Natural Gas / Methane Fuels: European Automotive Fuel Quality and Standardization Requirements

Erdgas / Methankraftstoffe: Europäische Kraftstoffqualitäts- und Normungsanforderungen für Kraftfahrahrzeuge

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Abstract: Due to highly volatile and mostly rising oil prices in the last decade, as well as potential reductions of automotive CO_2 emissions, natural gas (NG) usage as automotive fuel has grown significantly - in the form of compressed natural gas (CNG) or liquefied natural gas (LNG).

Furthermore downsizing of SI (spark ignition) engines for passenger cars is a mega trend in the automotive industry aiming at reduction of CO_2 emissions and fuel consumption while providing "fun to drive" at attractive cost of ownership. Downsizing offers increased potential when combined with alternative fuels like compressed natural gas (CNG).

A significant shift from oil based fuels to NG/methane as automotive fuel would increase NG demand considerably. If only half of European diesel/gasoline was replaced by NG, the NG market would increase by 35%. Full replacement would mean 70% increase. Thus automotive transportation has the potential to become the main customer for NG. Therefore NG standards need to be aligned with automotive requirements in order to ensure sufficient fuel quality at the retail stations. Otherwise the CO_2 reduction potential can be considerably limited by fuel quality issues.

Unfortunately the European standardization for NG as automotive fuel is not as advanced as the standardization for gasoline and diesel fuels.

For NG no final European standard has been issued so far. The NG fuel quality standardization is fragmented and carried out by the EU member states individually. European draft standards are under discussion and draft proposals have been issued recently. The responsibility for NG standardization is also very fragmented and handled by different standardization groups (CEN TC 234 and CEN TC 408). Currently three different standards are proposed. One standard is for gas grid quality (FprEN 16726:2015), another for bio-methane quality injected into the gas grid (prEN 16723-1:2014 E) and a third describes the automotive fuel quality at retail gas stations (prEN 16723-2:2014 E). Unfortunately all proposals contain different limits for critical components. This is very challenging, since automotive CNG is usually supplied by the gas grid. Critical deviations in the standards exist with regard to the sulfur content, the hydrogen content, the Lower Heating Value (LHV), the Wobbe Index (WI) and the Methane Number (describing the knock resistance of the fuel).

Furthermore some fundamental, required laboratory methods are not available yet, like methods for compressor oil and silicon determination.

A European automotive NG standard, well aligned with an appropriate future NG grid standard and an injection standard, is urgently required. Ideally identical parameters and limits as posted in this paper would be applied to all 3 of those standards.

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1 Prospects of NG as Automotive Fuel

Since Natural Gas (NG) reserves are estimated to be considerably longer lasting than oil reserves (200+ years vs. 70 years) [1], it is very likely that NG will be available for significantly lower cost than oil in the long term. Worldwide the NG price is already noticeably lower than the gasoline price (the average CNG automotive fuel retail price is 48% of the average gasoline price). [2]

The favourable fuel price conditions have already led to a worldwide vehicle population of 17.7 million Natural Gas Vehicles (NGVs). The annual growth rate between 2000 and 2012 was approximately 25 %. [2]

NG, which predominately consists of methane, has many combustion advantages vs. gasoline. Due to a more favourable C/H ratio than gasoline, NG combustion emits ~25% less CO_2 on a tank to wheel basis. Also feedgas emissions – in particular particle emissions – are significantly lower. Furthermore NG is very knock resistant and thus an ideal fuel for boosting and downsizing, which is not exploited by current NGVs on the European market. [3]

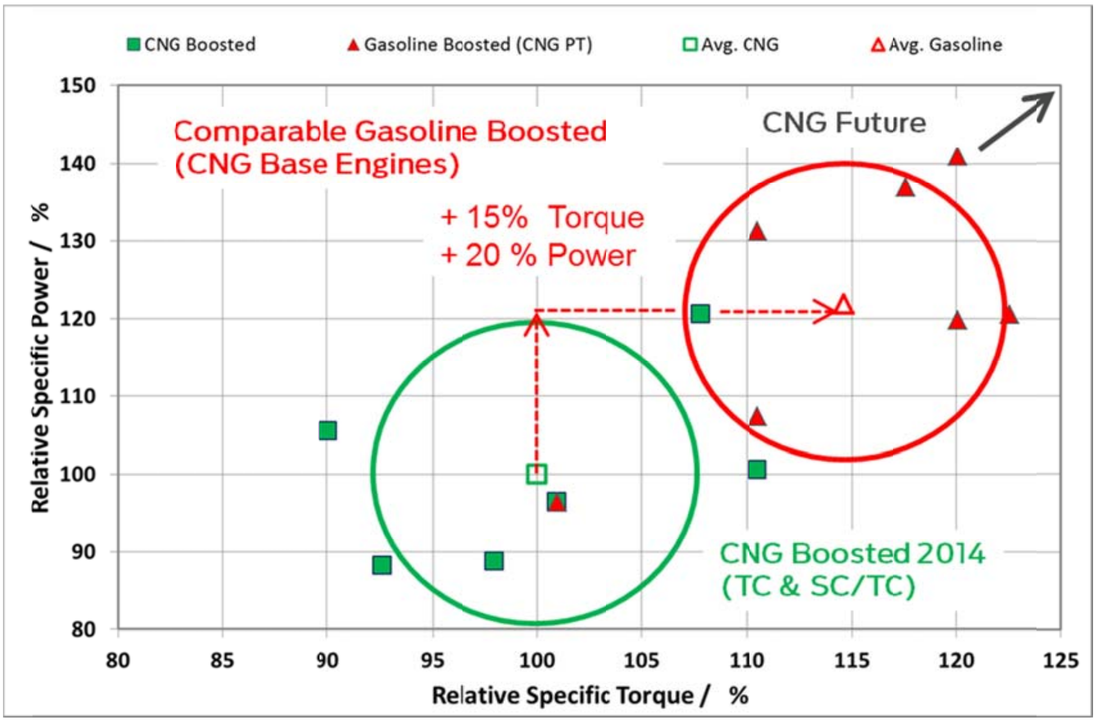


Fig 1: 2014 CNG vehicle market: CNG port fuel injection (CNG PFI) only [3]

