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ABATEMENT OF NOX EMISSIONS FROM VEHICLES

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INDEX

<u>1. INTRODUCTION</u>	3
1.1. SCOPE OF REPORT	5
<u>2. NO_x EMISSION CONTROL</u>	5
2.1. INTRODUCTION	5
2.2. EXHAUST GAS RECIRCULATION (EGR)	7
2.3. SELECTIVE CATALYTIC REDUCTION (SCR)	8
2.4. LEAN NO _x TRAP (LNT).....	11
<u>3. REAL WORLD EMISSIONS</u>	12
3.1. INTRODUCTION	12
3.2. NITROGEN OXIDES (NO _x)	13
3.2.1. CARS.....	13
3.2.2. LIGHT DUTY COMMERCIAL VEHICLES (VANS).....	14
3.2.3. HEAVY GOOD VEHICLES (HGVS).....	14
3.2.4. BUSES	16
3.3. NITROGEN DIOXIDE (NO ₂) EMISSIONS.....	17
3.3.1. INTRODUCTION	17
3.3.2. LIGHT DUTY VEHICLES	18
3.3.3. HEAVY DUTY VEHICLES	19
3.4. AMMONIA (NH ₃) EMISSIONS	19
<u>4. RETROFITTING BUSES</u>	20
<u>5. RECOMMENDATIONS</u>	21
5.1. LIGHT DUTY VEHICLES	21
5.2. HEAVY DUTY VEHICLES.....	22
5.3. NO ₂ EMISSIONS	22
5.4. EMISSION FACTORS	22
5.5. RETROFITTING.....	22
<u>6. SUMMARY AND CONCLUSIONS</u>	23
<u>7. REFERENCES</u>	25

1. INTRODUCTION

Most EU Member States (MSs) have locations which exceed the annual mean limit value for nitrogen dioxide (NO₂), with the highest concentrations occurring close to busy roads. MSs were obliged to meet this Limit Value by 2010, but according to the European Environment Agency (EEA, 2014) 20 MSs recorded exceedances at one or more monitoring stations in 2012. While the Limit Value was exceeded at only one rural background station and 2 % of all urban background stations, it was exceeded at 37 % of traffic stations, with a maximum annual mean concentration of 94 µg m⁻³, nearly two and a half times the limit value. The EEA concluded that NO₂ concentrations need to be substantially reduced for the annual Limit Value to be met.

Progress towards meeting this limit value has been more challenging than originally envisaged and a time extension for compliance up to 2015 has been requested by most MSs. Some cities, however, are unlikely to achieve the limit value until a decade or more after this date. In the United Kingdom (UK), for example, official estimates suggest that compliance in London, West Yorkshire and the West Midlands will not occur until after 2030 (Defra, 2014).

The principal reason for the lack of compliance is that road transport nitrogen oxides (NO_x) emissions have not declined as anticipated by the progressively more stringent vehicle emission limits introduced in Europe over the last two decades. Velders et al. (2011) for example, have estimated that in the Netherlands the higher than expected emissions from heavy duty vehicles have more than doubled the total road length with possible exceedance of the ambient NO₂ limit value; from approximately 100 km to 250 km along cities streets and motorways, by 2015.

Another contributory factor is the increase in the proportion of the vehicle NO_x emissions in the form of NO₂. This is thought to be due to a combination of increased diesel cars in the vehicle fleet and the use of certain exhaust after-treatment devices that increase NO₂ in the exhaust. The average proportion of new diesel cars sold in the European Union (EU) has increased from approximately 20% two decades ago to 50% in 2013 (ACEA, 2014), while the increasing use of diesel oxidation catalysts and particle filters on diesel vehicles has increased the proportion of the NO₂ (f-NO₂).

An increasing f-NO₂ in vehicle exhaust is an important factor influencing ambient NO₂ concentrations close to roads and can have a large effect on the exceedence of the NO₂ limit values in these locations (Carslaw & Rhys-Tyler, 2013).

