THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

# Emission Control for Hydrogen Internal Combustion Engines

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# Abstract

Hydrogen internal combustion engines have the potential to reduce greenhouse gas emissions from the transportation sectors, especially for heavy-duty road transport, such as long-haul trucks.

There are however still local emissions from hydrogen internal combustion engines that need to be minimized. These include nitrous oxides and particle emissions. Any high temperature combustion process that occurs in an excess of air has the potential to generate significant number of nitrogen oxides.
The particle emission on the other hand stems from the combustion of engine lubricating oil. This occurs in engine running on fossil fuel as well, but the effect on the emissions may be less significant when compared to those of incomplete burning of fossil fuels. The particle emission can also be significantly higher, as the short quenching distance of hydrogen flames allow combustion to take place closer to cylinder walls, as well as transferring more heat into the walls.

Another issue is that the exhaust after-treatment system can cause emission nitrous oxide, $N_2O$, or laughing gas, which is a very potent greenhouse gas with close to 300 times the greenhouse warming potential of carbon dioxide.

Experiments were conducted to measure the engine out emission, both regulated and unregulated of a direct injection, spark ignition hydrogen internal combustion engine. The results showed that air to fuel ratio is the most important factor for determining $NO_X$ emissions. At the same time, the particle emissions increased dramatically when the engine was throttled, and cylinder and intake pressure reduced to below crankcase pressure.

A heavy-duty, direct injection, spark ignition, engine fitted with ah exhaust after-treatment system has been used for measurement of the impact of engine operating parameters on the engine out emissions, as well as the effect of these engine parameters on the performance of the different components in the exhaust after-treatment system. The results indicates that low tailpipe emission is possible, although the experimental setup does not provide enough precision in operating conditions for the catalyst to quantify the effects of water content and catalyst efficiency, due to unsteady emission levels and confounding between variables.

# List of Publications

**Paper A** V. Berg, L. Koopmans, J. Sjöblom, and P. Dahlander, "Characterization of Gaseous and Particle Emissions of a Direct Injection Hydrogen Engine at Various Operating Conditions," *SAE International Journal of Advances and Current Practices in Mobility,* vol. 6, no. 3, pp. 1746-1757, 2023, doi: https://doi.org/10.4271/2023-32-0042.

**Paper B** V. Berg. "Experiments on Hydrogen Engine Exhaust After-Treatment Systems" (in-house report)

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# Contents

# 1 Introduction

## 1.1 Global Emissions

Global warming, due to increased greenhouse gas emissions, is a major challenge to societies worldwide. The impact of a hotter planet includes more frequent droughts, heavier precipitation and more intense storms, as well as a loss of biodiversity [1]. Despite efforts, the global emissions of greenhouse gases continue to increase, and there is evidence that the measures currently taken, and the technologies currently used is not enough to avoid adverse effects [2]. In 2018, the transport industry accounted for 24 % of global $CO_2$ emissions. Of these emissions, road passenger transport 45.1 % while freight accounted for 29.4 % [3].

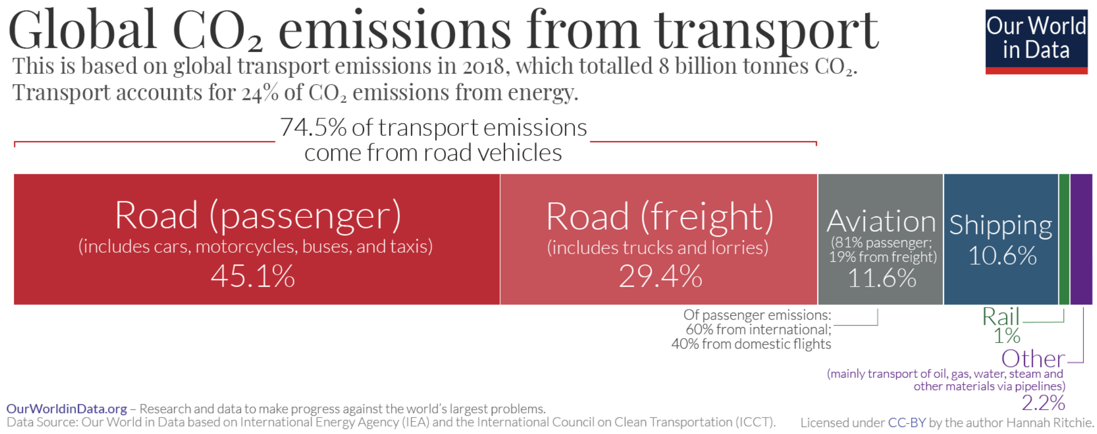


*Figure 1: Global emissions from transport [Hannah Ritchie (2020) - "Cars, planes, trains: where do $CO_2$ emissions from transport come from?" Published online at OurWorldinData.org. Retrieved from: 'https://ourworldindata.org/co2-emissions-from-transport']*

Solutions for personal use vehicles such as cars and motorcycles include pushing for electrification, through battery-electric vehicles (BEV) and hybridisation, which seems to be the favoured way forward. The market share of electric vehicles is increasing, especially in light-duty (car) sales [4].

Heavy-duty vehicles and especially vehicles that need high energy density, such as long-haul trucks, show much slower adoption of battery-electric powertrains, and there are challenges with regards to e.g. range that remains. This means that while electrification of the passenger car sales is progressing, and the market share is steadily increasing, the market share of electrified trucks is far smaller [4]. This makes investigating alternative approaches to carbon neutrality important.

The increased focus on greenhouse gas emissions has caused a renewed interest in renewable fuels, as efficiency improvements alone are incapable of reducing the emissions to harmless levels while running on fossil fuels.

Related to this, the transport sector has a unique set of challenges, most notably the energy density of the energy which limits the range weight of the vehicle or the load it can carry as well as the weight and the power density of the powertrain, including any aftertreatment system needed to reduce emissions to harmless levels.

An alternative to electric vehicles in heavy duty road transport, especially for long distances is to replace the fossil fuel with a renewable fuel.

Options include biofuels and electro-fuels. Biofuel options include alcohols for SI engines, and HVO and FAME for compression ignition engines, while the process for creating electro fuels start with hydrogen production. Depending on the desired fuel, the hydrogen will be combined with either carbon dioxide or nitrogen. The carbon containing pathways include the Fischer-Tropsch process to produce e-diesel and e-gasoline, while the Sabatier process can give methane. Methanol can be produced trough hydrogenation of $CO_2$, and is a pathway to DME, and e-gasoline and e-diesel as well [5, 6]. Long term and widespread adoption of these e-fuel would however require large scale CCS facilities as well [6].

To achieve zero tailpipe emissions, the fuel should be carbon free, which at present only leaves hydrogen and ammonia as the options.

Ammonia can be produced from green hydrogen trough the Haber-Bosch process [6]. Benefits of ammonia compared to carbon containing fuels is that the costly carbon capture is avoided, and that the technology is mature as the process is widely used for ammonia production for fertilizer. Compared to pure hydrogen, advantages of ammonia is a higher energy density. Ammonia can be used as a fuel for both spark ignition engines and compression ignition engines [7, 8]. Downsides of ammonia as a fuel include emissions of unburned ammonia, $N_2O$ emissions and high $NO_X$ emissions. In the case of compression ignition engines, long ignition delay times reduce efficiency and combustion stability [7, 8]. In the case of spark ignition engines low flame speed leads to low efficiency and high unburned $NH_3$ emissions at lean conditions, while rich operation can lead to very high $NO_X$ emissions [9]. For both types of engines, a complementary fuel may be needed, adding to the complexity of the system. Finally, ammonia is very toxic to human and aquatic life.

Hydrogen is the simplest possible form of electro fuel that can be produced and contains no carbon. It has therefore the potential to reach climate goals if the hydrogen is produced from renewable sources [10]. Hydrogen can be used to fuel either a fuel cell or a combustion engine. Both solutions have their relative benefits and drawbacks. The benefits of fuel cells are zero tank to wheel (TTW) greenhouse gas emissions and relatively high efficiency [11], however their drawbacks involve high cost of manufacturing, complexity, volumetric power density of the system and questions regarding longevity. An internal combustion engine may be more attractive in the near future, as the technology is mature and could be adapted to the new fuel. The benefits of an internal combustion engine operated on hydrogen are zero TTW $CO_2$ GHG emissions, a relatively high power-density, cost and robustness, and good efficiency at high power. The possibility of using hydrogen as a fuel has been known for over 200 years, although the impact on society has been marginal. In fact, what could be argued to be the first "car" ever, constructed in 1807 by François Isaac de Rivaz was powered by a hydrogen internal combustion engine [12]. Although there were no commercial breakthroughs, several car manufacturers, such as BMW [13], Mazda as well as suppliers [14] ran demonstration vehicles with internal combustion engines that had been converted to hydrogen in the early 20000's , proving the concept feasible.

However, carbon neutral fuel does not guarantee that the emissions of local pollutants are zero.

## 1.2 Local Emissions

Road transport is a major cause of emissions that affect local air quality, such as black carbon/soot (BC), and particles as well as emissions that contribute to acidification, such as oxides of nitrogen ($NO_X$). An overview of some of the local pollutants released in the European union, by sector, is shown in Figure 2.

