3-Methylphenanthrene	13,6	12,4
2-Methylphenanthrene	28,8	28,2
2-Methylanthracene	31,4	31,6
9-Methylphenanthrene	30,1	27,8
1-Methylphenanthrene	38,6	42,5
4H-cyclopenta(def)phenanthrene	6,6	7,3
9-Methylanthracene	0,0	0,0
2-PhenyInaphthalene	75,1	125,6
3,6-Dimethylphenanthrene	2,6	2,7
3,9-Dimethylphenanthrene	6,0	6,0
Fluoranthene	59,9	11,8
Pyrene	45,9	52,5
9,10-Dimethylanthracene	0,0	0,0
1-Methylfluoranthene	1,4	2,0
Benz(a)fluorene	0,5	0,6
Benz(b)fluorene	0,3	0,8
2-Methylpyrene	0,5	0,6
4-Methylpyrene	0,6	0,7
1-Methylpyrene	0,4	0,6
Benzo(ghi)fluoranthene	0,6	0,4
Benzo(c)phenanthrene	1,8	2,3
Benzo(b)naphto(1,2-d)thiop	0,0	0,0
Cyclopenta(cd)pyrene	0,9	0,4
Benz(a)anthracene	0,4	0,0
Chrysene	1,6	1,2
3-Methylchrysene	0,3	0,9
2-Methylchrysene	0,3	0,3
6-Methylchrysene	0,1	0,3
1-Methylchrysene	0,1	0,8
Benzo(b)fluoranthene	0,6	0,9
Benzo(k)fluoranthene	0,0	0,0
Benzo(e)pyrene	0,0	0,0
Benzo(a)pyrene	0,0	0,0
Perylene	0,0	0,6
Indeno(1,2,3-cd)fluoranthe	0,0	0,0
Indeno(1,2,3-cd)pyrene	0,0	0,0
Dibenz(a,h)anthracene	0,0	0,0
Picene	0,0	0,0
Benzo(ghi)perylene	0,0	0,0
Dibenzo(a,I)pyrene	0,0	0,0
Dibenzo(a,e)pyrene	0,0	0,0
Coronene	0,0	0,0
Dibenzo(a,i)pyrene	0,0	0,0
Dibenzo(a,h)pyrene	0,0	0,0
Total Volatile PAH	697,4	664,9

Appendix B1. Engine oil: Specific gas engine requirements and suitable quality

B1.1 Engine oil quality demands

Currently engines designed to run on CNG, or other methane rich fuels such as CBG, LNG etc., can be divided into three main groups, Bi-fuel, dedicated gas engines and dual-fuel engines. Current dual-fuel engines are often designed so that they can also run purely on diesel making the engine oil choice similar to the original diesel engine. The situation is similar for the Bi-fuel engine, then originating from a petrol engine and needing to keep oil recommendations that are in line with its base design.

When designing a dedicated gas engine for the commercial vehicle market most engines originate from a commercial heavy duty diesel engine.

That heavy duty diesel engine will have its oil quality recommendations, oil filtration system set up and engine oil service interval designed to the demands and requirements expected for a diesel engine. Therefore there is a need to look into the different areas affected when deciding on oil quality demands and service intervals for the gas engine, since the change in fuel and combustion system will result in changes in demands on engine oil and lubrication system. This section will concentrate on the oil quality demands on those commercial heavy duty gas engines originating from commercial heavy duty diesel engines.

B1.1.1 Differences; Diesel, Lean Burn and Stoichiometric Gas Engines

The combustion concept in a diesel, lean burn gas and/or stoichiometric gas engine is clearly different. This results in differences in the load on the engine oil. The differences can be exemplified by concentrating on 4 major parameters that are important for the oil formulation and degradation speed, and listing the general characteristics of the three different combustion concepts, diesel, lean burn gas and stoichiometric gas.

Diesel engines:

- High cylinder pressure (high blow-by¹)
- High soot load
- Relatively high NO_x levels in blow-by
- Relatively low thermal load

Lean Burn gas engines:

- Lower cylinder pressure than diesel (potential for reduced blow-by)
- Low soot load
- Low NO_x levels in blow-by
- Increased thermal load compared to diesel engines

Stoichiometric gas engines:

- Lower cylinder pressure than diesel (potential for reduced blow-by)

¹ Blow-by is the expression used when talking about the gas leakage past the pistons in an internal combustion engine. Often it is combustion gases that are forced past the piston ring area and into the crankcase during the combustion phase. Gases can also leak from the crankcase and up into the combustion chamber, for instance during engine breaking. This is the often referred to as negative blow-by.



- Low soot load

- High NO_x levels in blow-by

- Further increase of the thermal loads compared to Lean Burn gas engines

B1.1.2 Basic Engine Oil Demands

There are five basic tasks for engine oil to fulfil in an engine. They are to lubricate, cool, clean (clean up and/or keep clean), seal and protect the engine from corrosion.

B1.1.3 Lubricating

As the basic engine set up does not differ greatly, lubrication demands will be similar between diesel, lean burn gas and stoichiometric gas engines.

There are two main techniques used when lubricating an engine, hydrodynamic lubrication and boundary lubrication.

Hydrodynamic lubrication occurs when an oil film thick enough is present in between the moving surfaces. Ideally this is the case in for example crank shaft bearings. Then no surface to surface contact is present and the oil is carrying all loads. The friction losses are very small and wear is negligible.

Boundary lubrication occurs where the oil film is not thick enough to completely separate the moving surfaces. The most severe areas where boundary lubrication occurs are the cam and balancing chain and the connecting rod small end bearing.

A number of other areas have lubrication requirements that is somewhere between the hydrodynamic and the boundary lubrication. Typical examples are cam shaft lobes and piston rings. In those areas there is no oil pressure build up from the lubrication system, but the movement between the surfaces builds up a small oil film. That film is not enough to build a sufficient oil film thickness for a pure hydrodynamic lubrication band so there is still a need for functioning boundary lubrication.

An anti-wear additive in the oil is the key to proper boundary lubrication. All modern engine oils have anti wear additives. Often the additives are metal based.

B1.1.4 Cooling

The engine oil plays an important role in engine cooling. A role of thumb is that oil carries off about 40% of the heat that's generated by the engine. The task of cooling the engine will be increased with gas engines compared to diesel. That increase in cooling demands will be higher on stoichiometric than on lean burn gas engines. A dedicated diesel engine oil would not be suitable as the oxidation stability would not be sufficient.

B1.1.5 Keep Clean and Clean Up

Combustion components and other contaminates ends up in the crank case through blow-by. Those components need to be taken care of so that harmful deposits and abrasive particles are not left in the engine. The engine oil works with detergent and dispersant additives to solve this task. The detergent keeps the inside surfaces of the engine clean while the dispersants keeps the deposits in solution and stops particle flocculation. When particles forms that is big enough to be harmful, the engine oil filter is there to filtrate those particles away.

Combustion soot is significantly reduced when running CNG compared to traditional diesel combustion. That results in a reduced need of keep clean and clean up capability of the oil. Diesel engine oil would not be needed as there will be less clean up work. What is also important to keep in mind is the keep clean/clean up additive package of normal diesel engine oil is often not designed to work in temperatures that can be found in piston ring pack and turbo charger bearing houses of gas and petrol engines.

B1.1.6 Sealing

An important task for the oil is to assist in sealing the piston ring – cylinder liner area. Without oil in that area the task for the piston rings would be even more difficult. The engine oil viscosity is one important factor for this function. Other areas where the oil helps are in gaskets and stuffing boxes. As the basic engine set up in those areas will not be changed, lubrication oil demands will be the same for diesel, lean burn gas and stoichiometric gas engines.

B1.1.7 Corrosion Inhibitor

The engine oil coats internal engine parts to prevent surface rust on the inside of the engine which can be caused by blow-by products and water formed in combustion. It must also be capable of neutralizing the acids that are formed by combustion blow-by and oil oxidation at high temperatures. Basically all modern engine oils will manage to deliver the protection needed so there is no need for a special product in response to a lean burn or a stoichiometric gas engine set up.

B1.2 Additional Demands on Engine Oil

Engine oil will influence other engine functions as well. Some of the most obvious are, cold start function, exhaust aftertreatment system durability (due to oil consumption), fuel consumption and engine oil service intervals.

B1.2.1 Cold Start Function

The engine oils cold temperature viscosity and pumpability will influence the engines ability to start in cold temperatures. For gas engine applications the engine oil viscosity class 15W-40 is dominant. A few exceptions have been found where 10W-40 oil viscosity has been recommended as option.

The engine oil viscosity classes mentioned above are examples of multi grade engine oils. The viscosity for such oil is given as one cold viscosity class and one warm viscosity class. Taking 15W-40 as an example, the figure standing before the W (W stands for Winter) indicates the cold cranking viscosity. The lower the figures the easier the engine will be to crank at cold temperatures. The last figure, in this example 40, stands for the warm viscosity class. That viscosity is measured at 100°C, and can be looked at as the "working viscosity" of the oil.