As shown in Fig. 1 model year 2014 CNG passenger cars develop ~15% less specific torque and ~20% less specific power than the comparable gasoline engines in the same vehicles. The torque and power penalty is mainly caused by the reduced volumetric efficiency of CNG engines, which currently operate with port fuel injection systems (CNG displaces air).

With dedicated direct injection NG engines, designed for exploiting the high knock resistance of NG, an increased downsizing factor - even higher than on gasoline engines – can be achieved [3] [4]. The potential of dedicated, downsized CNG engines is currently thoroughly under investigation in the EU Horizon 2020 Project “GasOn”, where 4 automobile producers and more than 20 suppliers and research partners are going to demonstrate the efficiency potential of CNG as fuel until 2018.

NG is revealing further future sustainability potential as fuel as it can be blended with all types of renewable methane up to 100% blend rate (unlike ethanol/gasoline or bio-diesel/diesel). Renewable methane can be bio-methane or so called “power-to-gas methane” (PtG methane - methane produced out of renewable hydrogen). Bio-methane is one of the most land use efficient bio fuels with one of the best CO_2 avoidance factors. PtG methane is one of the most efficiently producible e-fuels.

All of those factors make methane a very interesting future automotive transportation fuel.

When NG consumption is compared with oil consumption worldwide (2987 Mtoe NG vs. 4130 Mtoe oil in 2012) or Europe wide (400 Mtoe NG vs. 611 Mtoe oil) [5] [6] – as shown in Fig. 2 - it becomes obvious that European oil consumption is approximately 1.5 times European NG consumption. That means a transition from oil based automotive fuels to NG/methane based automotive fuels would increase NG demand dramatically.

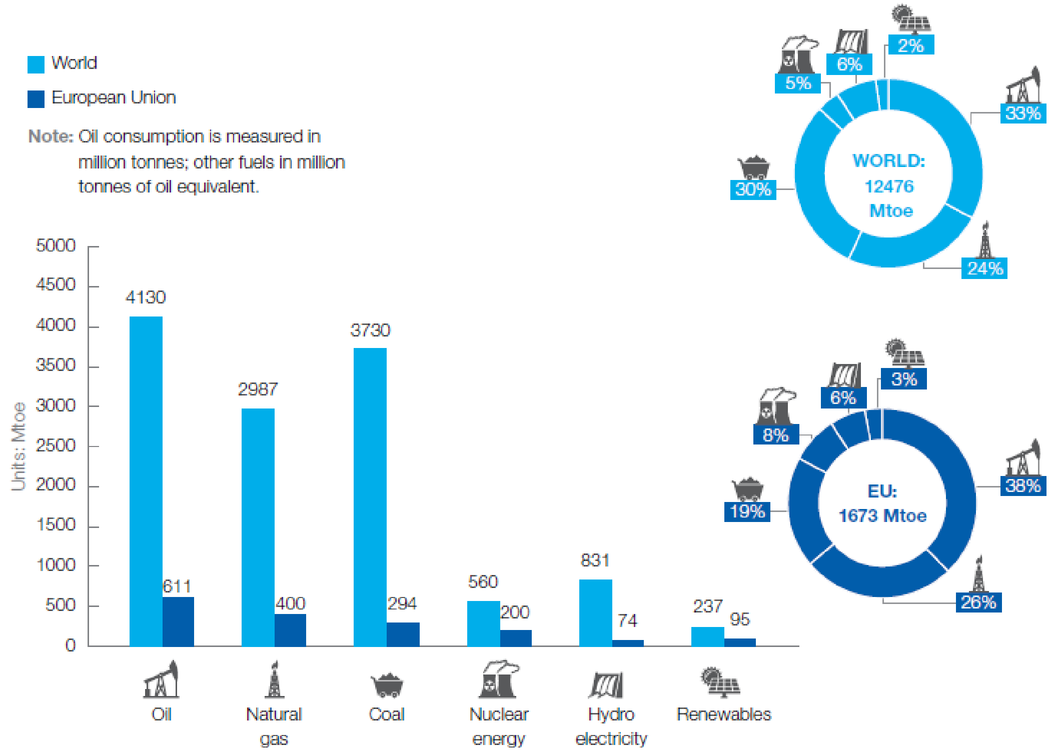
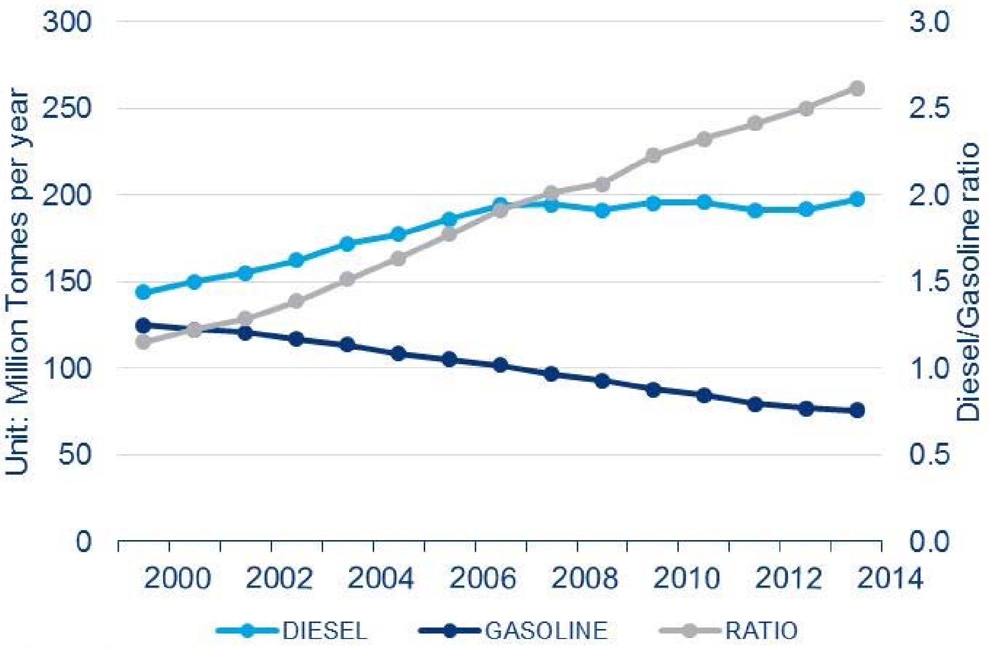