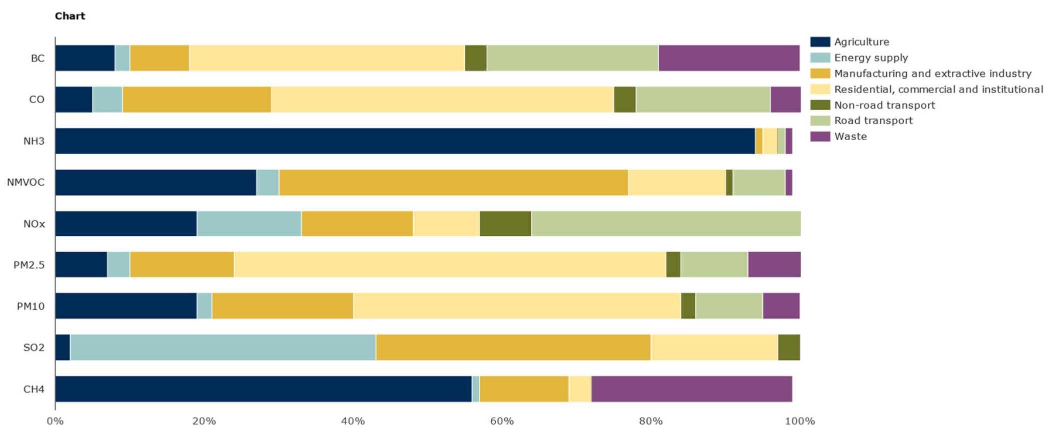


*Figure 2: Pollutants by sector in the EU. [EEA,* Sources and emissions of air pollutants in Europe*, 2022] [15]*

It is therefore important that any technology that replaces fossil fuels also solves to problem of local pollutant. To combat this, concurrently with the push to electrification, more stringent emission legislation is adopted in the European union, with the shift from the older EURO VI emission standard [16] to the newer EURO 7 standard [17]. The benefits of an internal combustion engine operated on hydrogen are zero TTW $CO_2$ GHG emissions, a relatively high power-density, cost and robustness, and good efficiency at high power. However, carbon neutral fuel does not guarantee that the emissions of local pollutants are zero.

Finally, trying to reduce the emissions of nitrogen oxides trough an exhaust after-treatment system introduces new challenges. Urea, dissolved in water, is commonly used as storage of ammonia, which is commonly used as the reducing agent in commercial systems. While ammonia is carbon free, the urea molecule contains a carbon atom. Excessive consumption of urea can thus give emission of carbon dioxide, meaning that even though the fuel is carbon free, the exhaust gases contain carbon above the limits for zero emission vehicles.

The main emissions of an internal combustion engine are oxides of nitrogen formed due to the high heat during combustion and the excess of oxygen and nitrogen present during lean combustion and particles.

### 1.2.1 Nitrogen Oxides

The emissions of a hydrogen-fuelled internal combustion engine mainly consist of nitrogen oxides ($NO_X$) and particulates.

Nitrogen oxides ($NO_X$) consist primarily of nitric oxide (NO) and nitrogen dioxide ($NO_2$). In the exhaust of a combustion engine, the largest fraction of total NOX will typically be NO, which formation is often described by the extended Zeldovich mechanism [18]:

$$N_2 + O_2 \leftrightarrow NO + \ N \tag{1.1}$$

$$N + O_2 \leftrightarrow NO + O \tag{1.2}$$

$$N + OH \leftrightarrow NO + H \tag{1.3}$$

The high activation energy of equation 1.1 and the high activation energy of the reverse reaction of equation 1.3 leads to a very strong temperature dependence, where high temperatures favour the formation of NO.

$NO_X$ emissions from hydrogen engines can be reduced to very low levels by running the engine at very lean conditions [19-23], lowering the peak temperatures in the combustion chamber. Very lean operation presents an obstacle to maintaining power output, which is already a challenge for most spark ignition hydrogen engine concepts and poses a challenge for a potential boosting system [24-26]. The lean mixtures will contain low exhaust enthalpy while high intake air flows at high pressures will be required.

Mitigation strategies follow two main strategies. The first is reducing the engine-out emissions as much as possible. The nitrogen oxides emissions can be reduced significantly trough lean combustion. While this is theoretically possible and can be achieved in stationary operation, it reduces an engines specific power output and places higher demands on a possible boosting system. It may not be able to stay below the limits at all operating points. The engine out particle emissions, while typically lower than a modern gasoline or diesel engine, can still be problematic [27-29].

### 1.2.2 Particles Emissions from Hydrogen Engines

The combustion of hydrogen could be expected to produce no particles, however incomplete combustion of lubricating oil in the cylinder leads to particle formation. Results with port injection have shown that gaseous fuel, and hydrogen especially can reduce the amount of particles emitted [30]. Results for direct injection engines, however, have showed that direct injection of hydrogen can lead to very fine particles in the sub 30 nm range being emitted [27, 28, 31].

These emissions of ultrafine particles can reach even higher numbers than those for times in number higher than in conventional gasoline powered engines [31]. An explanation proposed by the authors was that the high speed of the high-pressure hydrogen jet at the point where it impinges on the cylinder wall would remove oil from the cylinder wall. The turbulent jet could also entrain adjacent oil vapours into the jet, which in turn could increase emissions of particles [32].

With new emissions regulations, such as the new EURO 7 regulation [17], a number of new species are introduced in the legislation, and it is essential to investigate what the emission levels of these species are. Mitigation of particle emissions is done by particle filters, but in a hydrogen engine. However, many of the traditional means of accomplishing low engine-out emissions may not be relevant to hydrogen engines, as the particles are not formed as a result of incomplete combustion of the fuel.

## 1.3 Engine Technology in Hydrogen Engines

In this section several technologies relevant to this project are described. An overview of different approaches to carbon neutral road transportation is shown in Figure 3:

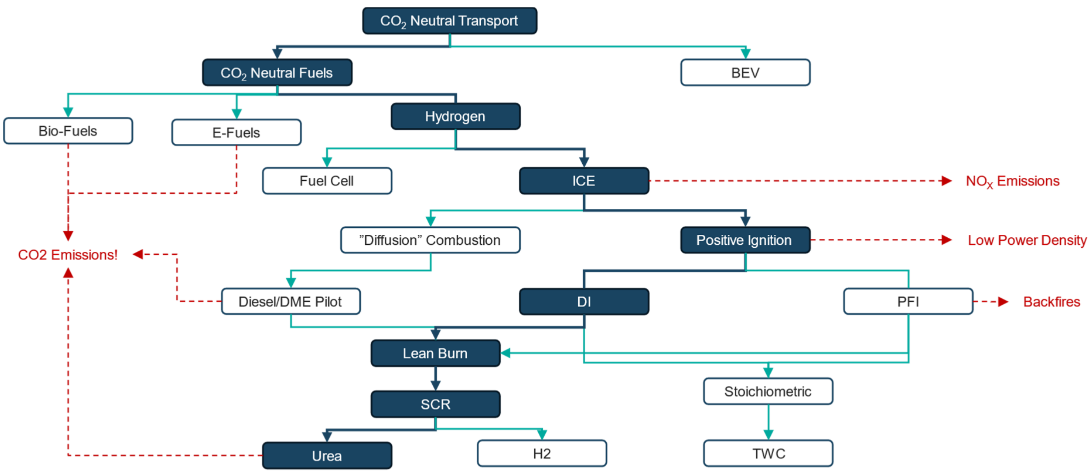


*Figure 3: Overview of different technologies for carbon neutral road transport.*

### 1.3.1 Ignition and Combustion Concepts

The simplest form of hydrogen internal combustion engine is a port fuel injected (PFI) engine. This was common approach in early research on hydrogen engines due to the simplicity of replacing a conventional fuel injector with a hydrogen one. Advantages, apart from simplicity is that this approach enables the hydrogen feed to be at relatively low pressures. This type of engine, however, has potential drawbacks in the form of low specific power and risk of backfires, due to long time in contact with hot gases and/or engine parts coupled with the low ignition energy of hydrogen [20]. The low ignition energy of hydrogen and wide flammability limits may even allow small amounts of fuel from the last cycle are still burning in the ring lands, igniting the incoming charge while the intake valve is still open [33].

DISI engines using low pressure direct injection (LPDI) can operate without the need for a pilot fuel, which is frequently needed in high pressure direct injection (HPDI) systems [21]. As most of the suggested fuels suitable for compression ignition, for example diesel and DME, contain carbon [20], $CO_2$ will be emitted. To avoid this, and the complexity of having to carry two fuels and a urea solution on board, and to enable close to zero greenhouse gas emissions, a low-pressure direct injection (LPDI), spark ignition engine concept is chosen for the work in this thesis. These systems can be considered a middle of the road approach, combining some of the advantages of port fuel injection and HPDI systems, and by injecting the fuel after the intake valve closes, volumetric power density can be increased over the port fuel injection systems [20, 23], while also decreasing the demands on the boosting system. Using DI to inject hydrogen after the intake valve closes is attractive for power density and to reduce pumping work but can lead to inhomogeneous mixing and increased $NO_X$ emissions [23, 34, 35], as well as impingement of the jet on the cylinder walls, leading to high particle emissions [31].

### 1.3.2 Boosting Systems

Operatizing a hydrogen internal combustion engine under very lean conditions at high loads provides a challenge for the engines boosting systems. The combination of high pressure and flow rate demand as well as low exhaust temperatures, and thus low energy to drive the turbine, can make it challenging to reach the target air/fuel ratio for $NO_X$ control [36]. The resulting boosting system.

Even though improvements in turbocharger design could allow lean operation of gasoline over a wide load range [37], the air/fuel ratio at high load may not be lean enough. The increases demand for boost even when there is low exhaust energy or quick response is needed may require solutions similar to those found in heavily downsized gasoline engines, and could use a both a supercharger and turbocharger [38] or a mechanically driven turbo [39].

### 1.3.3 EGR

Compared to lean operation, exhaust gas recirculation (EGR) implemented on hydrogen engines pose some challenges, including condensation of the vapor in the exhaust. The resulting reduction in $NO_X$ may also be less significant, compared to lean operation [36].