In cold climate countries it is common to recommend 5W-X or 10W-X engine oil viscosities (X is here representing the warm viscosity recommendation that needs to be in line with the engine demands). When deciding on engine oil viscosity requirements it is important to keep the cold starting ability in mind.

It is worth to mention that an oil with a large span between cold cranking viscosity and (W) and its "working viscosity" (100°C) either have a high quality base oil mix or uses high amounts of viscosity improver additives.



B1.2.2 Exhaust Aftertreatment System Durability

Exhaust aftertreatment durability is a complex area. Many different parameters will influence the durability of the system. One of those is contamination of the equipment with additives coming from consumed engine oil. To reduce the engine oil influence you could either reduce the oil consumption or reduce the amount of additives in the oil that has negative effect on the aftertreatment equipment. On most markets, engine oils with reduced metallic content have been released. They are often referred to as "low ash" or "low SAP's" oils. Usually the reduction of metallic additives are somewhere in the area of 30%, making it comparable to an oil consumption reduction of the same magnitude.

B1.2.3 Fuel Consumption

The fuel consumption improvements that can be done with the help of engine oil choice are not to be neglected. Low viscosity class, low shear stability of the oil and/or friction modifiers in the oil are the most common measures for fuel consumption reduction through engine oil choice. Other important features to keep control of are the engine oil temperature. To high temperature will reduce engine oil life and also result in a risk of too low oil viscosity. Too low oil temperature will result in higher internal friction losses (increased fuel consumption) and a risk of accumulation of fuel and water in the oil.

B1.2.4 Engine Oil Service Intervals

Engine oil service intervals are influenced by a number of different parameters. The most obvious one is the oil quality, but other parameters such as oil volume, customer driving pattern and engine hardware design will also have a large impact on the possible service intervals.

Oil Volume

There have been presentations on a number of different conferences showing models on how the engine oil is degraded. One of the most useful is the simplified model that is built on the assumption that one part of the oil is "destroyed" with every combustion. With that model in mind it is easy to understand the oil volumes influence on oil service life. Of course the reality is not quite as simple, oil volume will influence the oil temperature and also the oil's ability to evaporate fuel and water, so an increased oil volume will not result in a constant extension of the service intervals.

Customer Driving Pattern

The customer driving pattern has a huge effect on engine oil degradation. Here there is a difference between a diesel engine and a Spark Ignited (SI) engine (CNG as well as petrol engines) in what part of the engine usage is responsible for the oil degradation. Historically high load points have been degrading the oil in a diesel engine due to soot, but with today's aftertreatment equipment on diesel engines fuel dilution during regeneration has become an equally important issue.

On charged² SI engines, oil degradation is not so much affected by soot load but other factors like oxidation, nitration, combustion water, fuel dilution and small amounts of carbonic acid, sulphuric and sulphurous acids. Driving behaviour will affect the degradation load the engine oil needs to handle. High load will result in

² Charged engines is an expression used for engines using any means of charging system to increase the air mass in the cylinder. Most common systems are turbo chargers and super charger.

high temperatures and blow-by while low load will reduce the thermal load, but put a strain on the engine in other ways. On the contrary to what is normally believed a smooth driving behaviour, for example city traffic, is one of the worst case scenarios for a charged SI engine. When doing engine oil validation it is important to try to cover as many driving patterns as possible.

Engine Hardware Design

Engine hardware design will have an impact on oil degradation. Different features in the engine will have different impact on the oil. Three features to examine are, hot spots, cold spots and "crank case environment".

Hot Spots

Turbo charger bearing houses, piston ring area and the area underneath the piston crown are three potential areas where engine oil oxidation can take place. If the circumstances are extreme we will even see engine oil cooking in those areas. Oxidation will use up some of the oil additives. In many oil formulations those additives also fulfils other tasks (anti nitration and anti-wear). Oxidation will also lead to increased engine oil viscosity. Finally the oil might even form black sludge. If we go as far as engine oil cooking we will end up with cylinder liner seizures (cooking in piston ring groves) and turbo charger break down (cooking in bearing area on turbine side).

Cold Spots

Cold spots can also cause problems, especially if they affect the crank case ventilation system. Problem with cold spots is that they will condensate water. Combustion of CNG produces roughly twice the amount of water than the one generated in petrol and diesel combustion. Therefore the task of evacuating the water that travels down into the crank case as part of the blow-by gases is higher than on petrol and diesel engines. As engine oil has limited ability to carry water there is a risk of corrosion inside the engine. Also the oil degradation can speed up with the presence of water. One other "classic" problem is white sludge³, for instance on the oil filler cap or on the inside of the cam cover. As coolant water leakage also demonstrates itself in a similar way it is important to minimise the risk of white sludge due to combustion water trapped in the crank case as that white sludge might be mistaken for the much more severe white sludge coming from coolant system leakage.

Crank Case Environment

The crank case environment will also have a large effect on the engine oil degradation speed. Here the amount, and content, of the blow-by gases plays an important role. All engines have blow-by, and all blow-by contains reactive components, that will degrade the engine oil. Unburnt fuel, partly oxidised fuel, water, particles and nitrogen oxides will all degrade the oil. The measures that can be taken to decrease this engine oil degradation path way is to minimise the engine blow-by, and/or to design a crank case ventilation system that improves the crank

³ White sludge, a white – beige deposit with a consistency ranging between foam and paste. Formed by oil mixing with water. When occurring it is often found on cold areas in the engine where the water-oil emulsion will condensate, for instance oil filler caps and inside cam covers. It mst often occurs in engines run for short periods, i.e. not reaching working temperature of the oil. Combustion water entering the crankcase as part of the blow-by will then not be evaporated and vented away through the crankcase ventilation system but instead become accumulated in the crankcase and mix with the oil to form an emulsion.



case environment by ventilating away the unburnt fuel, water and, above all, the nitrogen oxides.

B1.3 Additive Technology, Cleaning and Cooling

In diesel engines the main contributor to the depletion of oil life has historically been soot. Engine oils have increased their ability to control soot, and figures of about 7% in heavy duty engine oils is today a quite common threshold for oil change. With the introduction of exhaust aftertreatment systems on diesel engines, fuel dilution into the engine oil has become an issue. Now that is as important as soot as a threshold for engine oil change.

CNG engines will not be producing any soot levels that causes concern. Also the fuel dilution issue that can occur in both diesel and petrol engines is not a problem in CNG engines.

However, the other classical petrol engine oil degradation parameters will be relevant and the engine oil additive package needs to be designed to take care of them. Compared to a dedicated diesel engine oil formulation, detergent and dispersant levels can be reduced while oxidation and nitration stability needs to be improved.

It comes down to: Ash content vs. oxidation/nitration stability or detergency/ dispersancy. Some OEM's are afraid of ash levels in oil. One reason for the fear of high sulphate ash levels might come from bad experience from highly specified diesel engine oils used in CNG engines. Some dispersants and detergents in diesel engine oil is not designed for high temperatures and can, if used in petrol or CNG engines, cause deposits and wear. So it is not the level of sulphated ash in particular that leads to problems but what kind of ash containing additives that is used. As an example we can look at normal petrol engine oil formulations where the sulphated ash content is much higher than what is usually found in CNG engine oils, but they still works fine in highly tuned petrol engines with or without turbo.

B1.4 Engine Oil Quality Specifications

API and ACEA hold the most commonly used engine oil specifications. Both of those organisations have specifications covering heavy duty diesel engines and passenger car gasoline engines. ACEA has a group of standards covering passenger car diesel engines while API does not have specific passenger car diesel engine specification. Neither API nor ACEA have any specific heavy duty CNG engine oil specifications and that might be one of the main reasons why there are a number of different OEM specific CNG engine oil specifications on the market.

Examples:

- Cummins CES 20074
- IVECO 18-1809, NG1
- Volvo CNG
- MAN M3271-1, and more

As discussed previously, a CNG engine oil would be similar to a petrol engine oil formulation, but still very few of the dedicated CNG engine oils available on the market meets API or ACEA passenger car (petrol) specifications. The reason for

this might me numerous, but two possible reasons come into mind immediately. Either the CNG engine oils simply do not have the right quality to meet the passenger car (petrol) requirements, or the extra cost that comes with an official API or ACEA approval have not been deemed as beneficial for the sales and therefore the oil and additive companies has opted for not applying for API and ACEA approval.

B1.5 Fuel Quality Impact on Engine Oil Degradation

It is easy to underestimate the effect the fuel quality have on engine oil degradation. This connection between engine oil degradation speed and fuel quality is true for petrol as well as diesel powered engines and has the potential to be even more relevant for CNG engines.

CNG fuel quality differs greatly and large parts of the world still do not have automotive fuel specifications covering CNG and other methane rich gases for use in an internal combustion engine.

Some of the fuel parameters have direct impact on the engine oil degradation speed.

In general, methane combustion produces very low amounts of particles and reactive combustion components and the corresponding need for detergents and dispersants would be low. However, CNG fuels are a mixture, and the heavier hydrocarbons will result in an increase of both particles and reactive combustion components that would increase the need for detergents and dispersants.