Fig 2: Worldwide Energy Consumption by Fuel Type in 2012 [5] [6]



Source: Wood Mackenzie

Fig 3: Road Fuel Demand in the EU [5] [7]

As can be seen in Fig. 3 [5] [7] the European gasoline demand in 2012 was approximately 80 Mt/y, while the diesel demand was about 200 Mt/y, which makes a total fuel demand of 280 Mt/y. If it is approximated that 280 Mt/y diesel and gasoline are equivalent to 280 Mtoe/y and half of the EU diesel/gasoline is replaced by NG approx. 140 Mtoe/y additional NG demand is generated, which is 35% of the actual NG consumption. Full replacement would lead to 280 Mtoe/y NG demand as automotive fuel which is 70% of the actual NG consumption

Therefore automotive transportation has the potential to become the main NG customer medium term, which implies that NG standards must be aligned with automotive requirements soon.

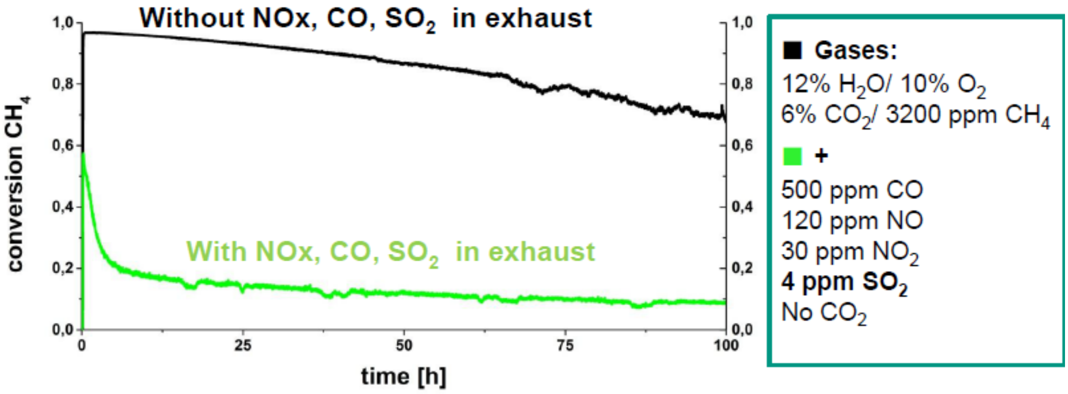
2 NG Quality Requirements

2.1 Catalyst Durability - Sulfur

In accordance with the European gasoline quality standard EN 228 [10] the automotive industry requires a maximum sulfur limit of 10 mg S per kg fuel. This limit is required in order to protect exhaust gas aftertreatment systems from sulfur poisoning. Higher sulfur concentrations in the fuel lead to increased sulfur loading in the exhaust gas and are hazardous for the aftertreatment durability.