Figure 1 shows the NO_x emissions from Euro 2¹ to Euro 6 gasoline and diesel passenger cars obtained using remote sensing data from Leeds in north east England (Tate, 2014). It clearly shows that NO_x emissions from gasoline cars have declined and that for the more recent cars the variation between cars built to the same EU emissions standard is small. This confirms earlier data from London (Carslaw & Rhys-Tyler, 2013) that showed that emissions from Euro 5 gasoline cars are approximately a factor of 20 lower than pre Euro 1 gasoline cars. It can be concluded that the emissions from modern gasoline cars are well controlled.

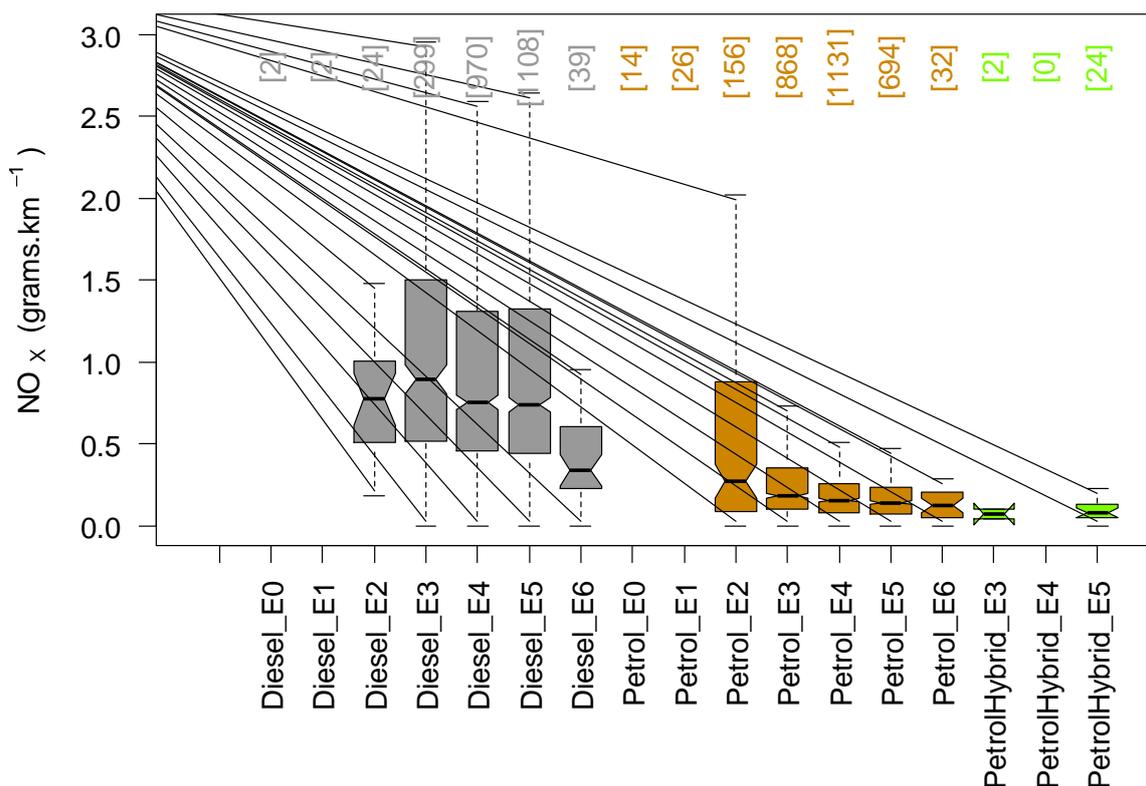


Figure 1-1 Box and whisker plot of diesel and gasoline passenger car NO_x emissions according to EU emission standard measured using remote sensing in Leeds, UK. In the shaded areas the horizontal line shows the median; the top and bottom of the shaded area show the 75th and 25th percentile respectively; the horizontal lines at the end of the dotted lines (whiskers) show the approximate 95th confidence limits (Tate, 2014)

Conversely Figure 1 shows that diesel car emissions have been poorly controlled with large variations between cars built to the same standard and between Euro 2 and Euro 5 there is little evidence of an improvement. Carslaw & Rhys-Tyler (2013) have shown that Euro 4 and 5 diesel cars have higher NO_x emissions than pre Euro 1 and Euro 1 diesel cars. There is

¹ Light duty vehicle emissions standards are denoted with Arabic numerals (1, 2, 3 etc.) and heavy duty engine emissions standards by Roman numerals (I, II, III etc.). Euro 1/I standards were first implemented in 1992, with the most recent Euro 6 and VI implemented from 2014 and 2013 respectively.

limited data that Euro 6 diesel cars have lower emissions, possibly 40% lower (Carslaw, 2014), but still well in excess of the type approval emission limit. Emissions of NO_x from Euro 5 gasoline cars are on average a factor of 10 less than the equivalent diesel cars (Carslaw & Rhys-Taylor, 2013).