The need for the cleaning and keep clean function of the engine oil is very much down to the purity of the CNG fuel. Depending on the fuel quality a rough recommendation for detergents and dispersants levels, reported as Total Base Number (TBN) value in unused oil, would be:

- A high quality natural gas fuel would not require high detergency and dispersancy ability of the oil. The only problem would be compressor oil contaminating the gas. Roughly 5 would be a recommended TBN.
- A natural gas with lower methane levels, and also most biogas qualities, would require a slightly higher TBN (~5.5 to 8). (due to more reactive HC in the blow-by)
- Landfill gases and other gas sources where there might be unusual contaminations (contaminations often not covered by standards) would require a TBN of roughly 10.

The values above refers back to the engine oil formulations that was used, and still are used in many oils, before the low ash oils appeared on the market. With the low ash formulations the importance of TBN has diminished and today the rule of thumb presented above is not valid for low ash formulations. Instead Total Acid Number (TAN) has grown in importance.



B1.6 Crankcase Ventilation Systems Influence on Engine Oil Degradation

When going from a Euro III diesel engine to a Euro V – VI CNG engine one of the new tasks to take care of is the crankcase ventilation gases. The ventilation gases needs to be incorporated in the engines total emissions.

One way of dealing with this is the introduction of a closed crankcase ventilation system (CCV). This system has the potential to, on one hand, totally destroy the engine oil life or, on the other hand, can extend the possible service interval of the oil⁴.

The CCV system has the task of evacuating the blow-by gases from the crankcase. Additional tasks are to evacuate fuel diluted into the oil (mainly for engines with liquid fuels) and to evacuate combustion water from the crankcase. If fuel and water is not evacuated the expected engine oil service interval will be shortened. We also mentioned the ability of the blow-by gases to degrade engine oil. The reactive components in the blow-by cause nitration and will also produce acids that will reduce TBN and increase TAN. Therefore it is beneficial with better evacuation of blow-by gases. One way of improving the ability to evacuate the crankcase is to introduce a "fresh air crank case ventilation system". It is simply a CCV system that introduces "fresh air" into the crankcase to dilute the blow-by and by that reduces the concentration of NOx and other oil degrading components in the crankcase.

B1.7 Engine oil consumption

One area that will differ when running an engine originating from a diesel engine design as a spark ignited gas engine is the throttling of the fuel inlet system. This will influence many things, but one thing worth to mention in this chapter is the possible influence on engine oil consumption.

The throttling will change the conditions for the crank case ventilation, the piston ring pack – cylinder liner, the inlet valve – valve stem and for the turbocharger bearing and sealing system.

Those four affected areas are the areas determining engine oil consumption. Any change to these systems can affect the base engine oil consumption and a CNG conversion will affect all four.

⁴A crankcase ventilation system has one major task, to vent away the combustion gases that leaks past the piston and down into the crankcase. But how well that is done will also influence a number of other parameters.

In the ventilation system there will be a need of an oil separator so that too much engine oil does not escape from the crankcase and into the atmosphere (directly or through the combustion chamber). Depending on the "quality" of the oil separator different amounts of oil will still escape, and also different amounts of the combustion derived water will be kept in the crankcase. High amounts of water in the oil will reduce the oil life. So the design of the oil separator is important for the engine oil life and oil consumption.

Engine oil life also depends on the crankcase environment. If there are high levels of combustion gases in the crankcase the oil degradation will be faster, one example is nitration that is driven by nitrogen oxides degrading the oil. A well designed crankcase ventilation system can improve the crankcase environment and by that increase the engine oil life while, on the other hand, a not so good designed crankcase ventilation system can reduce the engine oil life.

B1.8 Conclusion: Engine Oil Quality Demands

Engine oil recommendation will depend on fuel qualities, availability of oil qualities, targeted service intervals, service workshops ability to cope with multiple oil requirements and price sensitivity of customers.

When an OEM specifies the recommended oil qualities some of those parameters are clearly within the engine development team's responsibility. Other parameters need to be discussed and designed together with aftermarket and marketing in mind.

As a customer it is important to observe the specific engine oil quality recommendation. It might look as it is a "normal" diesel engine oil, but if the OEM recommends a specific engine oil quality they usually have a good reason.

However, some general rules of thumb are still worth discussing:

- Do not use just any standard diesel engine oil in gas engines.
- Choose from available gas engine oils or, if needed, opt for gasoline engine oils.
- It is recommended to use minimum Group II or Group III base oils

The thermal load on the oil will be higher with stoichiometric than with Lean burn gas engine set ups. It is then tempting to conclude that stoichiometric engines would need "better" engine oil. However, the engine oil demands is more based upon the fuel quality used, the base engine design (especially crankcase ventilation system quality) and the customer driving pattern than the specific difference in engine oil thermal load between a lean burn and a stoichiometric CNG engine. Because of this the engine oil recommendation would basically be the same regardless of choice of gas engine technology.

Availability of engine oil qualities, fuel qualities and expectations on service intervals will differ between different markets. The end result might be that different markets require different engine oil recommendations and consequently different service intervals.



Appendix B2. Engine oil formulation and consumption: Influence on particle emissions – A literature study with comments

In this appendix we will try to summarize the findings from a small literature study on particle number (PN) emissions from natural gas engines. The aim will be to compare the particle number and particle size distribution from different natural gas engines to engines using conventional liquid fuels. On top of that possible root causes for particle emissions from gas engines will be evaluated.

Historically particles and black smoke was considered to be one and the same. Analysing methods, Bosch number = darkening of filter paper and particle mass, was considered to evaluate the particle emissions. The smaller than 100 nm particulate size range is much smaller than the wavelengths of visible light, and so particulates of this size tend to have little light scattering power, and are often considered 'invisible'.

The importance of particle size distribution and particle number emissions was highlighted in the late 1990's and early part of the new millennium with reports on the effect that ultra-fine particles can have on human health [1, 2, 3, and 4].

From the different published papers that we have evaluated it stands clear that historically the notion was that a natural gas engine was cleaner than a diesel engine in every aspect. Even though benefits with using natural gas engines compared to the average diesel engine of that time are quite obvious some of the superiority claims made earlier can today seem a bit naive.

Still, some of the researchers were not that naive and the first couple of papers that is used in this literature study originate from the late 1990's.

To present a picture of the current knowledge you will find beneath extracts from 16 papers with, for the subject, interesting findings. But first a summary of those findings, pinpointing off the agreements and disparities and highlighting the areas where there are gaps in knowledge.

B2.1 Particles and their origin

Particles can originate from burning of fuel, from engine oil entering the inlet, combustion or exhaust system or particles originating from engine wear. The different papers under discussion have shown that all three pathways contribute to CNG engine particle emissions.

Also the particle size of CNG engine emissions seems to have a number maximum or peak in the 7 – 10 nm range. Some papers have found a second number peak for larger size particles, thus yielding a bimodal size distribution, often in the size range of 60 - 80 nm. In one paper the particle size analyser was able to detect particles at an even smaller particle size range. A particle number peak was found in the particle size range of 2 - 3 nm when testing a small bi-fuel passenger car. This size range of particles matches findings from tests with gas burners that also reported a number peak in the 1.5 - 3 nm size range, see chapter B2.12. In another paper the author also highlights the possibility that a particle size peak smaller than 7 - 10 nm exists from the tested gas engine, see chapter B2.4.1. Also in the paper quoted in chapter B2.8 there is evidence that a particle size peak is present below 10 nm. Most papers have reported that the particle size distribution also being lognormal, meaning that the median particle size is smaller than the average particle size.

The paper summarized in chapter B2.6 illustrates that the behaviour of carbon particles produced by an ethylene diffusion flame has different kinetic oxidation behaviour than carbon particles in diesel engine exhausts. The reason is put down to the presence of metallic particles in the diesel engine exhausts.

In many papers engine oil consumption has been highlighted as one of the sources for particle emissions. Particles originate from both the base oil and from the ash containing additives. Earlier measurements from SI engines suggest that approximately 10% of the PM emissions originate from the engine oil ash content.

As emission legislations get more stringent the oil consumption may make up more and more of the particle emissions allowed. An estimation made is that up to 40% of the PM emissions will be engine oil contributed in the 2007 American emission standard.

An interesting finding was that in some cases a small volume of engine oil combustion could reduce the PM emissions, but increase the PN, see chapter B2.6. It is clear that ash in the oil will produce particles if oil is consumed. With the upcoming more stringent emission regulations, engine oil consumption must be taken more seriously, perhaps forcing a redesign of the piston ring pack on heavy duty gas engines.

Particle number emissions corresponding to engine oil consumption is most prominent at low load while combustion derived particles becomes more dominant at high load, see chapters B2.3, B2.4 and B2.7.

The engine oil itself will also have an influence. Different oil formulations can influence the emissions. In this case it has been argued that PAH emission, as well as particle emissions, can be influenced by the oil formulation, see B2.6 and B2.7.