Water injection is also an alternative to EGR, and can avoid some of the drawbacks of an EGR system, while providing significant reduction in nitrous oxide emissions [34]. The drawback is that the vehicle must now carry additional water onboard, or complex systems for recovery of water from the exhaust stream.

## 1.4 Exhaust After-Treatment Systems

Given the complexities of maintaining lean enough engine operating conditions at high load and in transients, an exhaust after-treatment system (EATS) could still be needed to reach the emission targets under all conditions.

Such a system, for a lean burn, direct injection engine would be relatively similar to a conventional diesel engine EATS.

Another issue that is the focus of this paper is the engine out emissions, which can be high in nitrous oxides ($NO_X$). An approach that is attractive is to lean out the hydrogen mixture enough to reduce engine out emissions to acceptable levels [20, 23]. This typically reduces power density of the engine, which is already a challenge for most spark ignition concepts. Lean combustion also poses challenges for a potential turbo [24-26], as the lean mixtures will produce exhaust of low specific enthalpy while high intake air flows at high pressures will be required.

Apart from strategies to lower engine out emissions, an exhaust aftertreatment system (EATS) may be needed. A selective catalytic reaction (SCR) catalyst can be used to reduce the NOx emissions of a hydrogen engine, and has been proven to be able to reduce the emissions to the legal limit, both in stationary tests [24] and in the WLTC drive cycle [40].

### 1.4.1 Selective $NO_X$ Reduction

One key technology to reduce the emissions of nitrous oxides from the exhaust of any learn-burn internal combustion engine is the selective catalytic reduction (SCR) catalysts.

Urea, which decomposes to ammonia in the exhaust pipe upstream of the SCR catalyst contains carbon, which currently is a problem for a system that aim for zero carbon emissions. The urea consumption thus needs to be minimised.

The reactions that take place in an automotive selective catalytic reaction (SCR) catalyst are summarised below [41]:

$$\mathrm{NH_3 + NO + \frac{1}{4}O_2 \rightarrow\ N_2 + 3/2H_2O} \tag{1.4}$$

The reaction above is sometimes referred to as the **normal SCR reaction** and is what is typically dominating the reactions at high $NO/NO_X$ ratio. The reaction is desirable; however, it is not the fastest reaction pathway for reducing NO. In the presence of $NO_2$, another, faster reaction can take place. This is shown in equation ( 1.5):

$$NH_3 + \frac{1}{2}NO + \frac{1}{2}NO_2 \rightarrow N_2 + \frac{3}{2}H_2O \tag{1.5}$$

This reaction is sometimes referred to as the **fast SCR reaction** and is generally seen as the most desirable reaction in an SCR catalyst. As exhaust gases from an internal combustion engine, whether it is compression ignitions of diesel or spark ignition of gasoline typically contains mostly NO [18]. With only small amounts of $NO_2$, an oxidation catalyst is typically used. In addition to oxidising the hydrocarbon and carbon monoxide in diesel engine exhaust, it oxidises part of the NO into $NO_2$, according to the reaction in equation [18]. With only small amounts of $NO_2$, an oxidation catalyst is typically used. In addition to oxidising the hydrocarbon and carbon monoxide in diesel engine exhaust, it oxidises part of the NO into $NO_2$, according to the reaction in equation ( 1.6):

$$NO + \frac{1}{2}O_2 \rightarrow NO_2 \tag{1.6}$$

Achieving the desirable $NO_2$ to total $NO_X$ ratio is one of the challenges of designing a successful SCR system, as a $NO_2/NO_X$ ratio above 0.5 forces the reaction of $NO_2$ and ammonia instead, which is described in equation ( 1.7). This reaction pathway is slower than both the normal and fast SCR reaction.

$$NH_3 + \frac{4}{3}NO_2 \rightarrow \frac{7}{8}N_2 + \frac{3}{2}H_2O \tag{1.7}$$

This is however not the mayor drawback to high $NO_2$ content. An undesirable reaction can also occur if there is an excess of $NO_2$ This reaction is shown in ( 1.3):

$$NH_3 + NO_2 \rightarrow \frac{1}{2}N_2 + \frac{1}{2}N_2O + \frac{3}{2}H_2O \tag{1.8}$$

This describes the reaction between ammonia and $NO_2$ that causes the formation of $N_2O$. This could also occur locally, due to imperfect mixing of the reactants. The $N_2O$ released is a potent greenhouse gas, which means that control of this reaction is very important for any engine that uses an SCR system, but even more so for a hydrogen internal combustion engine.

Another issue with the reaction between ammonia and species in the exhaust, is the formation of ammonium nitrate, that can block channels in the SCR catalyst and reduce the catalyst efficiency. To combat this, it is important that urea is not injected at low temperatures. Tough cold start behaviour is not in the scope of this research project, this provides an incentive for finding methods to either reduce $NO_X$ emissions during cold start, as well as making sure that the catalyst reaches the light-off temperature as quickly as possible.

Urea deposits are typically of less concern in real world applications, where regeneration of diesel particle filters heat the exhaust gases downstream of the DPF to temperatures where urea deposits break down [41].

### 1.4.2 Water Impact on SCR Systems

Hydrogen internal combustion engines typically have very high water content in the exhaust gases, which can negatively affect conventional SCR systems, by both reducing the $NH_3$ storage capacity of the SCR catalyst and by reducing the conversion efficiency [42]. Water in the exhaust can be a hazard for most zeolites, and the high water concentration in the exhaust of a hydrogen combustion engine could be problematic. At low temperatures the zeolite remains stable even at high water concentrations. However, as high temperatures need to be avoided, urea deposits may be harder to avoid.

The presence of water in the exhaust has been found to improve the de-$NO_X$ performance of a copper-zeolite SCR system, while leading to increased $N_2O$ formation [43]. This was however tried at lower water concentrations than those present in a hydrogen internal combustion engine, especially at air to fuel ratios that lead to high $NO_X$ emissions [20, 24, 44-48].

### 1.4.3 Other EATS Components

In an EATS, there are also other catalytic components. These components are not the main focus of this thesis and only briefly described here.

- DOC (Diesel Oxidation Catalyst): Oxidizes CO and HC to $CO_2$ and water. Also oxidizes NO to $NO_2$ for improved performance in DPF and SCR.
- DPF (Diesel Particulate Filter): Captures and oxidizes the Particulate emissions, utilizing the $NO_2$ from the DOC to oxidize the particulates. Often, the DPF is also contains oxidation functionality to improve the SCR performance (as outlined above)
- ASC (Ammonia Slip Catalyst): Oxidizes any residual ammonia from the SCR catalyst, and is

## 1.5 Objective

This work focuses on the local emissions of hydrogen engines. While efficiency and fuel consumption of an engine will always remain important qualities, those qualities are outside of the scope of this work, that will instead focus on the local emissions.

Most of the research performed in this project so far has focused on a single engine operating point, reporting the emissions of common legislated species. This research focuses on investigating the total emissions from a direct injected hydrogen engine over a variety of operating points and investigating parameters affecting these emissions as well as to investigate the origin of the particulate emissions further.

The work in this licentiate thesis was of an experimental nature. The goal of the work in this thesis aimed to answer:

- What are the regulated and unregulated emissions of a hydrogen internal combustion engine?
- What engine operating parameters influences these emissions?
- What is the impact of the unique properties of the exhaust from hydrogen combustion on a conventional exhaust after-treatment system?

-

# 2 Experimental Setup

The experimental work performed has been done on two different single cylinder research engines at the laboratory at Chalmers. The first of the two is a light-duty engine like those found in passenger cars. It is on this engine the data used in the first publication was obtained. The other engine, where research on the implantation of an exhaust after-treatment system took place, is a heavy-duty engine similar to those typically found in commercial diesel trucks. Both engines have been modified to run on hydrogen.

## 2.1 Light-Duty Engine

The first engine used in the work presented in this thesis, and from which all the results included in the first publication were obtained, is shown in Figure 4. The engine is light duty research engine from AVL, with a piston and cylinder head typical of a direct injection, spark ignition passenger car. The modification made to the engine apart from safety features, were limited to a platina free spark plug and replacement of the centrally mounted fuel injector with a prototype hydrogen injector.

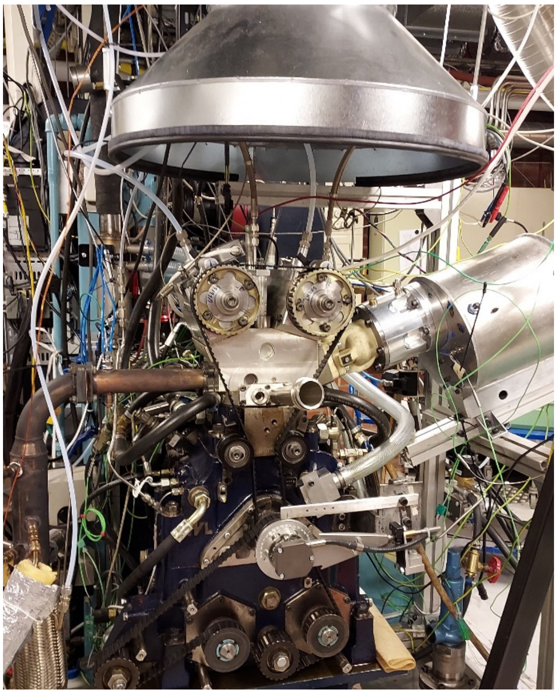

*Figure 4: Close-up of the light-duty engine used for the work presented in paper A.*

The engine's crankcase ventilation system was externally force vented and not fed back to the intake to reduce the risk of buildup of hydrogen in the crankcase. The forced ventilation was used to increase the air flow to the crankcase, thus diluting the hydrogen in the blow- by gases before it could reach the flammability limit.