A study from Zurich, in which annual remote sensing measurements were made at the same site between 2000 and 2012, concluded that NO_x emissions from diesel cars have increased over this period, despite the tightening of emissions standards. The NO_x emissions from pre-Euro 1 diesel cars were slightly lower than those from Euro 5 cars, with the highest real-world NO_x emissions from Euro 2 and 3 cars (Chen & Borcken-Kleefeld, 2014).

Carslaw & Rhys-Tyler (2013) measured the emissions from over 10,000 diesel passenger cars using remote vehicle emissions sensing and concluded that both engine size and manufacturer are important determinants of NO_x emissions. Carslaw et al. (2013) showed that NO_x emissions are also dependent on vehicle specific power. Diesel vehicles have become more powerful over recent years, increasing on average in the EU from approximately 77 kW to 97 kW between 2001 and 2013. Over the same period gasoline cars increased from 73 kW to 80 kW (International Council on Clean Transportation, 2014a).

Carslaw & Rhys-Tyler (2013) also showed that the trend in NO_x emissions from light duty commercial vehicles, which are predominantly diesel fuelled, and heavy duty vehicles (HDVs) have also shown little improvement from pre Euro 1/I to Euro 5/V.

1.1. Scope of Report

This report focuses on NO_x emission abatement technologies used on light and heavy duty diesel vehicles as it is clear from the above that gasoline cars have well controlled NO_x emissions. Commercial vehicles and buses are predominantly diesel fuelled.

2. NO_x EMISSION CONTROL

2.1. Introduction

If complete combustion of the fuel in an engine were possible vehicle exhaust would contain only carbon dioxide (CO₂) and water vapour. However, in reality vehicle exhaust also contains particulate matter (PM) and a range of other compounds depending on the fuel. Additionally NO_x is formed by oxidation of atmospheric nitrogen (N₂) at the high temperatures in an engine.

Controlling vehicle emissions has been an evolutionary process of gradual improvement to the combustion process, engine management system and, for gasoline cars catalytic after-treatment, over many decades. Emissions were initially reduced by controlling the formation

of pollutants in the engine, but as emission limits became more stringent the manufacturers have increasingly had to also remove pollutants in the engine exhaust using after-treatment devices.

There is a compromise during engine design between optimising for fuel consumption, emissions and performance. For diesel engines the emissions trade-off is between PM, NO_x and CO₂ emissions. Engine conditions with low PM and CO₂ emissions tend to be when high NO_x emissions occur, and vice versa.

For gasoline vehicles the use of three way catalysts (TWC) has been standard since the Euro I emission standard was introduced in 1992. These devices effectively control the NO_x, carbon monoxide and hydrocarbon emissions in the exhaust, but only within a very narrow ratio of air to fuel in the engine. This means that it is more difficult to reduce CO₂ emissions from a conventional gasoline vehicle.

Diesel engines are more fuel efficient because they use a higher air to fuel ratio (i.e. a leaner mixture). There is too much oxygen (O₂) in the exhaust for a TWC to control the NO_x emissions, and therefore different technologies have had to be developed. Reducing PM and NO_x emissions from diesel vehicles, while also reducing CO₂ emissions, has been one of the greatest challenges for the modern motor industry.

To reduce CO₂ emissions from gasoline vehicles direct injection (GDI) engines have been developed which also run on leaner air to fuel ratios than conventional gasoline engines, and have emission more similar to diesel engines. TWC also cannot be used with these engines.

The use of sophisticated electronically controlled fuel injection systems has allowed much more precise timing of when the fuel enters the combustion chamber and has resulted in the manufacture of engines with increased power, lower fuel consumption and lower emissions. They have also facilitated the use of GDI engines which require very complex engine management systems to manage the fuel injection under all operating modes.