Particle emissions from engines running on methane rich fuels (CNG, CBG, LNG etc.) are relevant, if considering number emissions rather than mass. If we look at particle mass emissions the gas engines gives a radical reduction in emissions compared to a diesel engine. But if we look at particle number emissions the difference between the different engine technologies and fuels are not that great. It is even so that in some cases a diesel engine with DPF can have lower particle number emissions than a gas engine without after treatment.

It is risky to compare results in between the different papers used in this literature study, but three of them have run NEDC cycle with light duty vehicles and the results on particle numbers are quite similar, in the range of 1×10^{11} to 4.2×10^{11} . That is less than what is regulated for a DI gasoline engine (6.0 $\times 10^{11}$ particles/km).

On the heavy duty side there is currently no PN requirement for gas engines, but comparing the results from some of the tests reported in the examined papers we can see that in many instances the indication is that it is not clear that the current (pre Euro VI) CNG engines will meet the diesel engine requirements in the Euro VI regulations (WHSC = 8.0×10^{11} and WHTC = 6.0×10^{11} particles/kWh).

B2.1.1 Conclusion and recommendations

Particle number emissions are still a concern even when converting engines to run on CNG. Particle numbers from a CNG engine are in the same order of magnitude as gasoline engines and diesel engines with DPF. It is also interesting to see the strong influence that engine oil consumption and engine oil formulation can have on both particulate mass and particulate number emissions.



It has also been shown that emissions from gas engines will differ depending on the gas quality they are using leading to the conclusion that the current emission certification procedure with multiple gas qualities to be run during the certification process makes sense.

One area that might still need to be investigated is the particle number measurement techniques and the way that the measuring equipment is mounted onto the test object. Particles have a tendency to agglomerate and condensate depending on the dilution method, gas temperature, gas flow and pipe length used.

In heavily polluted areas it can still be sensible to convert part of the vehicle fleet to gas, but it needs to be done in a correct way. All converted engines do not deliver the emission reductions hoped for, in some instances they can even increase emissions.

With fully implemented Euro VI emission requirements for both CI and SI engines both combustion technologies will be competing on a more level playing field (some small differences still exists). Then, perhaps, the full emission reduction potential of gas engines can be shown. But to be able to do that a fit for purpose automotive methane fuel standard needs to be implemented both in Europe and in the rest of the world. And the market fuel quality needs to fulfil those standards.

B2.1.2 Possible next steps

The goal of this literature study was to compare particle number and size distribution from natural gas engines, and to evaluate the engine oil influence on particle emissions from gas engines.

Particle size distribution in the exhausts from natural gas engines appear to be leaning towards smaller size particles compared to engines using liquid fuels (both SI and CI). As highlighted in some of the summarized papers there is a risk that gas engines have at least one particle number peak below the particle size distribution range measured today.

Papers summarized below have also touched upon the engine oil influence on particle emissions, but none have really managed to pinpoint the engine oil formulation's influence on particle emissions. Many of the findings have either been performed on petrol or diesel engines. Others using gas engines have often referred to engine oil formulations not really relevant for gas engines of today.

One possible next step could be to evaluate known and estimated health effects from particles in a size range below 10 nm, and depending on the findings proceed with particle number and distribution measurements on those size ranges that will have been shown to have large potential health effects.

Another possible next steep could be to set up an engine test project focusing on the influence of different base oil formulations and additive packages on particle mass and particle size distribution from gas engines.

B2.2 An Investigation to Determine the Exhaust Particulate Size Distributions for Diesel, Petrol, and Compressed Natural Gas Fuelled Vehicles

In the SAE 961085 paper (Greenwood et al., 1996) particle emissions from passenger cars was evaluated. The test program consisted of two diesel vehicles (one NA, one turbo), three petrol vehicles (all PFI and with TWC) and two CNG (one bifuel and one dedicated CNG). No information on the vehicles emission class was found in the paper, but as the paper dates from 1996 we can expect that the vehicles meet Euro 1 (1993) or at best Euro 2 (1996) emission standards. The particle size measurements covered particle sizes from 15 nm up to 700 nm. The particle size distribution across all three types of vehicles was "lognor-mal", meaning that the peak maximum particle number diameter was much lower than the mean diameter.

Not surprisingly the particle size distribution differed between the different technologies. Diesel having the largest size of the peak particles (NA: 70 - 90 nm, turbo 50 - 60 nm), followed by petrol (20 - 30 nm) and CNG (approximately 22 nm). Also the same order can be seen between the technologies when comparing particle numbers.

Vehicle	At idle	At medium load	At high load
	[#/nm³]	[#/nm³]	[#/nm ³]
IDI norm asp diesel	1,35 x10 ¹²	3,26 x 10 ¹²	3,02 x 10 ¹³
Petrol 3	1,47 x 10 ¹⁰	1,25 x 10 ¹²	1,78 x 10 ¹²
Bi-fuel CNG	3,58 x 10 ⁹	1,81 x 10 ¹¹	5,92 x 10 ¹¹

The author's conclusion was that all three types of vehicles produces significant numbers of particulates, especially at high road loads.

B2.3 Elemental composition of combustion emissions from spark ignition vehicles

In the paper "Elemental composition of combustion emissions from spark ignition vehicles" (Ristovski et al., 1999) the particle number was not examined and the vehicles used had petrol engines. The reason for refereeing to this paper is the evaluation method used to determining the origin of the particles measured.

The five-mode steady state cycle was adopted and modified from the US EPA Final ASM Test Procedure Documentation. In this case the result is five vehicle sped/loads have been investigated, idle, 40 km/h, 60 km/h, 80 km/h and 120 km/h. Particles smaller than 1 µm was collected onto filters and analysed.

A bulk aerosol chemistry study has been performed. The concentrations of the following trace elements were measured: Boron, Calcium, Titanium, Chromium, Manganese, Iron, Cobalt, Nickel, Copper, Zinc, Cadmium, Barium, and Lead. The petrol particles have been found to be spherical. The most abundant elements were Fe, Ca, and Zn. Ca and Zn have their origin from the additives in lubricating oil while Fe must come from engine wear (directly into the exhaust gases from the cylinders or carried into the combustion/exhaust via the oil consumption). It is interesting to note that although particle concentrations increased with 3 orders of magnitude from 40 to 120 km/h, emissions of metals did not significantly increase. This would indicate that most of the particle emission at higher loads comes from the unburned fuel and consists mainly of carbon either in elemental or organic form.

Iron particles were observed in very small size bellow 20nm. Calcium and Zinc, as the most abundant elements besides Fe, were observed in larger size around 100nm. It is interesting to note that particles of pure Zn were rarely observed. Zn was usually in larger particles in a mixture with other elements.

Similar to the findings in the SAE 2006-01-3397 paper, it can be seen that engine oil consumption contributes to the particle emissions.



B2.4 Particle Emissions from Natural Gas Engines

In the paper "Particle Emissions from Natural Gas Engines" (Ristovski et al., 2000) emissions from two large CNG SI engines have been investigated. The first engine was of an older type (Waukesha 5115GL Constant Torque Engine) without any emission control, while the second (Deutz, model TBG 616 K) was an ECU controlled engine with emission controls.

As no standards for emission measurements of this type of engine were available at the time, it was decided to do the measurements of size distribution and concentration of particles at five power settings of the engine: 35%, 50%, 65%, 80% and 100% of full power. Three consecutive samples were taken at each setting with the SMPS and, five with the APS, see below for explanation. The engines were cycled up from the lowest power setting to the highest and then down again to allow repeat sampling for the same power setting.

Two different instruments were used for particle size measurements. Particle size distributions were measured by the TSI Model 3934 Scanning Mobility Particle Sizer (SMPS) in the size range 0.015-0.7 μ m, and by the TSI Model 3030 Aerodynamic Particle Sizer (APS) in the size range 0.5-30 μ m.

Samples for elemental composition of particles were collected on filters directly from the output of the electrostatic classifier of the SMPS. Concentrations of the following trace elements were measured: Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Zn, As, Si, Mo, Cd, Sb, Ba, Hg, and Pb.

Polycyclic Aromatic Hydrocarbons were collected by bubbling the undiluted exhaust sample through a solution of chloroform solvent. The samples were then analysed by a Gas Chromatography Mass Spectrometer.

B2.4.1 Particle emissions

Particle emissions in the APS range (0.5-30 μ m) were very low, at a level below 2 particles/cm³ which is comparable with an average ambient concentration.

The particle number emissions measured in the $0.015 - 0.7 \mu m$ range was in the order of 10^6 to 10^7 particles/cm³. When looking at the analysing data collected with the SMPS equipment ($0.015 - 0.7 \mu m$) an odd behaviour is however observed, especially for the Deutz engine. The particle number results on "35% power" and "50% power" differs quite drastically from the measurement done when going through the power settings with increasing power, to the repeats when going stepwise down through the power settings. The particle numbers are two magnitudes higher on "the way up".

The author's explanation was that this was due to the decrease in particle diameter below the measuring range of the SMPS, as a result of which a certain number of particles were not measured. The instrument registered the upper part of the particle distribution, while the majority of the emitted particles were below $0.015 \mu m$.