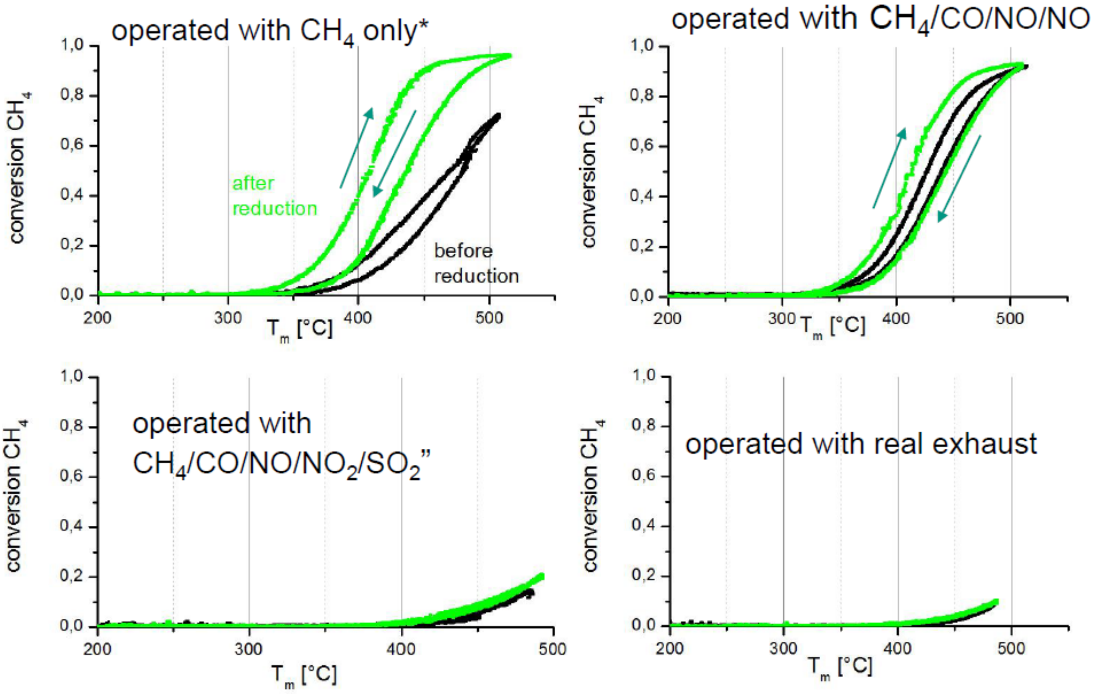
In Fig. 4 the effect of sulfur poisoning on a catalyst is shown. The presence of 4 ppm SO_2 in the exhaust gas (which is equivalent to approximately 30 ppm sulphur in the fuel) leads to a 91% reduction in CH_4 conversion rate after 100 hrs aging under severe laboratory conditions. [8] This is a considerable loss in methane conversion efficiency.

As shown in Fig. 5 the regeneration of the aged catalysts by reduction with H_2 at 400°C for 1 h, has not been successful, while it was successful without the presence of SO_2. Therefore no regeneration of the SO_2 poisoned catalyst could be achieved. [8]



- Strong deactivation of catalyst within 10 h in the presence of SO_2
- Only 9 % of methane converted after 100 h of operation at 450°C
- CO completely oxidized in all cases

Fig. 4: Catalyst Durability – Sulfur Effect on Conversion Efficiency [8]



- Regeneration achieved if SO_2 was not present in operation

Fig. 5: Regeneration of aged catalysts by H_2 reduction at 400°C for 1h [8]

In vehicle tests with De-NOx catalysts it has been demonstrated that 30 ppm of sulfur can lead to significant conversion efficiency reduction even after short distances. [9]

Therefore well established automotive fuel quality standards already contain sensible sulfur limits:

- 10 mg/kg in European gasoline standard EN 228 [10]
- 10 mg/kg in European diesel standard EN 590 [11]
- 10 mg/kg in German automotive NG standard DIN 51624 [14]

Thus NG as automotive fuel ideally must not contain more sulfur than 10 mg/kg or at least 10 mg/m^3.

The species of sulfur found in NG are usually: hydrogen sulfide, carbonyl sulfide, mercaptans, tetrahydrothiophene, carbon disulfide. There are mainly 2 origins of those sulfur species [9]:

- Natural sulfur: due to organic decomposition process → traces of sulfur → typically cleaned or processed close to the extraction points.
- Sulfur additive for odorization (for safety, since NG is odorless).

For most NG supplied to Europe the amount of natural sulfur is usually below the required limit of 10 mg/m^3. But NG is odorized for safety reasons. The majority of odorants are based on sulfur organic compounds, although sulfur free odorants are commercially available and are used e.g. in Germany, where 20…25 % of the odorants are already sulfur free [30]. In the draft automotive standard prEN 16723-2:2014 E it is proposed to apply a maximum sulfur limit only for non-odorized gas. For such gas a maximum content of 20 mg/m^3 total sulfur is proposed. The total amount of sulfur in the odorized gas, which usually is significantly higher, is not specified. For the automotive industry the lack of any limit for odorized NG is unacceptable, since most automotive CNG is supplied by the grid and sulfur is hazardous for the durability of exhaust gas aftertreatment components. For the final version of a standard an absolute total sulfur maximum of 10 mg/m^3 for the delivered (odorized) gas is required (in analogy to gasoline and diesel and as already introduced in the German automotive NG standard DIN 51624 [14]).

Country	**Sulfur (mean) / mg/m³**	**Sulfur (max. observed) / mg/m³**	**Components**
Belgium	**2.7**	**8**	**Total Sulfur**
Germany	**1.5**	**5**	**H2S + COS**
Netherlands	**1.5**	**6**	**Total Sulfur**
UK	**3.3**		**Total Sulfur**
Italy	**25**	**35**	**Total Sulfur**
Spain (odorized !)	**11**	**25.7**	**Total Sulfur**
Denmark	2.6		H2S
France	< 5	14	H2S

Table 1: Mean and maximum total Sulfur levels observed in different EU member states [9]

As shown in Table 1, non-odorized mean sulfur levels are usually below 10 mg/m³ in most EU member states. 10 mg/m³ are exceeded significantly mainly by sulfur entry via conventional odorization, which can be avoided since sulfur free odorants are commercially available. Furthermore sulfur peaks can be cushioned by controlled NG conditioning at NG grid entry points.

Another theoretical option would be to remove sulfur at the NG filling station [9]. But the disadvantage of this method is the complete removal of sulfur including all odorizers. Re-odorization would be required after sufur removal for safety reasons. The whole process would increase the investment costs and maintenance costs of the already expensive NG filling stations considerably and therefore hinder the expansion of NG station infrastructure significantly.