As the higher loads in the experiments necessitated intake pressures above atmospheric, there would be no point with pressure lower than the crankcase to vent the gases to, which required the crankcase to be vented to the low pressure created by the test cell's exhaust fan. Venting upstream of the compressor turbine may solve this issue on engine equipped with turbochargers, but the test cell was limited to pressurised air from an external compressor, rendering this approach impossible. As such, any effect recirculating the blow-by gases could not be investigated.

The hydrogen injection pressure was controlled by a mechanical pressure regulator. The injector was fed through a single pipe, without any buffer reservoir. The volume of the pipe from the pressure regulator to the injector was large enough to avoid significant pressure drop during injection.

*Table 1: Light-duty engine specifications.*

| **Light Duty Engine** | | |
|---|---|---|
| Displacement | 0.475 | dm3 |
| Bore | 90 | mm |
| Stroke | 82 | mm |
| Conrod length | 139 | mm |
| Compression ratio | 10:01 | - |
| Intake valve opening | -364 | °ATDC |
| Intake valve closing | -142 | °ATDC |
| Exhaust valve opening | 145 | °ATDC |
| exhaust valve closing | 357 | °ATDC |

## 2.2 Heavy-Duty Engine

The engine used in this study is a heavy-duty hydrogen engine, based on an AVL research engine. The single cylinder engine is typical of those found in commercial diesel trucks, apart from modifications made to operate the engine in hydrogen.

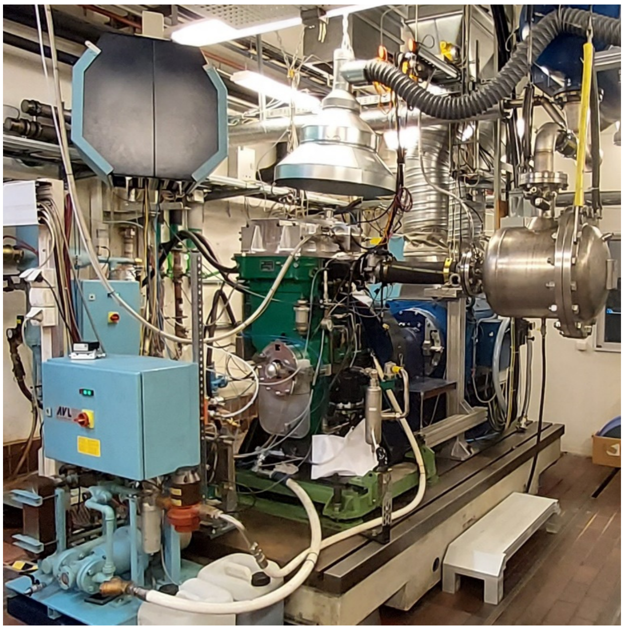

*Figure 5: The heavy-duty engine used for the exhaust aftertreatment tests.*

The most important changes made to the engine was a prototype piston the lowers the compression ratio and lacks the typical diesel bowl. The head has also been modified to allow the fitting of a spark plug and a prototype, single hole hydrogen injector.

The fuel supply consists of tanks of compressed hydrogen, a PLC-controlled pressure regulator, stainless steel lines and a small buffer tank just upstream of the injector. The function of the buffer tank is to avoid sharp pressure drops during injections, by allowing a larger volume of hydrogen close to the injector. This also simplifies the operation of the pressure regulator, as the pressure at the hydrogen bottles fluctuates less.

Test bed operation is done through an AVL PUMA system, and all engine operating parameters are set manually. A summary of the engine data is given in Table 2:

*Table 2: Summary of heavy-duty engine data.*

| **Heavy-Duty Engine** | | |
|---|---|---|
| Displacement | 2.13 | dm3 |
| Bore | 131 | mm |
| Stroke | 158 | mm |
| Conrod length | 259 | mm |
| Compression ratio | 10.5:1 | - |

The engine was fitted with an exhaust aftertreatment system consisting of an oxidising catalyst and a selective catalytic reaction system.

## 2.3 Exhaust After-Treatment Systems

The EATS used for the heavy-duty experiments is shown in Figure 6.

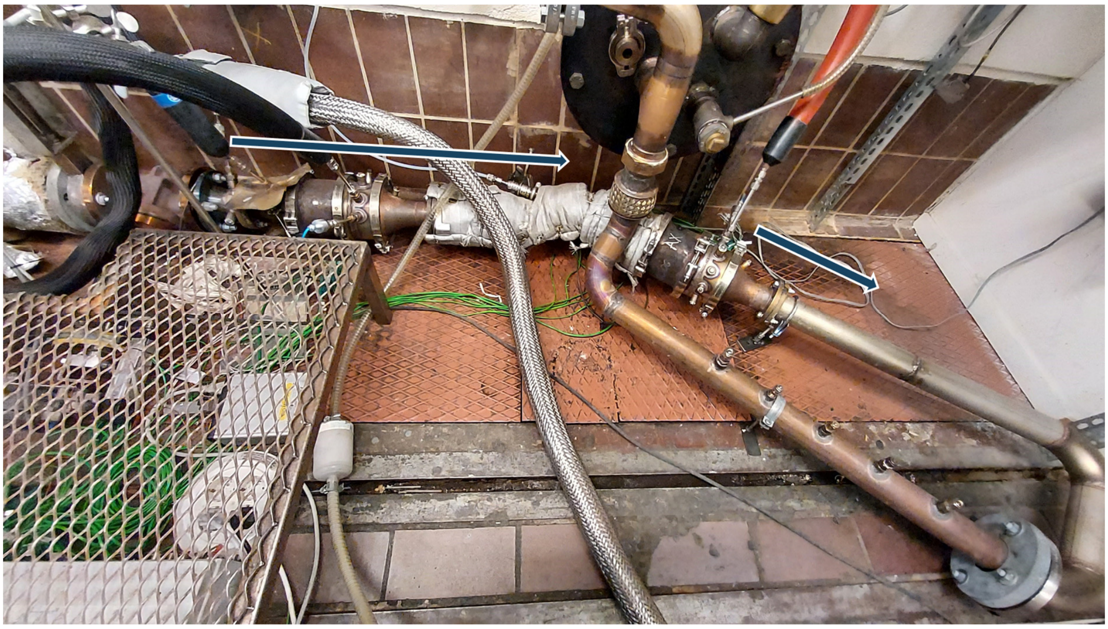

*Figure 6: Complete EATS for the heavy-duty experiments. Arrows indicate the exhaust flow direction.*

The catalyst were provided by Johnson Matthey. The DOC used was a custom-made DOC 40g /ft3 – platinum only on alumina. The SCR was a commercial Cu zeolite catalyst.

Urea was fed to the system by a simple system that used compressed air to pressurize a urea solution. The solution under pressure was fed to an injector, mounted in a production spec urea mixing pipe from a commercial vehicle.

Control of the system was done manually by changing the frequency and duty-cycle of the power supply to the injector. To check the accuracy of the system, different settings for frequency, accuracy and pressure was tested with water. The result of these tests showed promising results. It was decided to stick with the higher injection pressure of 5 bar, as higher pressures should give finer atomization of the pray, analogous to a fuel injector.

The tests were repeated for a commercial urea/water solution and only showed deviations between points for less than 1 % for repeated points. The duty cycle was shown to be the sole factor influencing flow rate, except for very short injection durations.

The urea system was calibrated trough repeated measurements using a scale and measurement of injection duration for different pressures and opening frequencies. More details can be found in the appendix 1 of paper B.

## 2.4 Emission Measurement Systems

Several measurement systems have been used to measure the emissions, see the appended papers. Unsteady $NO_X$ emissions from the engines have required detailed analysis of the precision of the instruments to assess the system performance.

## 2.5 Design of Experiments

### 2.5.1 Light-Duty Engine

For a detailed description of the operating points of light-duty engine experiments see the attached paper, *Characterization of Gaseous and Particle Emissions of a Direct Injection Hydrogen Engine at Various Operating Conditions*. One of the key problems of hydrogen internal combustion engine vehicles, namely that of low volumetric energy density and the associated difficulty of fuel storage made minimising the number of experiments even more crucial than when operating on liquid fuel.

Design of experiments (DoE) is a common term for approaches that aim to produce the most valuable data from as few experimental runs as possible. As the hydrogen engines used were completely new systems, the process can be divided into the following steps:

- Identify parameters of interest
- Investigate the limits of said parameters
- Use a suitable design in these parameters

The design chosen was a face centred cubic design, for the run of experiments carried out in the first and second paper. The advantages of the cubic design, as opposed to a central composite design that made it favourable in these cases, is that finding the limits and corresponding load points is easier, with fewer unique combinations of engine settings. It also allows some parameters, that are difficult to keep constant to be more clearly.

As the maximum brake torque (MBT) spark timing was not known, the spark timing was swept for all points.

A design of experiments was made using the parameters that influences the engine-out emissions the most. These were deemed to be λ, load and start of injection. The combustion phasing was swept through spark timing for each load point.

The final run of experiments on the light-duty engine was a sweep of start of injection timing at a constant load of 9 bar IMEP at constant λ of 2, to investigate the effect of SOI on emissions and performance.

### 2.5.2 Heavy-Duty Engine and EATS

For the heavy-duty engine, a DoE approach was taken from the start. Since the goal of these experiments were the investigation of key engine operating parameters on the efficiency of the catalyst, the first objective was to determine which parameters were most influential. A difficulty arises in these kinds of experimental plans, when there is no direct control of the parameters of interest. Table 3 shows a breakdown of the engine parameters and their effect of the exhaust stream that are important for the catalyst. Not included are temperatures, which has significant effect on the catalyst performance, but are not directly controllable.