Exhaust gas recirculation (EGR) has been used for several decades in diesel engines, but has become increasingly sophisticated as NO_x emissions limits have reduced. EGR combined with engine improvements is considered capable of meeting the Euro 6 NO_x emission limits for passenger cars (International Council for Clean Transportation, 2014b), but not the recent HDV emission standards.

There are two after-treatment systems to control NO_x emissions in commercial use; selective catalytic reduction (SCR) and lean NO_x traps (LNT)². Each has advantages but also disadvantages. In general, from Euro IV SCR has been used on HDVs. For passenger cars

² Also known as NO_x Adsorbers Catalysts (NACs)

EGR and/or SCR or LNT are used to meet the Euro 6 standards. These abatement techniques are described in the following sections.

To reduce PM emissions diesel vehicles have been fitted with oxidation catalysts for many years. These devices remove the soluble organic fraction of the particles, but to reduce PM emissions further diesel particle filters (DPFs) are required. DPFs are standard equipment for new diesel cars in the EU but have not been required to meet the HDV emission standards until Euro VI. However, a large number of HDVs have been retrofitted with DPFs, particularly those that operate in urban areas.

The most common type of DPF fitted to HDVs is the continuously regenerating trap (CRT[®]). It was first used commercially in 1995 and retrofitted to buses in Sweden, and then in London and Germany. The filter needs to be regenerated periodically to avoid the buildup of back pressure in the engine. In the presence of O₂ regeneration occurs when the temperature exceeds approximately 550°C. However, diesel exhaust temperatures are generally considerably lower. To reduce the regeneration temperature to within the normal exhaust temperature range NO₂ is used. This, however, increases the f-NO₂ in the exhaust.

For this reason a number of cities have retrofitted buses with both CRT and SCR (known as SCRT) that allows filter regeneration without increasing NO₂ emissions. For example, the Transport Authority of Barcelona (TMB) retrofitted 411 Euro II and III diesel buses with 'low NO₂ SCRT'. These systems have also been used in the UK, Belgium, and Scandinavia.

For cars and vans passive regeneration of the DPF takes place automatically when the vehicle is driven on motorways or other fast roads when the exhaust temperature is high. Under other driving conditions, when the PM loading on the DPF is approximately 45%, the vehicle's engine management system will inject fuel into the engine after combustion to increase the exhaust temperature. If the journey is too short to complete regeneration a warning light will come on to show that the DPF is partially blocked. Typically driving for 10 minutes above approximately 65 km h⁻¹ regenerates the DPF.

2.2. Exhaust gas recirculation (EGR)

For many years the main method for reducing NO_x emissions from both heavy and light duty diesel engines was EGR. NO_x emissions are mainly produced at the high engine temperatures generated when the vehicle is operating under load. EGR returns a proportion of the exhaust gas to the combustion chamber, reducing the amount of O₂, which reduces the combustion temperature and hence NO_x emissions. This, however, increases the fuel consumption. In some EGR systems the exhaust gas is cooled to further reduce NO_x emissions.

The efficiency of the EGR depends on the amount of recirculated exhaust gas and its temperature. Disadvantages of EGR include the increase in engine-out PM emissions (if a DPF is installed it needs to be regenerated more frequently) and higher CO₂ emissions. Possibly of greater significance is that it principally reduces NO_x formation during low load

operation and not during real-world high-load events (Zheng et al., 2004). Therefore there is concern that Euro 6 diesel cars that rely solely on EGR to control NO_x will have high emissions under motorway driving conditions.

2.3. Selective catalytic reduction (SCR)

The main after-treatment used to reduce NO_x emission from diesel engines is selective catalytic reduction (SCR). It continuously removes NO_x by creating a rich microclimate where NO_x is converted to N₂ using ammonia (NH₃), while the overall exhaust remains lean. NH₃ is produced from an aqueous solution of urea stored in a tank on-board the vehicle. The urea solution is injected into the exhaust stream upstream of the SCR where it forms gaseous NH₃ which is stored on the catalyst. The NO_x from the engine reacts with the stored NH₃ to produce N₂ and water.