B2.4.2 Metallic ash

The metallic content on the collection filters was analysed. Similarly as reported in literature (5), the most abundant element was calcium which is a component of detergent additives. Significant amounts of barium, which is also present in lubricating oils (5), were detected, although concentrations of barium were about an order

of magnitude smaller than of calcium. Concentrations of other detectable elements, including titanium, cadmium, antimony and lead, were about two to three orders of magnitude lower than the one of calcium.

Analysis of metallic ash emissions showed that, higher emissions were observed for the lower power modes (both 35% up and down), for all detectable elements. Metallic ash particles are formed through homogeneous nucleation of metals, which are introduced into the combustion chamber through the engine lubricating oil (5; 6). Recent measurements on spark ignition engines showed that a significant fraction, approximately 10%, of the total exhaust particulate mass emissions is composed of solid metallic ash (5). These particles present nucleation centres for further growth.

The emissions of metallic ash measured in this study can be compared with particulate emissions in the submicrometer region (SMPS measurements). For the two modes at 35% power, the first one (increasing power) had a CMD of 0.025 μ m and a total particle concentration of 2.49×10⁶/cm³, while the second mode (decreasing power) had a CMD of 0.017 μ m, with the majority of particles below the measuring range of the instrumentation.

As metallic ash concentrations were the same, in the limit of error, for both modes, one can conclude that the metallic ash particles that were formed in this combustion process were in the ultrafine size range below 0.020 μ m. These particles presented nucleation centres for further growth of larger particles. The total number-weighted nucleation potential for these particles can be calculated from the measured mass concentration of metallic ash. Assuming that these metallic particles had a diameter of about 0.010 μ m, they would form particle concentration of which would be in the order of 10⁶ particles/cm³. This number corresponds to the total concentration measured in this experiment. Studies by Graskow, et al. (6) reported the total particle concentration calculated from engine oil consumption, to be of the order of 10⁹ particles/cm³. The calculations were based on the assumption that all metals compounds that are introduced into the combustion chamber, form particles that survive into the tailpipe.

B2.4.3 PAH

None of the organic gases which are often associated with the gas combustion process were detected, e.g., formaldehyde, acetaldehyde, and to a lesser extent, benzene and 1,3-butadiene. Instead, a large number of higher molecular weight compounds were identified as possibly characteristic of the emission gases. While the exact identification of these compounds was not possible, it appears that the majority of the compounds were esters attached to a benzene ring. Their sources can only be hypothesised, but it is likely, that they are the result of any oils used in the engine.

It is possible to calculate the relative proportions of the molecules for each power setting from the gas chromatograms. The organic molecules determined for the lower power settings, 35% up and 35% down, were found to represent only 1.5 and 8.3%, respectively, of the total concentration of the organic molecules, relative to the 100% power setting. The trend in emission of organic compounds was opposite to the trend in metallic ash emission. This indicates that the majority of particles emitted at lower power settings come from metals introduced through engine oil, while emissions at high power settings are mostly of organic origin.



B2.5 Performance and Emissions Evaluation of Compressed Natural Gas and Clean Diesel Buses at New York City's Metropolitan Transit Authority

In the SAE 2003-01-0300 paper (Lanni et al., 2003) unregulated emissions from diesel buses and CNG buses was analysed. Both the diesel and CNG buses tested in the program were chosen from the existing fleet at Mother Clara Hale depot in Manhattan. The diesel buses were 1999 40 foot Orion model V transit buses. The CNG buses were 1999 40 foot New Flyer CLF 40 transit buses. All of the buses had 1999 Detroit Diesel Series 50 engines. The diesel buses were equipped with a standard oxidation catalyst and either with or without a continuously regenerating diesel particulate trap (CRDPF). The CNG buses had no catalytic aftertreatment installed.

The vehicles underwent chassis dynamometer emissions testing. Each bus was tested with both the Central Business District (CBD) and New York Bus (NYB) cycles. Both are time based cycles operating for approximately 10 minutes each. The CBD cycle covers 2 miles for an average speed of 6 mph. The NYB cycle is more representative of actual operation in New York City, and covers 0.5 miles for an average speed of 1.5 mph.

Particle size distribution measurements were performed using two distinct instruments and methods. The Scanning Mobility Particle Sizer (SMPS), TSI Model # 3934, measures mobility diameter through the range 0.005 to 1 micron. It can produce a complete size distribution, or can measure one pre-selected size in real time. The second instrument used was the Electrical Low Pressure Impactor (ELPI), from Dekati Ltd. Finland, which measures aerodynamic diameter using an impactor. It has twelve stages, each covering a subset of the size range between 0.035 – 10 micron.

B2.5.1 Carbonyl emissions

The CNG buses generally produce more carbonyls, up to one order of magnitude higher than diesel buses. Furthermore, greater than 96% of the total carbonyl emissions from CNG buses were formaldehyde, in contrast to 60-65% formaldehyde for the diesel buses. Individual carbonyl emissions from CNG buses were actually 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. For the CRDPF bus tested, the reduction of carbonyls by the oxidation catalyst in the CRDPF was so efficient that most carbonyls were below the method detection limit. 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 [7].

B2.5.2 PAH/nPAH Emissions

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 as shown in Figure 6. This observation agrees with the higher PAH emission from CNG buses reported by others [7, 8]. If more data are found to support this observation, it may indicate the presence of a pathway leading to the elevated PAH levels in CNG bus exhaust due to possible engine oil burning or thermal synthesis during CNG combustion. Further analysis of the CNG fuel and engine oil may help indicate whether this is the case.

B2.5.3 Particle sizing comparison

Measurements of the particle size distribution performed during 30 mph steady state runs using the SMPS showed relatively little difference between CNG and Series 50 CRDPF buses. Number concentrations for all buses were of the same order of magnitude with the exception of those for CNG bus #854, which were lower. Observed modes for the distributions ranged from 10 to 30 nm, with an apparent shifting of the mode towards smaller particle diameters for the CNG buses. Measurements of the particle size distribution were also performed during CBD and NYB driving cycles using the SMPS and the ELPI.

The results were comparable for Series 50 CRDPF and CNG buses for particles in the 270 to 6800 nm size range. In the 30 to 180 nm range, average number concentrations from two of the CNG buses were lower than those from the CRDPF buses by up to an order of magnitude.

Compared to CRDPF buses, higher emission of carbonyl, benzene, toluene and PAHs from CNG buses without oxidation catalysts may contribute to the reported unfavourable toxic potentials from CNG buses. The reduction in PM of the CNG buses was approximately equal to that of the Series 50 CRDPF buses, i.e., a 90% reduction from Series 50 buses without CRDPF using 30 or 300 ppm sulphur fuel.

B2.6 The Influence of Engine Lubricating Oil on Diesel Nanoparticle Emissions and kinetics of Oxidation

In the SAE 2003-01-3179 paper (Jung et al., 2003) the engine oil and its additives influence on oxidation rate in a diesel engine combustion engine was investigated.

Although the oxidation rates of Diesel particles have been measured numerous times in the past the influence of engine oil metals on the kinetics of Diesel particle oxidation has previously been overlooked.

It is often assumed that the oxidation kinetics of carbon particles in Diesel exhaust is similar to the kinetics of carbon oxidation in diffusion flame studies [9, 10].

However, a recent study by Higgins et al. [11] showed that the kinetics of oxidation of carbon particles in Diesel exhaust were quite different from that of carbon particles from an ethylene diffusion flame, and from the Nagle and Strickland-Constable kinetic model [12]. Higgins et al [11] suggested this might be due to the presence of metal compounds in Diesel exhaust particles.

Engine oil is continuously consumed in the combustion chamber. Although oil consumption typically is only about 0.2 % of fuel consumption, it contributes disproportionally to the exhaust particulate matter. The combustion efficiency of engine oil is considerably lower than that of Diesel fuel and engine oil contains metals that form solid particles. Engine oil is composed of a base oil and an additive package. In general, the base oil is composed of petroleum-derived mineral oils, whereas the additive package is composed of various chemicals, including metal compounds (such as Ca, Mg, and Zn) [12]. Most of these contribute to engine oil ash.

The typical ash content of earlier "high ash" engine oil is about 1 %. A material balance based on 1 % engine oil ash content, 0.2 % relative engine oil consumption rate, 250 g/kWh fuel consumption rate and 0.13 g/kWh PM (Particulate Matter) (US PM standard for 2004) gives 0.005 g/kWh ash emissions. Under these conditions, the ash would constitute 4 % of the current PM, assuming that all of the ash appeared in the PM standard. Published results show up to 10 % ash by mass



[13]. For 2007, the PM standard drops to 0.013 g/kWh. If engine oil ash consumption is assumed to be at the same level as in the material balance above, it could constitute 40 % of the PM emissions. This suggests growing importance of the engine oil to PM emissions.