*Table 3: Engine operating parameters and secondary (catalyst parameter) effects.*

| Engine Parameter | $NO_X$ | Space velocity | Water content |
|---|---|---|---|
| **RPM** | → | ↑ | → |
| **Intake Pressure (lambda)** | ↓ | ↑ | ↓ |
| **MFB 50** | ↑ | → | → |

Given that the stable engine operation was limited to a relatively narrow range of load, speed and SOI, due to abnormal combustion phenomena as well as exhaust back-pressure, the range of settings for the parameters that could be run was limited.

It was decided that instead of manipulating the air/fuel ratio, the intake pressure would be used as the variable of interest. During all the runs, the fuel injected per stroke was to remain constant. Variations on load would thus be caused only by changes in engine efficiency. The resulting design in shown Figure 7 below:

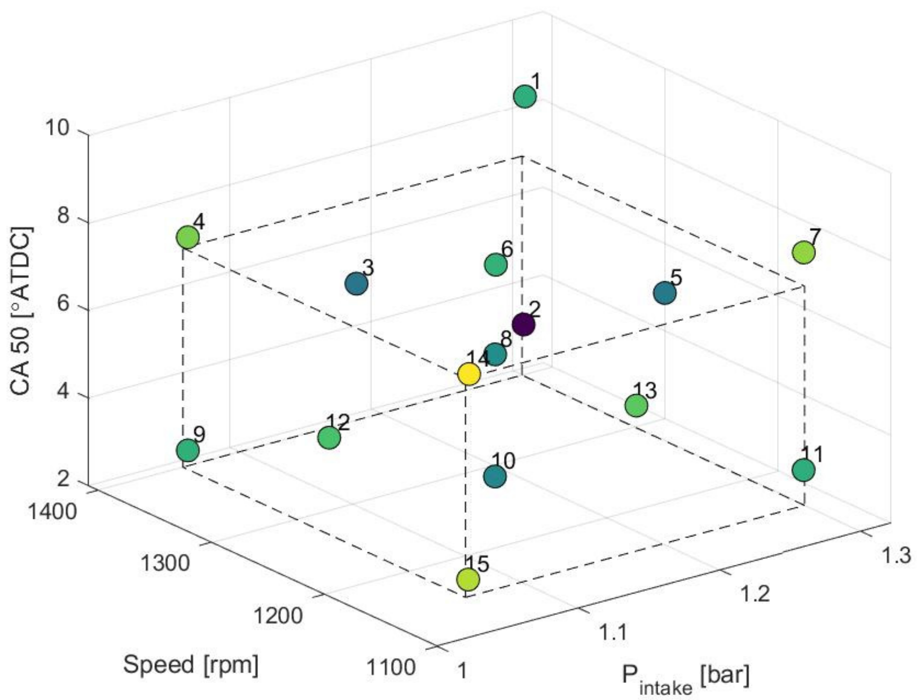


*Figure 7: Face-centred cubic design of experiments for the heavy-duty engine experiments. The experiment point number is shown next to the point.*

The intake pressure was limited to 1300 mbar, which corresponded to a λ of approximately 2.4, as the $NO_X$ emissions at this point was 50-100 ppm. It was deemed unnecessary to go to leaner conditions, as the emission levels, and especially the differences between the upstream and downstream positions relative to the catalysts would become small compared to the range of the instruments. At lower pressures, the limiting factor was the minimum back pressure in the exhaust system. The $NO_X$ emissions at this air/fuel ratio was significant, which rendered it unnecessary to go richer. It was also desirable, to be able to compare the operating point to Results and Discussion

# 3 Results

## 3.1 Summary of paper A

**Characterization of Gaseous and Particle Emissions of a Direct Injection Hydrogen Engine at Various Operating Conditions** [44]

This paper aimed to investigate the effects of operation parameters such as load, speed, injection timing, air- fuel (A/F) ratio and ignition timing on the emissions and performance of a direct injection spark ignition internal combustion engine operated on pure hydrogen. Three experimental campaigns are included in this paper.

The first, an investigation on the effects on the emissions and on the efficiency of the engine over a range of speed and load at a constant A/F ratio. The second campaign was a design of experiments aimed at investigating the combined effects of load, A/F and start of injection at constant engine speed. For both campaigns the spark timing was varied, as the effects of combustion phasing was, and the optimal MFB 50 point was unknown for the engine. Finally, sweeps of injection timing and ignition timing at constant load and speed were performed.

The results of these experimental runs showed that a hydrogen internal combustion engine can be operated in modes that produce very low emissions, but that an exhaust after-treatment system will be required to consistently reach emission limits. The indicated specific $NO_X$ emissions are shown in Figure 8:

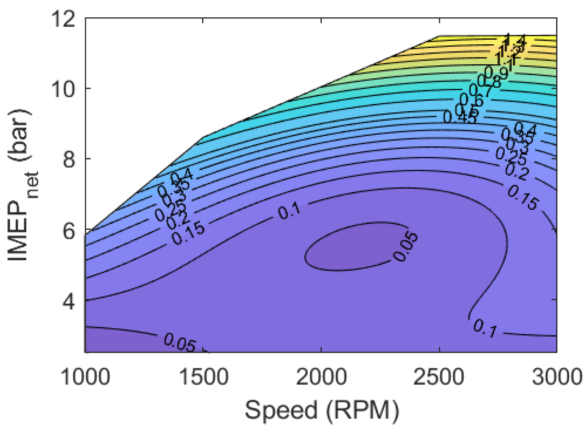


*Figure 8: Indicated specific $NO_X$ emission over the engine map.*

The engine-out $NO_X$ emissions was found to be dependent on air/fuel ratio as expected, however, the engine needed to be operated at very lean conditions. A four-cylinder engine, similar in specification to the light duty engine, equipped with an advanced boosting system reached λ 1.4 at slightly higher load, but at lower intake pressures and significantly higher exhaust temperatures [37]. The engine-out $NO_X$ emissions was found to be dependent on air/fuel ratio as expected, however, the engine needed to be operated at very lean conditions for the emissions to be below the limits, as shown in Figure 9:

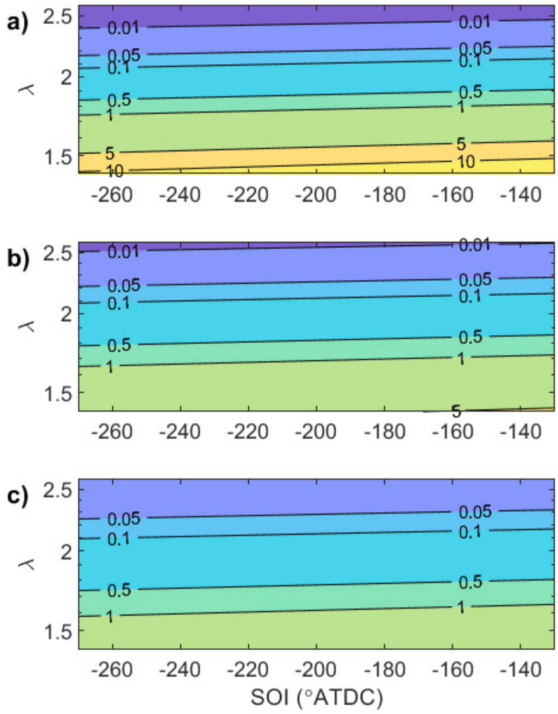


*Figure 9: Indicated specific $NO_X$ emissions [g/kWh] a) 9 bar IMEP, b) 6 abr IMEP, c) 4.5 bar IMEP*

The particle emissions at low loads proved especially problematic, with particle numbers that were far higher than acceptable, especially considering particle emissions at part load. Exhaust gases were sampled on filters and analysed for trace metals as well as fraction of elemental to organic carbon. Through this analysis, the cause of the particle emissions was confirmed to be engine oil. The increased emission at low load, which coincided with throttled operation of the engine could be caused by higher oil flow through intake valve stem seals and past piston rings driven by the sub-atmospheric pressure in during the intake stroke. Figure 10 shows a summary of the particle emissions for all load points of the light duty engine as a function of load. It shows clearly why throttled operation of a hydrogen internal combustion engine should be avoided.

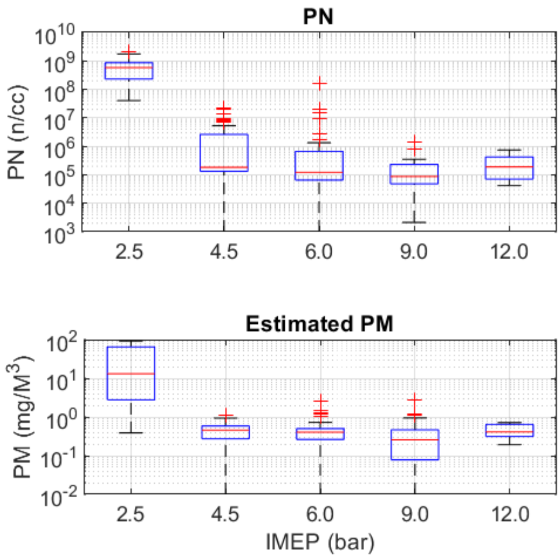


*Figure 10: Particle numbers and particle mass emissions for the light duty engine tests.*

The emissions of the currently unregulated emission were found to be low, and within the error margin of the measurement instruments.

Indicated efficiencies reaching higher than 40 % were possible for a large range of loads and operating conditions.

## 3.2 Summary of Paper B

### Experiments on Hydrogen Engine Exhaust After-Treatment Systems

An investigation into the effect of some of the unique properties of a hydrogen internal combustion engine, including high levels of water int the exhaust gases, on the efficiency of an SCR system similar to commercially available system was performed. A design of experiments approach was used, and face centred cubic design in intake pressure, combustion phasing and engine speed was designed to maximise the spread in $NO_X$ emissions, water content and space velocity, while using the catalyst temperature as an uncontrolled variable.