2.2 Energy Content – Wobbe Index

The Inferior Wobbe Index (WI) is specified as the inferior calorific value, on a volumetric basis, at specified reference conditions, divided by the square root of the relative density at the same specified metering reference conditions (dry air density) [12]. The WI is a measure of heat input to gas appliances derived from the orifice flow equation. Heat input for different natural gas compositions is the same if they have the same WI, and operate under the same gas pressure [12]. The WI has a considerable impact on injector flow rate demand [13] and is an important parameter for the dimensioning of NG engine injection systems. The WI is especially important for engines operated with open loop fuel metering control, typically gas engines based on diesel technology, since the WI determines the output power for such engines.

Thus regulation of WI is beneficial for automotive usage, whereby the Inferior Wobbe Index (WI) is better suited for automotive purposes than the Superior Wobbe Index (WS) as usual in the gas industry [13]. For automotive applications it is proposed to limit the Wobbe Index from 41.9 to 49.0 MJ/m³ for H-Gas and to set a lower limit of 40.5 MJ/m³ for L-Gas (calculated based on a LHV of 39 MJ/kg and a gas-density of 0.83/kg/m³)

2.3 Energy Content – Lower Heating Value (LHV)

The energy content of NG as automotive fuel is one of the dominant factors determining the mileage range of a NG vehicle. Mileage is in particular important for passenger vehicles because NG needs to be stored as a compressed gas (CNG) and therefore requires considerable more volume on board than competing liquid fuels. In order to enable a sufficient driving range of CNG vehicles, a limitation of the minimum energy content of automotive NG is required. Furthermore in Europe CNG is usually sold in "€ / kg", which also requires a limitation of the energy content related to the fuel mass in order to enable fuel cost transparency.

The usual fuel characteristic number to describe the energy content of a fuel is the Lower Heating Value (aka Lower Calorific Value) which should also be used to describe the energy content of NG. For high caloric H-Gas a Minimum Net Calorific Value of 44 MJ/kg is sufficient. For the low caloric L-Gas – available in some niche markets - a Minimum Net Calorific Value of 39 MJ/kg is required. Similar limits are already standardized in Germany for automotive NG (DIN 51624) [14].

2.4 Knock Resistance - Methane Number

The Research Octane Number (RON) [15] and Motor Octane Number (MON) [16] as used for gasoline fuel are insufficient to describe the knock behavior of NG, since the RON and MON scale ends at approximately 120 before typical NG starts to knock in a standard CFR engine [14], [17], [18], [19], [20]. For RON and MON higher than 120, no primary reference fuels (PRF) are available.

This was recognized in the 1960s when the Methane Number (MN) was developed to describe the knock resistance of NG. Instead of iso-octane / n-heptane mixtures, H_2/CH_4 mixtures are used as PRF. Basically two different methods have been in common use so far which lead to different results, the so called “AVL method” [14] [17] [18] [19] [20] and the so called “GRI method” [12] [21] [22].

Recently it has been agreed in Europe to apply the so called “MWM method”, a further development of the “AVL method”, which is thoroughly described in the appendix of the European proposed standard “Gas infrastructure — Quality of gas - Group H”, “FprEN 16726:2015” [23]. The introduction of the “MWM method” is also under consideration for other parts of the world.

Natural gas and bio-methane typically are considerably more knock resistant than gasoline. Therefore the fuel efficiency of dedicated spark ignited (SI) NG engines can be improved beyond what is possible for gasoline engines. Dedicated NG spark ignited engines are typically optimized for a methane number of 70 and can reach 40% efficiency with stoichiometric combustion systems. With lean combustion systems even higher efficiency can be achieved. As shown in Fig. 6, knock restricts optimum engine operation and causes efficiency degradation below MN 70 with a Compression Ratio (CR) of 12.5 on a boosted CNG SI engine. MN 65 instead of MN 70 causes a performance degradation of appox.10 %.

For EU V dual fuel engines (NG port fuel injection and diesel, diesel substitution with NG) the effect is even bigger. As displayed in Fig. 7 the amount of diesel which can be substituted by NG (Diesel Substitution Factor) is strongly dependant on the Methane Number.

Engines using the dual fuel technique can reach higher efficiencies than SI engines, close to the efficiency of the best diesel engines when the Methane Number is at least ~80. In the example in Fig. 7 a Diesel Substitution Factor above 75% and nearly diesel like efficiency is achieved for MN >83 while dual fuel operation below MN 78 is not possible.