In some cases, engine oil is intentionally mixed with fuel to extend engine oil change intervals. An on-board continuous oil management system called CENTI-NELTM was developed by Cummins [19, 20]. The system takes a small amount of used oil out of the lubrication system and sends it to the fuel tank, where it is blended with Diesel fuel. At the same time, fresh oil is sent from a makeup tank to the engine, replacing the used oil. This system has been shown not to increase PM mass emissions.

In this study the Diesel fuel was dosed with engine oil (2 % by volume). This was done to intentionally increase the oil consumption rate and the loading of metal components from engine oil on the DPM with combustion, in order to better observe the effect of engine oil (specifically metal components from engine oil) on the oxidation of Diesel particles downstream of the engine and on the exhaust size distribution. The oxidation rate of 2 % engine oil dosed Diesel particles was compared to that of undosed Diesel particles measured in the previous study [11]. The size distribution was also compared with that for undosed Diesel particles.

The SMPS measures mobility equivalent particle diameter in the diameter range of 7 to 300 nm during a 4-minute (2 minutes up, 2 minutes down) scan time

Comparing tests run with the 2% doped fuel to tests run on the un-doped fuel the volume concentration of accumulation mode particles was reduced by nearly a factor of two while the total number increased by about a factor of 10, mainly as a result of the formation of a large nuclei mode. The same trend was observed by Kytö et al. [14] when they blended 2 % of engine oil with Diesel fuel. They discovered that the dosage of engine oil that gave the lowest particulate mass (meaning the reduction of accumulation mode particles), produced the highest number of particles (which means the appearance of large nuclei mode).

Calcium is the most abundant species present in the engine oil used in these tests, and is known to suppress the formation of carbonaceous Diesel particles [15] and to enhance their oxidation [15, 16] when it is blended with fuel. Miyamoto et al. [15] concluded that Ca and Ba are the most effective additives for enhancing oxidation and suppressing the formation of soot among the metallic additives (Ca, Ba, Mg, Fe, Ni, Mn, Cu) investigated. This study shows that metals in engine oil blended with fuel may play a similar role.

A somewhat surprising outcome of this study is the significant reduction of PM volume emissions (which are proportional to mass) with engine oil dosing. These experiments were done with clean engine oil. It would be interesting to examine the effects of aged engine oil, like that used in the CENTINELTM system, and to further investigate the kinetics of oxidation at a lower level of engine oil dosing.

A disturbing result of engine oil dosing is the formation of a large solid nuclei mode. A number of studies [17, 18] suggest that very small particles, especially solid particles, may be more toxic than larger ones.

B2.7 Particle Emissions of a TDI-Engine with Different Lubrication Oils

In the SAE 2005-01-1100 paper (Czerwinski et al., 2005) the goal was to determine how different engine oils influenced the particulate emissions and the contribution of oil to the total particle emission.

Tests were performed with three commercial oils from Lubrizol (GB). Additionally the effect of three further oils was investigated.

For the tracer study the tests were repeated with two doped lubrication oils containing a mixture of the elements Pb, Cd, Ba and Sb. The oil-based solution was added in a 1:58 ratio to the lubrication oil.

In this study a modern VW TDI 1.9 L engine was used for the nanoparticle investigations.

Ultra low sulphur (ULSD) Diesel fuel (EN590 - S < 10 ppm) was used for all tests.

Following conclusions can be pointed out:

- The oxidation catalyst reduces the nanoparticle emissions due to the diffusion-deposition and oxidation of the soluble organic fraction (SOF).
- Particle emissions differed from approximately 5x10⁸ to 1,5x10⁹ [part/cm³] simply by using different engine oil.
- Reduced sulphur and lower additive content of the lubrication oil don't necessarily reduce the particle emission of the engine.

The complex influences of oil quality on the particle emissions offer several open questions for further research.

The results of oil tracing show, that the engine oil has a considerable contribution to the PM, particularly at the part load operating conditions of the engine.

B2.8 Evaluation of Regulated Materials and Ultra Fine Particle Emission from Trial Production of Heavy-Duty CNG Engine

In the SAE 2006-01-3397 paper (Tonegawa et al., 2006) the connection between engine oil consumption and particle number emissions is demonstrated in a simple way when particle emissions are shown before and after an engine oil consumption improvement was done.

The tests were performed on a prototype CNG engine origination from a Nissan inline 6 cyl diesel engine with 13,1 l displacement (GE13TA). The test was run using the Japanese JE05 heavy duty on-road cycle.

Before the measures to reduce the engine oil consumption the particle size distribution was bimodal with a first peak at approximately 7 nm and a second peak at 70 - 80 nm. The reduction in oil consumption resulted in a strong reduction of the second particle number peak, also the first peak was reduced but was still quite high. The authors concluded that quite a large portion of the particle emissions from a CNG engine can be contributed to engine oil consumption. It was also highlighted that the particle measuring technique was important, for instance they saw a strong influence on agglomeration and condensation depending on the dilution method used.



B2.9 Particle Characterisation of Modern CNG, Gasoline and Diesel Passenger Cars

In the SAE 2007-24-0123 paper (Schreiber et al., 2007) tailpipe particle measurements was investigated using Euro-4 passenger cars. The investigation included compressed natural gas fuelled (bifuel vehicles) spark-ignition vehicles, gasoline port-injection spark-ignition vehicles, gasoline direct injection spark-ignition vehicles as well as compression ignited diesel vehicles with and without particle filters. All vehicles were newer than 7 years and had mileages lower than 100,000 km.

The reported results are mainly based on measurements during the New European Driving Cycle (NEDC), and the real-world Common Artemis Driving Cycle (CADC). Besides the NEDC and CADC, constant speed driving tests were performed.

Particle number and soot mass was measured in the NEDC and CADC cycles. Particle number size distributions were recorded at constant vehicle speed operation.

A condensation particle counter (TSI INC., CPC3022A) was used for particle number evaluation (NEDC, CADC). This instrument has single particle detection and a 50% counting efficiency for 7nm diameter particles.

During constant driving test measurements a SMPS consisting of a differential mobility analyser (DMA, model 3071, TSI Inc.) and a condensation particle counter (CPC, model 3025, TSI Inc.), size range: 10-430 nm, was also used.

As expected, the measured particle emissions from diesel powered cars without a particle filter were the highest. Lower by more than one order of magnitude was the particulate emission of the gasoline direct injected vehicles.

These emissions were significantly higher than the corresponding ones from conventional gasoline MPFI cars. Particle emissions of the CNG and the diesel DPF-equipped vehicles are clearly the lowest.

Average particle number values from the NEDC cycle can be seen in the table below.

Vehicle type	CNG	MPFI SI	DI SI	Diesel w/o DPF	Diesel with DPF (after regen)	Diesel with DPF (loaded)
Particle number	3,8x10 ¹¹	8,5x10 ¹¹	2,7x10 ¹²	4,8x10 ¹³	5,6x10 ¹¹	2,7x10 ¹¹

At typical urban and extra urban driving conditions CNG and diesel fuelled DPF equipped vehicles have the lowest particle number emissions. Particle number emissions of MPI gasoline vehicles are, depending on the driving cycle, at least twice as high. DI gasoline vehicles have one order of magnitude higher particulate emissions, while diesel vehicles without DPFs emit even one order of magnitude more particles.

Emission patterns change at highway driving where CNG and gasoline fuelled vehicles has comparable particle emissions, one order of magnitude higher than diesel DPF equipped vehicles. Diesel vehicles without DPF have also at highway driving the highest particulate emissions (two orders of magnitude higher than their DPF equipped counterparts). The volatile component of the particles is during highway driving substantially higher than during urban or extra urban driving.

Specific for natural gas fuelled (bifuel) spark-ignition Euro-4 vehicles we have identified.

- Particles emissions lower than diesel fuelled DPF equipped passenger cars at low speed driving conditions (CADC urban phase).
- Particle emissions in the order of magnitude of the diesel Fuelled DPF vehicles provided the DPF was loaded and not freshly regenerated at extra urban road conditions (CADC extra urban phase).
- Disproportionately higher particle emissions similar to gasoline DI passenger cars at typical highway conditions (CADC highway phase).

The particle emissions at high speed conditions of the CNG vehicles consist of an accumulation mode, mostly consistent of soot, and a nucleation mode consisting of volatile aerosols. The particles emitted by the DI gasoline cars have similar structure.

The main reason identified for the high particle emission of the CNG vehicles at high vehicle speeds was the enrichment of the fuel-air mixture at higher loads.

B2.10 Regulated and Non-regulated Emissions from Euro 4 Alternative Fuel Vehicles

In the SAE 2008-01-1770 paper (Karlsson et al., 2008) one CNG/petrol and two ethanol/petrol passenger cars were tested using the NEDC and Artemis cycles.

Conventional petrol, compressed biogas (CBG), Swedish summer (E85) and winter ethanol fuels (E85W) were used. The tests were performed at 22°C and - 7°C.

Regulated emissions, particulate mass (PM), particle number (PN), CH₄, HC speciation and aldehyde were measured. The tested vehicles were standard Euro 4 production vehicles from three different manufacturers. All three vehicles were equipped with three-way catalysts (TWC).