The engine out $NO_X$ emissions was found to be primarily influenced by the intake pressure, and thus the air flow ratio, and the combustion phasing as intended in the experimental design. The engine speed was shown to have little effect on the levels of $NO_X$ in the exhaust. The influence of combustion phasing and intake pressure is illustrated in Figure 11:

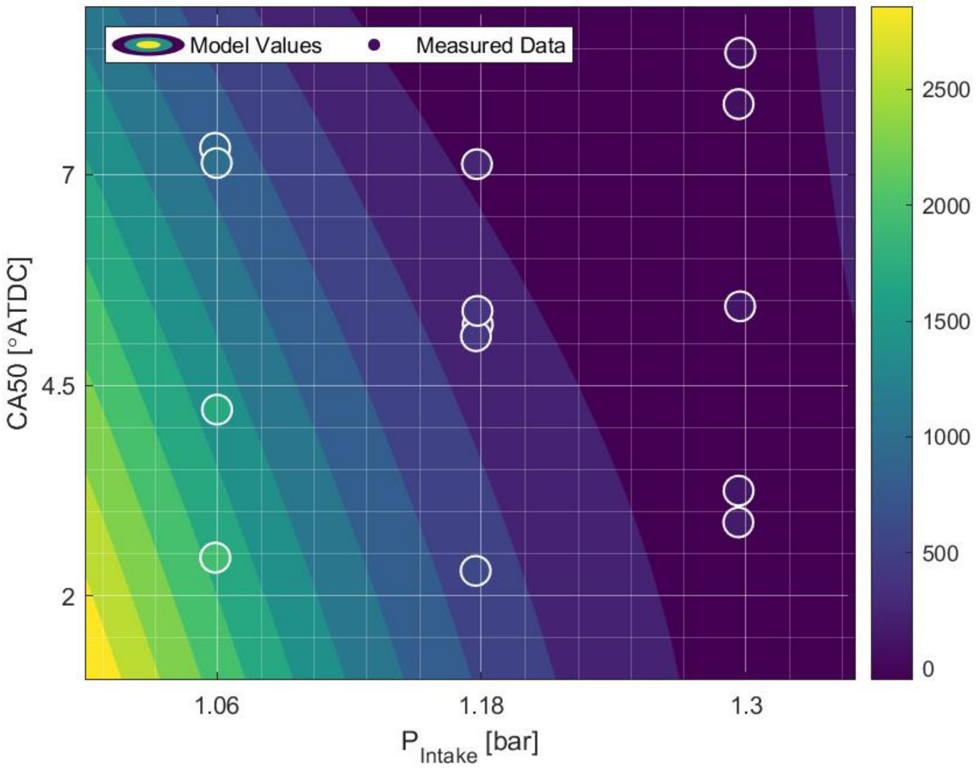


*Figure 11: $NO_X$ emissions, modelled and measured values from the design of experiments.*

The main goal of the design of experiments, apart from the investigation of engine out emissions, was to investigate the impact of water and heat as well as $NO_X$ levels in the oxidising catalyst. The engine however showed large variations in fuel flow over time, which caused the separation of water and $NO_X$ levels to be smaller than intended, as the values would fluctuate over time. A small effect of water levels on the downstream $NO_2$ to $NO_X$ ratio was found, and it seems that water increases the oxidation. The effect was however small, and in is not included in Figure 12. However, the downstream $NO_2$ to $NO_X$ ratio, which is important for the SCR system could still be estimated with decent accuracy, with an R2 of 0.96. The two most influential parameters were found the be temperature, as expected, and total $NO_X$ levels, which indicates that the oxidation of NO to $NO_2$ is rate limited in the catalyst. Curiously, the space velocity did not have any statistically observable influence on the $NO_2$ to $NO_X$ ratio.

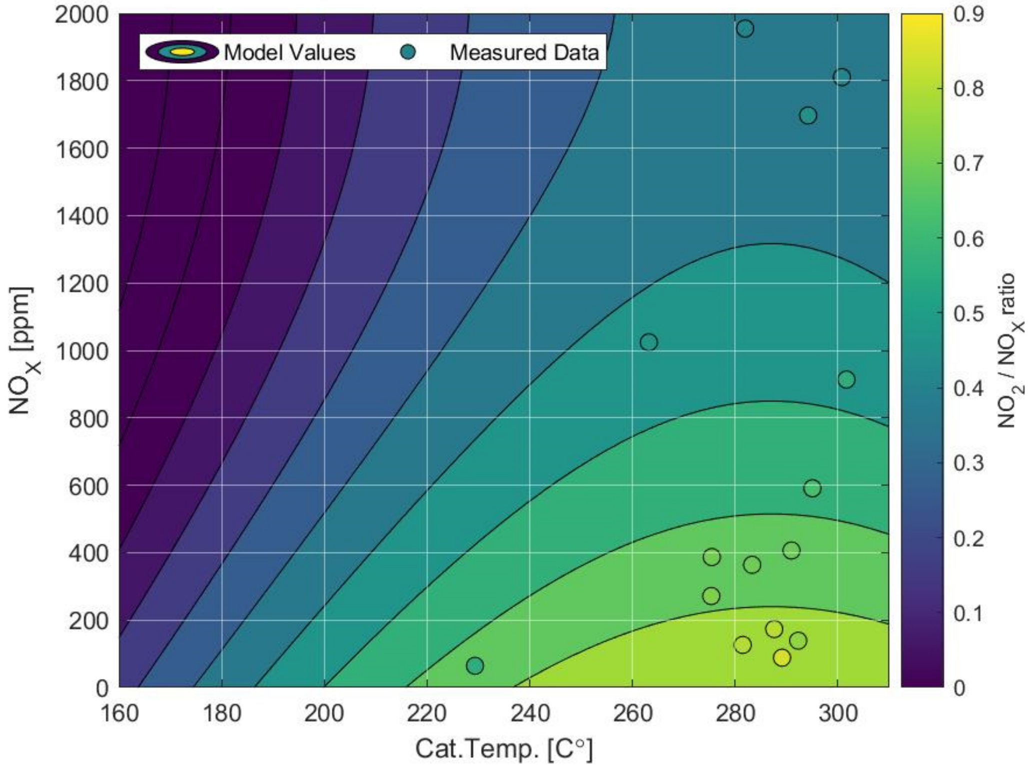


*Figure 12: $NO_2$ to $NO_X$ ratio downstream of the oxidising catalyst.*

The output from this model was used to estimate the $NO_2$ to $NO_X$ ratio the SCR system was subjected to in the SCR runs.

The runs measuring $NO_X$ conversion in the SCR system were run for one hour. For brevity, only the high $NO_X$, high water, high ammonia to $NO_X$ ratio run is shown in Figure 13.

The top graph in the figure shows the emission of nitrogen oxides, measured continuously with the FTIR as well as intermittent measurement with the CLD in the AVL AMA. The second graph shows the measured ammonia slip and $N_2O$ generation in, measured with the FTIR. As the FTIR only has one sample point, it had to be switched between the sampling point downstream of the oxidising catalyst but upstream of the urea injector and SCR catalyst. The switches can be seen in the jumps in the curves, but the sampling points is also shown in the bottom window, where sampling point and ANR is shown. The ANR was calculated from the urea flow and the engine out $NO_X$ emission measured by the AMA and is thus only available for the short periods the CLD was measuring.

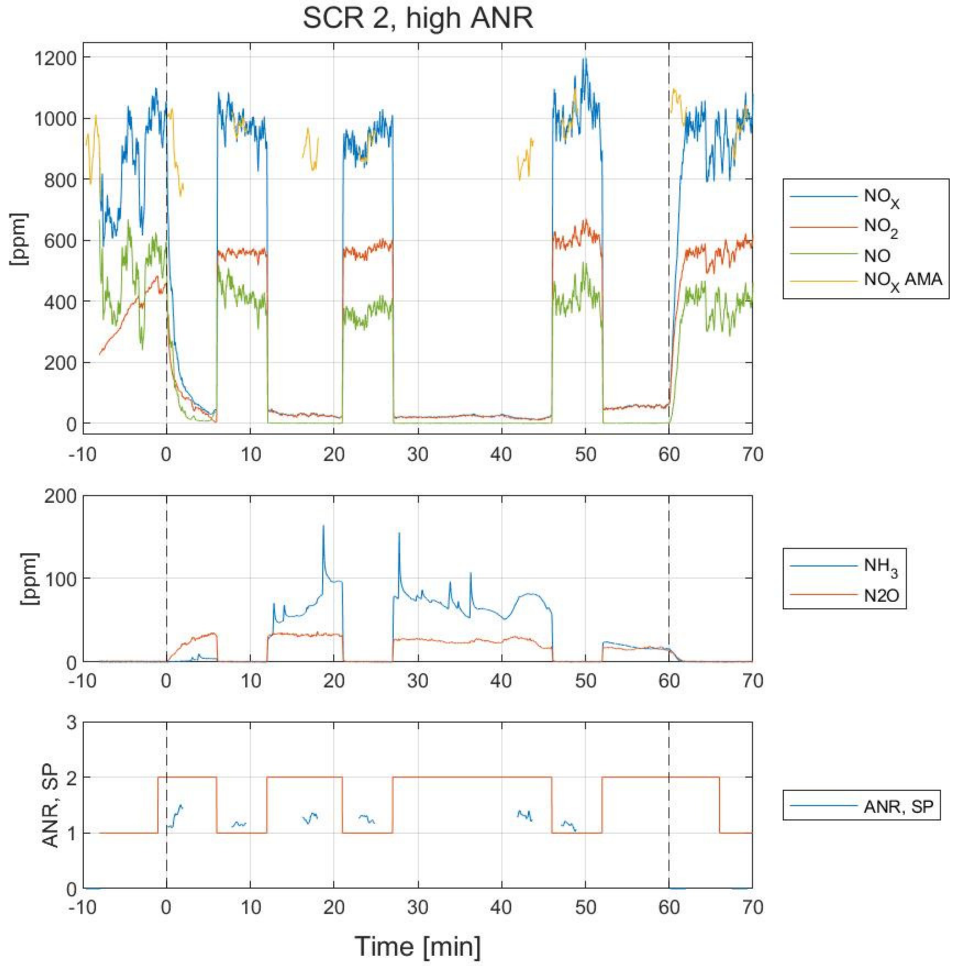


*Figure 13: Nitrous species measured upstream and downstream of the SCR catalyst.*

The results shown that a very high NO conversion can be achieved, albeit at high ANRs. The $NO_2$ however, is harder to reduce, especially in the absence of NO, as the only pathway to reduction is the slow reduction mechanism.