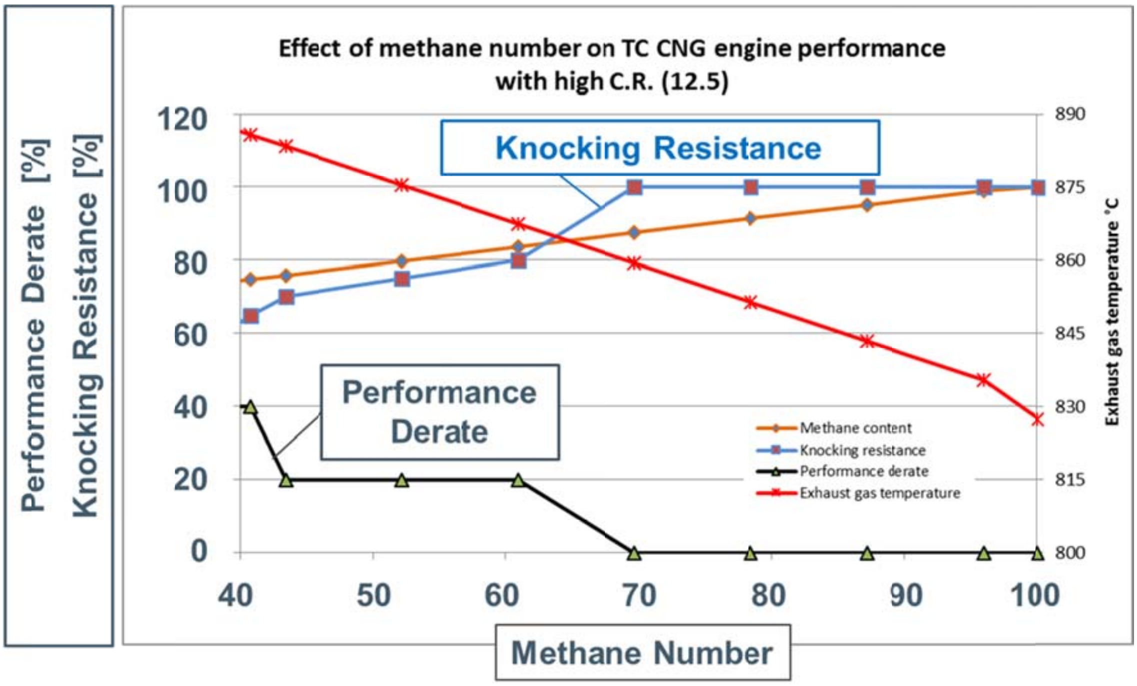


Fig. 6: Knock restriction and thus efficiency degradation below MN 70 with moderate CR 12.5 for a boosted CNG SI engine (Source: CRF)

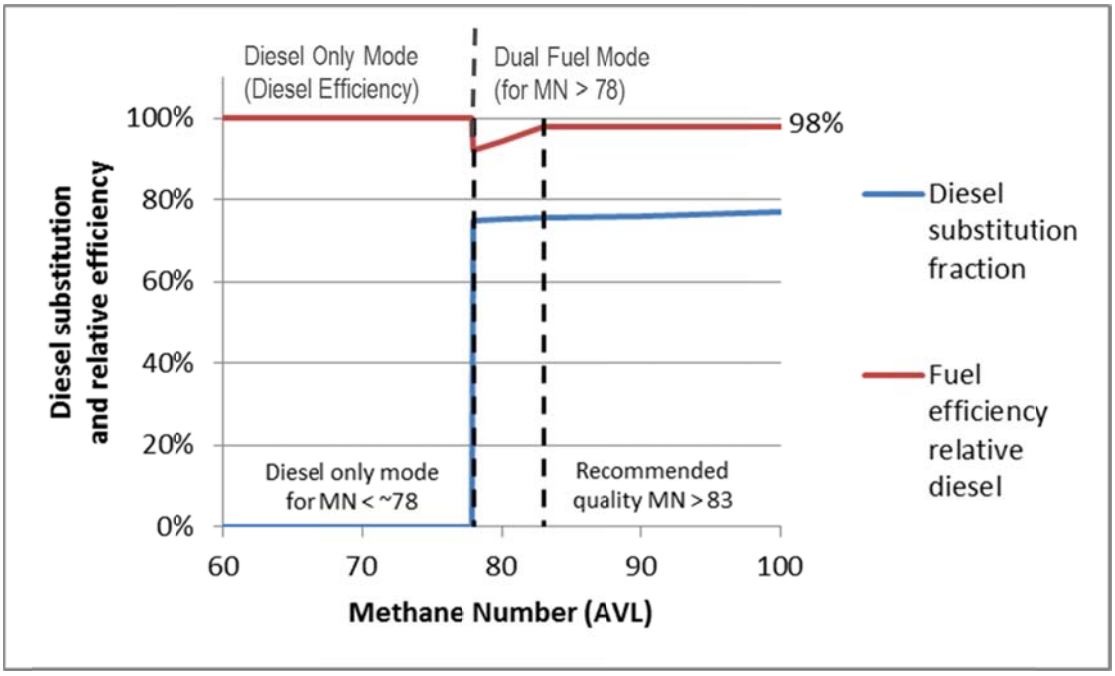


Fig. 7: Influence on Methane Number on Dual Fuel engine performance (Source: Volvo)

The lowest Methane Numbers generally occur in LNG (which can be directly used as LNG or re-evaporated to CNG). The worldwide LNG trading volume is approximately 7% of the worldwide NG market. [24]

As displayed in Fig. 8, only 3% of the worldwide LNG has a quality below MN 70 [24].

Therefore most NG (~ 99.8%) is delivered with MN above 70 [24]. Furthermore bio-methane and power-to-gas-methane are usually produced with a MN above 80. Thus automotive methane fuel – NG, bio-methane and any other methane - should at least provide a Minimum Methane Number of 70 (regular grade) at any point of sale. Underperforming NG can be conditioned before entering the grid. For dedicated applications (e.g. dual fuel engines or high compression ratio boosted spark ignition engines) an additional highly knock resistant grade with a Minimum Methane Number of 80 (premium grade) would be very beneficial to make further CO_2 reduction accessible for automotive applications. Those high MN grades can be distributed separately from the NG grid.