It is worth to point out that the vehicle always started in petrol mode even if gas fuel was chosen as operating fuel. The CBG fuel led to significant CO and NO_x reductions and somewhat higher HC emissions compared to petrol fuel, the results being in accordance with published studies [19, 20]. The high HC emissions from CBG were due to that the main composition of the fuel was CH₄, a compound which is very difficult to oxidise in the catalytic converter. Indeed 74 % of the HC emissions from the tests with CBG consisted of CH₄. Although petrol fuel led to lower HC emissions, the 1,3-butadiene, benzene and toluene emissions were higher compared to CBG.

PN and PM measurements showed that petrol fuel led to higher PM and PN emission compared to CBG.

	Petrol NEDC	CGB NEDC	Petrol NEDC-7	CBG NEDC-7
PMP PN [#/km]	3x10 ¹¹	1x10 ¹¹	3x10 ¹²	7x10 ¹¹

The results were expected since petrol is a better precursor for particle formation than CBG. The petrol PN emissions were in the similar range as those reported for the Euro 3 and Euro 4 petrol vehicles.

B2.11 Particle and Gaseous Emissions from Compressed Natural Gas and Ultralow Sulphur Diesel-Fuelled Buses at Four Steady Engine Loads

In the paper "Particle and Gaseous Emissions from Compressed Natural Gas and Ultralow Sulphur Diesel-Fuelled Buses at Four Steady Engine Loads" (Jayaratne



et al., 2009) exhaust emissions from thirteen compressed natural gas (CNG) and nine ultralow sulphur diesel in-service transport buses were monitored on a chassis dynamometer.

Measurements were carried out at idle and at three steady engine loads of 25%, 50% and 100% of maximum power at a fixed speed of 60 km/h. Emission factors were estimated for particle mass and number, carbon dioxide and oxides of nitrogen for two types of CNG buses (Scania and MAN, compatible with Euro 2 and 3 emission standards, respectively) and two types of diesel buses (Volvo Pre-Euro/Euro1 and Mercedez OC500 Euro3).

Particle number analyses was done using 1/ a TSI 3022 condensation particle counter (CPC) that measured the total particle number concentration in the size range 5 nm to about 4 μ m, and 2/ a TSI Scanning Mobility Particle Sizer (SMPS) consisting of a TSI 3085 Electrostatic Classifier and a TSI 3025 CPC. The SMPS sample and sheath air flow rates were set to measure particle size distribution in the range 5-160 nm.

All emission factors increased with load. The median particle mass emission factor for the CNG buses was less than 1% of that from the diesel buses at all loads. However, the particle number emission factors did not show a statistically significant difference between buses operating on the two types of fuel. In this paper particle number emission factors are presented at four steady state engine loads for CNG buses. Median values ranged from the order of 10^{12} particles/min at idle to 10^{15} particles/km at full power. Most of the particles observed in the CNG emissions were in the nanoparticle size range and likely to be composed of volatile organic compounds. The CO₂ emission factors were about 20% to 30% greater for the diesel buses over the CNG buses, while the oxides of nitrogen emission factors did not show any difference due to the large variation between buses.

In the present study, the particle number emission rate/factors in each of the four modes were greater for the diesel buses over the CNG buses, but the difference was not statistically significant in three of the four modes.

B2.12 Ultrafine particle emission from combustion devices burning natural gas

In the paper "Ultrafine particle emission from combustion devices burning natural gas" (Minutolo et al., 2010) natural gas have been burned to mimic the use in kitchen stoves. This example of methane burning is not typical for burning in a combustion engine, but the result is relevant as it shows that even burning of relatively pure methane will produce particles and in this case the particles need to be formed during the combustion of the methane fuel. The particles formed are very small and the size distribution is bimodal. Depending on the air/fuel ratio and the boiler/stove design the peak particle size differs slightly, with one device showing a first peak at 3.5 nm and a second peak at around 15 nm and the second device having its two peaks at around 1.5 - 2 nm and 10 - 15 nm respectively. Especially the first particle peak (1.5 – 2 nm and 3.5 nm) is a particle size that often is missing from combustion engine emission measurements. The authors also tested an internal combustion engine (vehicle - Fiat Punto 60 Natural Power). Also there they found particle emission behaviour with particle size distribution of a bimodal behaviour. That was especially true for the "high speed tests" (corresponding to 120 km/h). Also in the internal combustion engine they found a peak in particle emissions at around the same size as found in the burner tests, in this case

around 3 nm. Their conclusion was that all devices they examined showed that methane and natural gas combustion, even when conducted in overall lean premixed conditions can produce particles with a mean size of 2 - 3 nm, and in a very high number concentration.

B2.13 Characterization of Toxicity as a function of Volatility of Ultrafine PM Emissions from Compressed Natural Gas Vehicles

In this paper from Centre for Alternative Fuels, Engines and Emissions (CAFEE) (2011) a CARB funded study focusing primarily on characterizing the toxicity of the volatile fraction of PM from advanced heavy-duty natural gas engines is reported. The objective of the study also includes characterizing the unregulated species of the exhaust together with number concentration and size distribution of ultrafine nanoparticle emissions. The work plan involved the chassis dynamometer testing of two heavy-duty natural gas transit buses that are compliant with the USEPA 2010 emissions regulation. Since the project was focused on investigating the effect of volatility on toxicity, the test procedure included a thermal denuder to remove the volatile component prior to collection on the filters.

Immediately downstream of the exhaust manifold all particles are mainly composed of carbonaceous particles (soot), and metallic ash. As the exhaust cools, soluble organic compounds (SOP) condense and adsorb to the surfaces of carbonaceous particles or nucleate to form nanoparticles (less than 50 nanometres). Further downstream, the agglomeration process decreases the number of particles, but does not affect the total mass. The nucleation process increases the number of particles, but does not affect the total mass. Kittelson and Abdul Khalek [21] found that the formation of nanoparticles was affected by the amount of soot in the exhaust stream. As the soot mass increased the number of nanoparticles decreased. Lack of carbonaceous soot particles and the presence of engine-oil (heavy-organics) based nanoparticles are very characteristic of CNG fuelled engine exhaust fumes.

It is well recognized that a considerable amount of the soluble organic fraction (SOF) is derived from the lubrication oil. Jung et al. [22] reported that lubrication oil additives, such as calcium, can increase the oxidation rate of the lubrication oil, resulting in a decrease of the SOF mass.

The amount of elemental carbon in gaseous fuelled engine exhaust is negligible compared to the organic carbon fraction. Ionic species and metals such as sulphates, phosphates, oxides of calcium, zinc, and magnesium are produced in the combustion chamber as a result of additives in the engine lubricating oil.

In a conventional diesel engine Essig et al. [23] found that a significant portion of the SOF in the exhaust PM came from the lubrication oil. In a typical engine exhaust PM the SOF is mostly made of poly-nuclear aromatic hydrocarbons (PAH).

Lubrication oil consumption has become an issue of major importance because of the tighter emission requirements imposed on modern engines. Experiments have shown that gas-fuelled engines have the same level of oil consumption as gasoline engines, operating under the same conditions [24].

The source of oil consumption in internal combustion can be divided into the following sources: piston-cylinder, valve train, crankcase ventilation system, and turbo charger. The most important is the piston-cylinder, which accounts for approximately 80 percent of the total oil consumption [25].



The exhaust of CNG vehicles are characterized by higher concentrations of organic carbon than elemental carbon. This fact could also be observed in the Elemental and Organic Carbon results of this study. The particle size distribution reflects a similar trend with a particle diameter peak of 10 nm with minimal variation in particle number throughout the cycle.

Both buses represented a similar consistent 10 nm peak over the UDDS cycle. A comparison of gravimetric PM from both buses revealed higher PM emissions from bus 2 and a similar trend is reflected in the PM size distribution of bus 2 with a well-defined accumulation mode with minimal variation in concentration over the transient UDDS cycle.

The steady state cycle resulted in very low particle numbers that were close to the detection limit of the instrument. The 10 nm peak observed in the transient cycle is also observed during the cruise cycle with the same order of magnitude in average particle number. However, a greater variability in particle concentration is also observed, due to measurement close to detection limits of the instrument.

B2.14 Emissions and Combustion Behavior of a Bi-Fuel Gasoline and Natural Gas Spark Ignition Engine

In the SAE 2011-24-0212 paper (Prati et al., 2011) a Fiat Panda 1.2 NP was tested using both CNG and petrol fuel. Emission factors were measured over the type-approval New European Driving Cycle (NEDC) and on real-world Common Artemis Driving Cycles (CADC).

Particulates were characterised as total particle number and size distribution. This measurement was carried out by an Electrical Low Pressure Impactor (Dekati ELPI) which is able to count particles with an aerodynamic diameter between 7 nm and 10 μ m, sizing them on 12 dimensional stages.

Particle Number (PN) emissions are higher for gasoline, with the exception of the Artemis Motorway. It is interesting to note that for gasoline the largest PN emissions occur during the NEDC, as a consequence of the cold start, whereas the high speed driving cycle (Artemis Motorway) brings larger emissions for CNG. Moreover by fuelling the vehicle with gasoline, PN emission, over NEDC, exceeds the Euro 5b standard limit (6×10^{11} particles/km). This limit for PN is currently prescribed in Europe only for diesel passenger cars.