The downside of the high ANR is the ammonia slip that occurs and is seen in the middle curve. The ammonia slip can be seen to increase until around 20 minutes into the measurement, at which point it remains relatively stable. The decreased ammonia slip at the end of the period is most likely caused by the increased engine-out $NO_X$ emissions, as more ammonia is being consumed in the reduction reactions. It does however indicate a problem with the setup, namely that the crude ammonia injection system is hard to adapt to variations in $NO_X$ emission over time, and this instability in $NO_X$ levels proved to be an obstacle to accurate evaluation of the water concentrations as well as ANR on the efficiency of the SCR system.

The unstable $NO_X$ emission exaggerated the confounding between the $NO_X$ and water concentrations, causing their impacts of water on the systems efficiency to be impossible to separate, trends cold still be observed in the data. The total $NO_X$ reduction efficiency as a function of the most important parameters is shown in Figure 14:

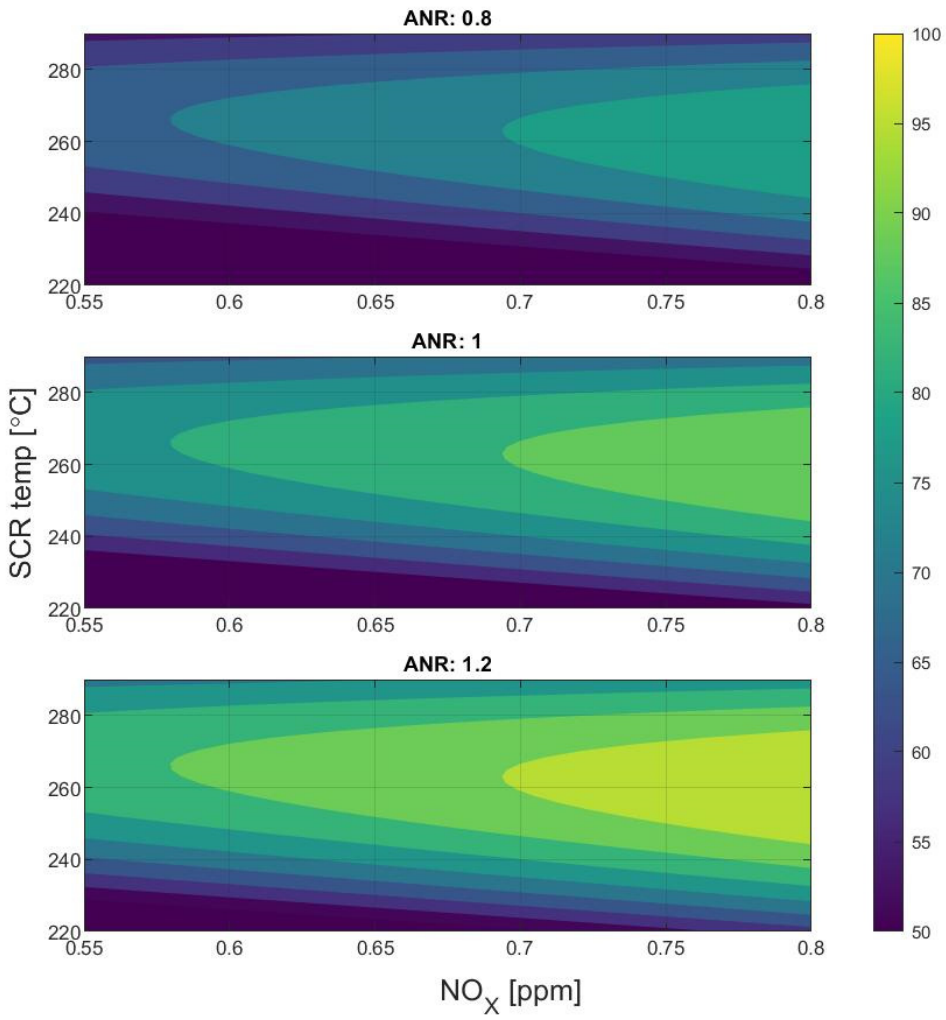


*Figure 14: $NO_X$ conversion efficiency of the SCR system*

The $NO_X$ efficiency improves with higher $NO_X$ levels and or higher water content, as those factors are strongly correlated. Inclusion of water in the model, however, leads to a model with very high condition number, indicating a very sensitive model. It was still shown that a high $NO_X$ reduction could be achieved for high urea injections, as would be expected. While the $NO_2$ to $NO_X$ ratio is expected to impact the rate the reactions and thus the efficiency of the system, no statistically significant impact was found on the total conversion efficiency. The NO and NO2 reduction however, showed an impact from the ratio, with a high ratio improving NO efficiency and a low ratio facilitating high $NO_2$ efficiency.

## 3.3 Discussion

### 3.3.1 Importance of stable $NO_X$ emissions

The SCR measurements, as shown in Figure 13, shows unstable levels of $NO_X$ upstream of the catalyst. This causes unsteady emissions and leads to poor control of the ANR over time. This in turn leads to poor control over the amount of ammonia stored in the catalyst and is an obstacle to comparing the results and evaluating how the NOX conversion efficiency of the catalyst is affected by the conditions upstream, including any impact of water content etc. that was part of the research questions.

Investigating the causes of the variations in engine out $NO_X$ levels shows that the fuel flow rate of the engine is a most likely cause of the fluctuations in $NO_X$. The first issue that was found was periodic fluctuations in pressure in the fuel line. This gave rise to a saw-tooth shaped $NO_X$ fluctuation. Modifications to the fuel system and the control system managed to reduce the pressure fluctuations to a very low level, but some variations in fuel flow persisted, and appeared to be time change over longer time periods. To verify this, the setup of the AVL measurement system was modified to allow longer time series to be measured. A figure showing how the fuel pressure and $NO_X$ varies over time, and over a step in intake pressure, is shown in Figure 15:

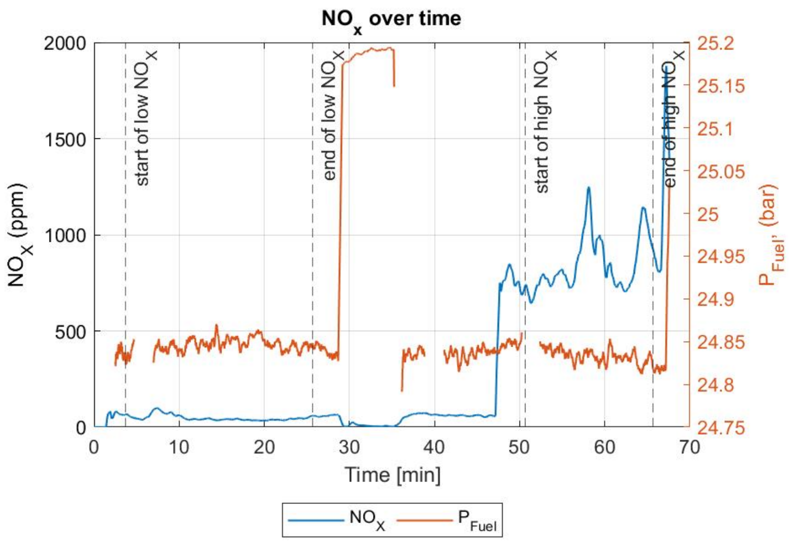


*Figure 15: Fuel pressure and engine out $NO_X$ emissions. The step in $NO_X$ emissions at approximately 13:10 is due to change in intake pressure. Two periods chosen for further analysis is indicated by the vertical lines and labels.*

It is evident from the figure that the intake pressure is relatively stable over time, with maximum changes in the range of ±0.05 bar, corresponding to a relative change of only 0.2 %. It seems unlikely that the fuel pressure regulation should be the cause of the variations. A closer look at the one of the two sections indicated in Figure 15, including fuel flow measurements from the Coriolis fuel flow meter is shown in Figure 16

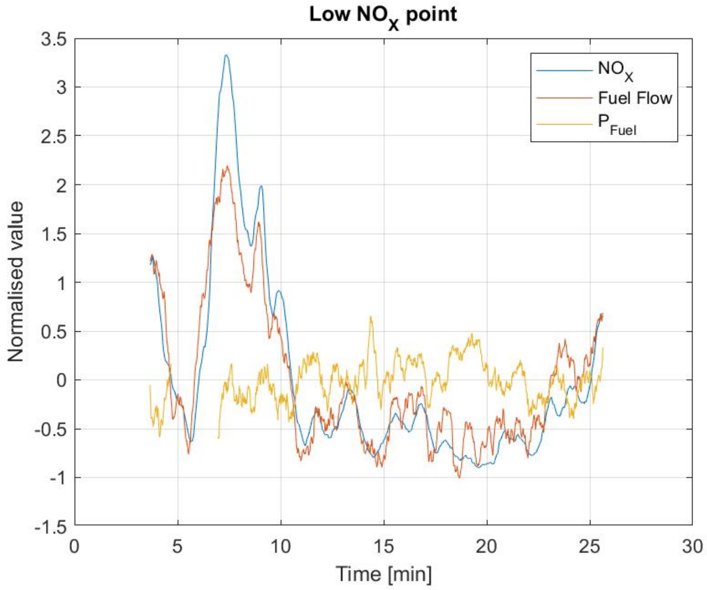


*Figure 16: Normalized $NO_X$ emissions, fuel flow and fuel pressure for the low $NO_X$ section*

It is evident that the changes in $NO_X$ emissions over time corresponds to changes in fuel flow. The fuel flow, however, shows no correlation with the fuel pressure in the rail, which was the earliest hypothesis for the flow variations. Significant effort has been made to achieve stable fuel flow, but the root cause is yet to be found. The results are similar for the high $NO_X$ sections, which is not shown here.