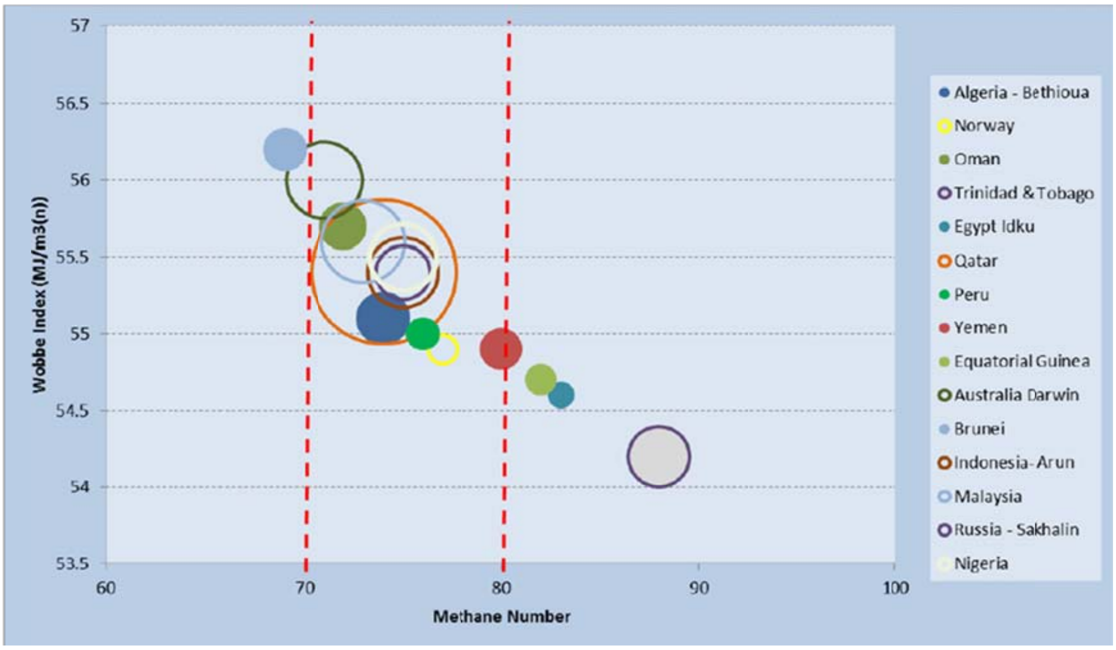


Fig. 8: Methane Number vs. Wobbe Index for LNG Qualities (LNG from different countries and amounts exported worldwide - based on average composition for 2013 and MWM calculation method for MN) [24]

2.5 Steel Tank Safety – Hydrogen

Hydrogen is another critical component in NG. It reduces the Methane Number; but more importantly it degrades high-strength steel CNG tanks (due to H_2 embrittlement). In accordance to ECE 110 [25] hydrogen shall be limited to 2% by volume when CNG tank cylinders are manufactured from steel with an ultimate tensile strength exceeding 950 MPa and for dry gas (water limited to less than 32 mg/m^3; pressure dew point of -9°C at 20 MPa), which is the usual gas quality of automotive NG. For wet NG (water content > 32 mg/m^3), the hydrogen limit would be 0.1% by volume.

2.6 Cleanliness – Compressor Oil

In order to avoid issues such as injector closing delay [27], pre-ignition etc., a limitation on compressor oil content is required. Unfortunately there is currently no standardized compressor oil concentration test method agreed. Reasonable and applicable test methods as well as agreed limits for compressor oil are urgently required.

Since the risk of oil contamination occurs at high pressures only (>100 bar) a compressor oil limit is only sensibly applicable to the automotive standard (no issue in the NG grid but during NG compression to vehicle tank pressure 200 bar). [26]

2.7 Cleanliness –Siloxane (Silicon Content)

Today CNG is blended with biogas, and the amount of biogas injected into the existing natural gas grids is growing; the fraction of biogas in automotive CNG fuel can be high (up to 100%). Those biogases can contain compounds which are not present in fossil-sourced NG. One species of those critical components are siloxanes. Siloxanes are widely used in numerous chemical products and end up in landfills and in the sludge of waste water treatment plants. As a result siloxanes can be found in bio-gases produced from landfill and wastewater sludge. Siloxanes can also be present in biogases from other sources when for example silicone based anti-foaming agents are used during biogas production. Siloxane can severely harm the lambda sensors of vehicles. Therefore the automotive industry requires a maximum limit of 0.1 mg/m^3.

Since there currently is no standardized test method available for measuring silicon at that low level and current bio-methane production processes cannot guarantee less than 0.5 mg/m³, the draft European automotive standard prEN 16723-2 (E) proposes a limit of up to 0.5 mg/m³, and the injection standard prEN 16723-1 (E) proposes up to 1 mg/m³.

As discussed for hydrogen, as well as for sulfur and silicon, the proposed automotive standard “prEN 16723-2:2014 E” and the proposed injection standard “prEN16723-1:2014 E” contain different limits. But according to the EU Directive “2014/94/EC” additional natural gas re-fuelling points are supposed to be put into place and to be supplied from the existing well-developed natural gas distribution networks in the EU. Therefore the standard on natural gas and bio-methane injected into the grid “prEN 16723-1:2014 E”, coupled with the standard for the quality of grid gas FprEN 16726:2015, will be the basis for what will be delivered for use in vehicles and is specified in the automotive standard “prEN 16723-2:2014 E”. Deviations of the automotive limits to the grid and injection limits may require dedicated treatment facilities at refuelling stations, which is technically not feasible for every parameter of poor quality grid gas. Even if it may be technically feasible, it would definitely lead to considerable costs for refuelling station operators. Since infrastructure costs are already high, any additional financial burden on the infrastructure will be very detrimental to the expansion of the alternative fuelling infrastructure.