B2.15 Influence of Different Natural Gas Blends on the Regulated Emissions, Particle Number and Size Distribution Emissions from a Refuse Hauler Truck

In the SAE 2012-01-1583 paper (Karavalakis et al., 2012) compared regulated and unregulated emissions from a 2001 Cummins 8.3L C Gas Plus lean burn spark ignited engine with an oxidation catalyst when running on 7 different methane rich fuels. The waste hauler was tested on each fuel on the William H. Martin (WHM) refuse truck driving cycle.

The William H. Martin Refuse Truck Cycle (RTC) was developed by West Virginia University to simulate waste hauler operation. The cycle consists of a transport segment, a curbside pickup segment, and a compaction segment.

Emissions measurements were obtained using CE-CERT's Mobile Emissions Laboratory (MEL) [17-18]. For all tests, standard emissions measurements of total hydrocarbons (THC), non-methane hydrocarbons (NMHC), methane (CH₄), carbon monoxide (CO), oxides of nitrogen (NO_x), carbon dioxide (CO₂), and particulate matter (PM), were measured.

Particle number counts were measured with the 3776 CPC (TSI) with a 2.5 nm cut point. Additionally, a nano scanning mobility particle sizer (nano-SMPS) was used to obtain information about how the particle size distributions differ between gas blends. The size range of the nano-SMPS was 4 to 70 nm in electrical mobility diameter.

Emissions of NO_x are a key concern for heavy-duty engines and can be affected by the methane number and the Wobbe number. Under the present test conditions, fuel composition had a significant influence on NO_x emissions. In general, NO_x emissions exhibited an increase as methane number decreased and Wobbe number increased during all three phases of the Refuse Truck Cycle.

THC emissions followed the same pattern in all three phases of the Refuse Truck Cycle. The gases containing higher hydrocarbons, such as CNG3, CNG4, CNG5, and CNG6, produced lower THC emissions than the baseline CNG1 and CNG2 and CNG7 gases. As discussed later in the paper, the THC emissions were predominantly CH_4 .

Compared to CNG1, CNG2, and CNG7, statistically significant reductions in PM emissions were found for CNG4, CNG5, and CNG6 gases for the waste hauler. CNG3 also showed statistically significant reductions relative to CNG1 and CNG2 gases, but not compared to CNG7.

The experimental results show that PN emissions followed the same pattern for all three phases of the Refuse Truck Cycle. For all three phases of the cycle, statistically significant reductions in PN emissions for CNG3 and CNG4 were found compared to the CNG1, CNG2, CNG5, CNG6, and CNG7 gases. The PN results followed the PM mass reductions for CNG3 and CNG4, but not for CNG5 and CNG6. Gases CNG3 and CNG4 have higher levels of higher hydrocarbons compared to CNG1, CNG2, and CNG7, but not compared to CNG5 and CNG6. It should be noted that PN emissions were approximately an order of magnitude higher for the transport phase compared to the curbside phase of this cycle. The higher PN emissions during the transport phase may be attributed to the higher load that the vehicles experienced during this mode of operation. The PN emissions from the curbside phase likely include a contribution from lubricating oil combustion during the idling periods of this mode.

The particle size distributions were averaged over 6 repeats for each fuel composition. Particle size distributions measured with a nano-SMPS showed a peak in the nucleation mode range, ranging from 7-12 nm for all gases. There was some indication of an up-going trend near the upper size limit of the nano-SMPS, which indicates the possible presence of an accumulation mode larger than 70 nm in the particle size distributions that could not be measured by the nano-SMPS.

Further measurements with instruments covering a larger size range are needed to investigate the accumulation mode particles in natural gas engines. It is also unclear whether the nucleation mode particles are mainly ash particles, or volatile particles composed of hydrocarbons originating from the lubricating oil, or combinations of both.



B2.16 Particle and gaseous emissions from individual diesel and CNG buses

In the paper "Particle and gaseous emissions from individual diesel and CNG buses" (Hallquist et al., 2013) size-resolved particle and gaseous emissions from 28 individual diesel-fuelled and 7 compressed natural gas (CNG)-fuelled buses, selected from an in-use bus fleet, were characterized for real-world dilution scenarios.

The method used was based on using CO₂ as a tracer of exhaust gas dilution. The particles were sampled by using an extractive sampling method and analysed with high time resolution instrumentation EEPS (10 Hz) and CO₂ with a non-dispersive infrared gas analyser (LI-840, LI-COR Inc. 1 Hz). The gaseous constituents (CO, HC and NO) were measured by using a remote sensing device (AccuScan RSD 3000, Environmental System Products Inc.). Nitrogen oxides, NO_x, were estimated from NO by using default NO₂ /NO_x ratios from the road vehicle emission model HBEFA3.1. The buses studied were diesel fuelled Euro III–V and CNG-fuelled Enhanced Environmentally Friendly Vehicles (EEVs) with different after-treatment, including selective catalytic reduction (SCR), exhaust gas recirculation (EGR) and with and without diesel particulate filter (DPF).

The primary driving mode applied in this study was accelerating mode. However, regarding the particle emissions also a constant speed mode was analysed. The investigated CNG buses emitted on average a higher number of particles but less mass compared to the diesel-fuelled buses. Emission factors for number of particles (EF_{PN}) were EF_{PN}, DPF = $4.4\pm3.5\times10^{14}$, EF_{PN}, no DPF = $2.1\pm1.0\times10^{15}$ and EF_{PN}, CNG = $7.8\pm5.7\times10^{15}$ [# / kg fuel]. In the accelerating mode, size resolved emission factors (EFs) showed unimodal number size distributions with peak diameters of 70–90 nm and 10 nm for diesel and CNG buses, respectively. For the constant speed mode, bimodal average number size distributions were obtained for the diesel buses with peak modes of ~10 nm and ~ 60 nm.

Emission factors for NO_x expressed as NO₂ equivalents for the diesel buses were on average 27±7 g/kg fuel and for the CNG buses 41±26 g/kg fuel. An antirelationship between EF_{NOx} and EF_{PM} was observed especially for buses with no DPF, and there was a positive relationship between EF_{PM} and EF_{CO}

B2.17 Comparison of Regulated Emissions and Particulate Matter of Gasoline/CNG Dual-Fuel Taxi Over New European Driving Cycle

In the SAE 2014-01-1467 paper (Wang et al., 2014) regulated emissions and particulate matter from five CNG/gasoline bi-fuel taxis⁵ over NEDC were reported. In addition, fuel economy of CNG and gasoline fuelling was quantitatively and qualitatively analyzed taking Beijing as an example. Finally, some implications and recommendations for future adoption of CNG vehicles were proposed.

For counting particle number purpose, a DEKATI ELPI (Electric Low Pressure Impactor) was adopted. ELPI was capable of capturing particles ranging from 7 nm to 10 μ m in diameter. Sample probe of the ELPI was affixed to the center of the dilution tunnel.

⁵ Hyundai Elantra, powered by 1.6L spark-ignited engines incorporated with 5-gear manual gearboxes. The taxis were registered in May and June, 2013. Please note that the author is not using the proper term for bifuel operation in the title.

It was found that the adoption of CNG decreased CO emissions during both cold- and warm-start conditions. Compared with gasoline-fuelled vehicles, the percentage of CO decrease was 58% and 22% in warm- and cold-start tests, respectively.

The use of CNG gave rise to evident increases in HC emissions in both cold- and warm-start tests. HC emissions emitted by CNG-fuelled taxis were 74% higher than gasoline-fuelled results in cold-start tests, and this percentage further climbed to 188% in warm-start tests.

In both the cold- and warm-start tests, CNG fuelled vehicles emitted higher NO_x emissions than gasoline-powered ones. In cold-start tests, CNG-fuelled vehicles emitted approximately 243% more NOx emissions. In warm-start conditions, gap between different fuelling regimes narrowed, NO_x from CNG-powered vehicles was only 29% higher. For both fuels, heavier NO_x emissions were yielded in the cold-start tests, extraordinarily so when CNG was used.

It could be observed that particle numbers ranged from 7.37×10^{11} to 1.07×10^{12} for the two fuels depending on start conditions. Generally, the number of particles captured in the gasoline-fuelled tests was 19-30% smaller than those in the CNG-fuelled tests. For both fuels, cold-start tests produced larger number of particles than the warm-start tests.

The results showed that using CNG as an alternative fuel was very helpful to curb CO and CO₂ emissions. However, changing to CNG would lead to higher emissions of HC and NO_x. From a number point of view, using CNG as an alternative to gasoline was not beneficial to abating particles, but limited by cold dilution method employed in the current research, conclusions on particulate matter should be further examined by introducing other method and technologies. At last, comparing to gasoline, CNG is cheap to use. This character would be the main incentive of implementing alternative fuels.



- B2.18 References for Appendix B2
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