### 3.3.2 Discussion on the Limitations of Operating Points

The heavy-duty engine used in the lab was limited in the range of operating points that could be run at the beginning of the experimental campaign. Maximum load was limited by prevalent pre-ignitions to around 7 bar IMEP, while the maximum engine speed was limited to 1400 rpm. Severe combustion instability at low engine speeds limited the engine to speeds over 1150 rpm.

Further constraints were imposed by less severe, but still important factors. In trying to keep the load constant (and as high as possible) the minimum air/fuel ratio that could be reached without imposing high pumping losses was limited by the exhaust back pressure. The main reasoning for the avoidance of throttled operation at this load was that throttling would impose an unnecessary (and obvious) efficiency penalty, and this mode of operation would be avoided.

The maximum advance of the spark timing was limited such that the minimum combustion phasing was 2° ATDC, to ensure that all operating points could be run with a small margin to knock.

The $NO_X$ emissions was also a limiting factor. As combustion phasing was retarded and the intake and exhaust pressure were increased, the $NO_X$ emissions were reduced. Any later combustion phasing or higher pressures would result in near-zero $NO_X$ emissions which, while desirable, renders investigation of the effect of e.g. water content in the exhaust gases and their impact on catalyst efficiency impossible.

### 3.3.3 Discussion of SCR Performance

Fuel flow proved to be very large cause of the variations. For the experiments presented in the paper B, some of the variations could be explained by the fuel pressure changing. Updates to the fuel supply did however not remove the variations completely, and fluctuation s in both $NO_X$ emissions and fuel flow persisted. After extensive searching for the error sources, the fuel injector and/or driver was identified as the cause.

The unsteady fuel flow, and the associated unsteady levels of $NO_X$ in the exhaust made separating the effects of $NO_X$ and water difficult in practice. If the engine-out emissions would have been stable, the difference in total $NO_X$ due to the combustion phasing would still have been smaller than the impact on $NO_X$ of the air/fuel ratio.

The variations also introduced an additional difficulty in that time synchronization of data turns into a potentially severe source of error

### 3.3.4 Discussion on Particle Measurements

Particle measurements using the DMS 500 have been met with very large variations between repeated tests at similar engine operating parameters. This held true for both the light-duty and heavy-duty engine used in the research presented here. The variation between the repetition presented and obstacle to evaluating the effect of different engine setting on PN emissions.

The spark plug also showed evidence of excessive oil consumption, in the form of oily black deposits. Figure 17 shows a photograph of the spark plug, which was removed after the light-duty engine tests.

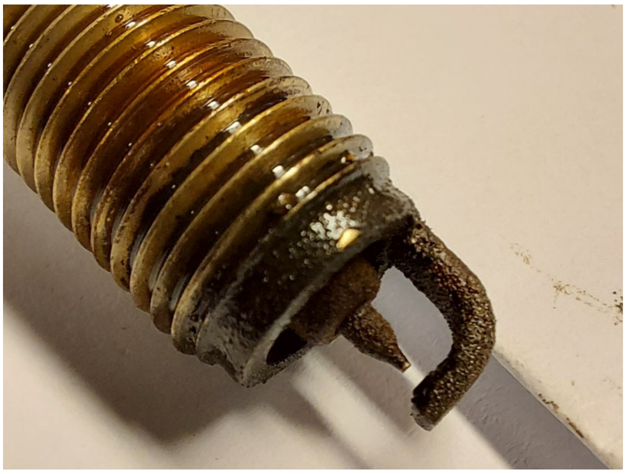

*Figure 17: Spark plug from the light-duty engine, showing fouling by engine oil.*

The spark plug would be expected to be clean as there should be no components in the fuel could give rise to the deposits. However, the spark plug shows signs of oil fouling, indicating that a significant amount of engine oil was presented in the cylinder during the light duty engine tests.

An alternative approach for investigating operating parameters and their effect on the heavy-duty engine could be to perform steps in the parameters and observing the response. The steps could then be repeated multiple times in a relatively short duration, at different starting levels of PN. This could, potentially, give an insight into the impact of one engine parameter at a time on the emissions. The other approach is to simply repeat the measurements more times. The history effect on the PN will however necessitate a long sampling period at each point.

# 4 Conclusions

The conclusions can be separated into two parts. Conclusions from paper 1

- An engine converted to hydrogen combustion engine can reach indicated efficiencies greater than 40% without significant modification.
- The unregulated emissions (engine out) that can be measured trough FTIR are very low, and around the detection limit.
- The particle emissions increase drastically at throttled operation, possibly due to the increased oil inflow into the cylinder caused by the pressure difference over piston rings and valve stem seals during the intake stroke.

The results described in the technical report shows that:

- The inclusion of an oxidation catalyst in a hydrogen exhaust after-treatment system can be problematic due to very high NO to $NO_2$ oxidation at low temperatures.
- The engine out $NO_X$ emissions of the heavy-duty engine showed very large sensitivity to the injected fuel amount, with changes of a few percent almost doubling the $NO_X$ emissions.
- The conversion efficiency of the SCR catalyst in reducing NO and $NO_2$ depends on the upstream $NO_2$ to $NO_X$ ratio.
- The total NOX reduction was found to be dependent on temperature and ANR, with a small sensitivity to total $NO_X$ or water levels. The concentrations of water and $NO_X$ in the exhaust is too high to be able to tell which is mor important from the data.
- High $NO_X$ reduction could be achieved for high urea injections, as would be expected.
- $N_2O$ was created and ammonia slipped past the catalyst at high ANRs.

The potential for low emissions of LPDI hydrogen engine have been shown at steady state. The research presented in this thesis has focused on the reduction of the emissions that are produced, but the trends in e.g. $NO_X$ as a function of air/fuel ratio indicate that there are operating conditions where the engine-out emissions may be below future limits.

# 5 Future Work

In the future of this research project, two areas need more attention. One is the composition of the particle emissions. Some analysis of the particle emissions from a light duty -engine was performed and presented in the first publication. This confirmed that the source of the emissions was the engine lubricating oil. Future work will involve analysis of the engine parameters that affect the PN emissions and means to minimize these emissions. Furthermore, more detailed analysis of the chemical composition of the particles at different particle size ranges using e.g. TOF-SIMS [49] will be performed.

There are also plans for analysis of deposits formed on the DOC, that could give information regarding the particle buildup and/or damage due to the high water concentration in the exhaust gases.

Finally, the SCR catalysts will be investigated further. The influence of the unique characteristics of hydrogen combustion on the exhaust gases, and their influence on the efficiency of the SCR system will be investigated for different, commercially available catalysts chemistries. A heated, uncoated catalyst will be used in place of the oxidising catalyst together with a water injection system. This will enable independent control of temperature, $NO_X$ and water in the exhaust.

The works presented here has however only been performed on steady state conditions, and especially under transient engine operations, the role of an exhaust-aftertreatment system would be crucial.

## 5.1 Outlook

The results from this thesis show promising results from hydrogen LPDI engine and the use of an EATS.

The tailpipe emissions could probably meet the EURO 7 legislation using current technologies.

Other challenges for the broad implementation of hydrogen engines include:

- Infrastructure: supply of hydrogen at filling stations
- Safety: implementation of robust safety systems for on road usage
- $N_2O$ will not be a problem using pure hydrogen, but other concepts (e.g. dual fuel concepts, including ammonia) may create emissions of $N_2O$ that may be a concern.

To reach the climate goals, fossil fuels need to be faced out as soon as possible, and hydrogen is a viable, affordable and possible energy carrier for transportation. This will require drastic changes in society and the energy system. Hydrogen engines will hopefully contribute to one piece in this transition.

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# Abbreviations

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|---|---|
| ANR | Ammonia to $NO_X$ Ratio |
| ASC | Ammonia Slip Catalyst |
| ATDC | After top dead centre |
| DOC | Diesel oxidation catalyst |
| DPF | Diesel Particulate Pilter |
| EATS | Exhaust after-treatment system |
| EGR | Exhaust Gas Recirculation |
| HPDI | High pressure direct injection |
| IMEP | Indicated Mean Effective Pressure |
| LPDI | Low pressure direct injection |
| MFB | Mass fraction burned |
| MBT | Maximum Brake Torque |
| PFI | Port-fuelled Injection |
| SCR | Selective catalytic reaction |
| SI | Spark ignition |
| SOI | Start of Injection |
| TTW | Tank to Wheel |
| WLTC | Word-harmonized Light-vehicles Test Cycle |
| WTW | Well to Wheel |

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