In order to implement a sensible silicon limit into all 3 standards, a capable silicon concentration determination method needs to be developed. Therefore data of statistical silicon occurrence in current bio gas production should be gathered. As long as no relaxing data are available a limit of 0.1 mg/m³ should be introduced to protect lambda sensors.

3 European Standardization Status

European NG standardization is fragmented and handled by 2 standardization groups:

- CEN TC 234: Grid Standard
- CEN TC 408: (Bio-) Methane Injection Standard and Automotive Standard

Currently 3 different standards are proposed for Europe:

- Quality of CNG in the European grid: FprEN 16726:2015 (E) (TC 234)
- Quality of (bio-) methane injected into the grid: prEN 16723-1:2014 E (TC 408)
- Automotive NG / (bio-) methane fuel retail quality: prEN 16723-2:2014 E (TC 408)

Standardization issues from an automotive point of view are currently as follows:

Automotive NG / methane fuel retail quality: prEN 16723-2:2014 E (TC 408) [28]

- No Wobbe Index limit in requirement table
- No Lower Heating Value in requirement table
- Silicon limit not agreed. Proposed 0.5 mg/m³ limit is too high. No method agreed.
- H_2: max. 2% n/n is reasonable, but it should be specified in "% v/v" as in ECE110
- Sulfur: no limit agreed. Footnote: "difference between the automotive needs (10 mgS/m³ including odorization) and the values the gas industry may provide (30 mg/m³ including odorization)"
- Methane Number: 65 is too low. Footnote: "…only a small fraction of the distributed natural gas has a MN below 70 (MWM) "
- Compressor Oil: No limit. No sufficient method.

Quality of bio-methane injected into the grid: prEN 16723-1:2014 E (TC 408) [29]

- No Wobbe Index in requirement table
- No Lower Heating Value in requirement table.
- Silicon limit not agreed. Proposed 1 mg/m³ limit is too high. No method agreed.
- H_2: not in requirement table.
- Sulfur: not in requirement table.

- Methane Number: not in requirement table.
- Purpose of the complete standard is questionable.

Quality of NG in the European grid: FprEN 16726:2015 (E) (TC 234) [23]

- No Wobbe Index in requirement table
- No Lower Heating Value in requirement table.
- Silicon not in requirement table
- No H_2 in requirement table. Just a note in Annex E: "[…] admixture of up to 10 % by volume of hydrogen to natural gas is possible in some parts of the natural gas system […], steel tanks in natural gas vehicles: specification UN ECE R 110 stipulates a limit value for hydrogen of 2 vol%".
- Sulfur: limit only <u>before</u> odorization 20 mg/m³ is too high, no limit <u>after</u> odorization.
    - Footnote: "[…], for existing practices with respect to transmission of odorized gas between high pressure networks higher sulfur content value up to 30 mg/m³ may be accepted"
    - Grid standard needs to ensure automotive NG quality in order to ensure sufficient quality for connected filling stations.
    - Desulfurization at retail stations is economically unrealistic.
    - Upper limit must be specified after odorization. Should be 10 mg/m³.
- Methane Number 65 is too low.

  Positive: MWM method sufficiently laid out in "Annex A".

4 Summary & Conclusions

- NG (+ renewable methane) as fuel has a considerable greenhouse gas (GHG) and cost reduction potential. Thus it is in the focus of many OEMs for future automotive transportation, in particular as fuel for dedicated, highly efficient NG engines. e.g. downsized SI engines for passenger car applications (→ Horizon 2020 GasOn EU project) and as dual fuel engines for HD long haul trucks
- Any significant shift from oil based fuels to NG/methane as automotive fuel would increase the NG demand considerably.
- Automotive transportation has the potential to become the main NG customer. Thus NG standards need to be aligned with automotive needs.
- European methane standardization is fragmented and handled by different standardization groups (CEN TC 234 and CEN TC 408).
- Currently 3 different standards are proposed for Europe: grid (FprEN 16726:2015), bio-methane injection (prEN 16723-1:2014 E) and automotive (prEN 16723-2:2014 E) quality.
- All standards – including grid standard - need to ensure automotive NG quality.
- Upgraded standards with appropriate limits are required for: Wobbe Index, Lower Heating Value, silicon, sulfur, H_2, Methane Number, and Compressor Oil.
- Fuel Quality Directive for methane fuels is recommended.

5 Recommendations

Parameter	**Unit**	**Min**	**Max**	**prEN 16723-2**	**prEN 16723-1**	**FprEN 16726**	**Comment**
Net Wobbe Index (H-Gas)	MJ/m³	41.9	49.0	+	+	+	
Net Wobbe Index (L-Gas)	MJ/m³	40.5	-	+	+	+	no upper limit (transition to H-Gas)
Lower Heating Value (H-Gas)	MJ/kg	44	-	+	+	+	
Lower Heating Value (L-Gas)	MJ/kg	39	-	+	+	+	
Sulfur Total	mg/m³	-	10	+	+	+	including odorization
Methane Number (high grade)	MWM	80	-	+	+	+	dual fuel requirement, non-grid distribution
Methane number (regular grade)	MWM	70	-	+	+	+	
Total Siloxanes (calculated as Si)	mg/m³	-	0.1	+	+	+	capable test method to be agreed
Hydrogen	% v/v	-	2	+	+	+	according to ECE 110
Compressor oil	mg/m³	-	tbd.	+	-	-	method and limits to be agreed (automotive standard only)

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