Since 2008, when the first generation of vehicles fitted with SCR were introduced, a distribution network for urea solution, known as diesel emission fluid (DEF) or AdBlue® has developed. On-board systems alert HDV drivers when to re-fill the DEF tank. For light duty vehicles the tank is refilled during the annual service. The SCR system needs to be tuned to ensure that little NH₃ passes through the catalyst and is emitted from the vehicle. This is known as NH₃ slip.

There have been substantial advances in SCR technology over recent years but there remain a number of challenges particularly related to its performance under low-temperature conditions and the need to precisely match urea injection with NO_x emissions (Johnson, 2014).

The temperature of diesel exhaust gas is relatively low. To convert urea to NH₃ requires the exhaust gases to be above approximately 190 °C and the SCR at a slightly higher temperature to begin to control NO_x, but the system is more efficient at higher temperatures. The engine needs to be warm before the SCR starts to control the NO_x emissions, which at low vehicle speeds may take many minutes. In addition, when a vehicle is idling the exhaust temperature may be as low as 100 °C. SCR systems do not work well during short urban trips with low engine loads. As EGR does work well under these conditions, a SCR combined with EGR may be necessary for Euro 6 cars to have low in-service emissions under all driving conditions.

SCR catalysts were typically based on vanadium pentoxide (V₂O₅). These catalysts were relatively low cost but originally had poor low temperature performance. This has been improved by optimising the proportion of NO₂ in the exhaust to approximately 50% of the NO_x using an oxidation catalyst ahead of the SCR catalyst (Johnson, 2008).

According to Görsmann (2015) copper-zeolites and iron-zeolites³ are beginning to be used to produce better NO_x reduction, particularly when used with a DPF, as they are more stable at the high temperatures produced during DPF regeneration. Cu-zeolites show little sensitivity to the NO₂/NO_x ratio, tolerate high NH₃/NO_x ratios, have low NH₃ emissions, good low temperature activity and thermal stability, but there are concerns that Cu-zeolites may increase the emissions of nitrous oxide (N₂O), a potent greenhouse gas. Fe-zeolites have poorer low temperature performance and also require the f-NO₂ in the exhaust to be increased using an oxidation catalyst. Conversely, they have better high temperature performance and are more resistant to sulphur (S) in the fuel or lubricants than Cu-zeolites. There is no ideal catalyst. Daimler, for example has chosen to use Fe-zeolites to meet the Euro VI emission limits for their engines, whilst other engine manufacturers have chosen Cu-zeolites. In the future, however, manufacturers may choose to use both types of zeolites together to take advantage of their different characteristics.

Good NO_x reduction rates can be achieved under steady-state engine conditions, and initially SCR systems used a fixed ratio of DEF to NO_x. It is much more difficult to control NO_x emissions under the transient conditions typical of urban driving. This requires good control of the amount of DEF injected to provide a high NO_x reduction rate while keeping emissions of NH₃ to a minimum. The first generation DEF control systems used a NO_x sensor downstream of the SCR to control the rate of injection. However, during transient operation there was too much time delay in the response of the system to effectively control the emissions. Second generation controls have been developed to reduce this response time. For example, a NH₃ sensor may be placed in the middle of the SCR to allow a rapid increase in DEF dosing until NH₃ is detected by the sensor. This results in faster SCR control and higher NO_x conversion efficiency, while minimising the risk of NH₃ emissions. However the time lag remains of the order of a minute. It should be noted that it is not currently possible to detect the amount of NH₃ stored on the catalyst using on-board sensors. In addition, according to Skaf et al. (2014) NH₃ interferes with NO_x sensors. Therefore on-board measurements of NO_x in the exhaust may reflect high NH₃ emissions as well as NO_x.

In the future it is likely that there will be increasing integration of engine and after-treatment control systems to optimise the performance of the SCR system and enable fuel economy benefits through increased engine-out emissions but low post SCR NO_x (Stanton et al., 2013). Thermal management of after-treatment devices through engine management will become an essential element of NO_x control to enable SCR to operate at peak efficiency over a wide range of driving conditions.

³ Zeolites are micro-porous aluminium silicate minerals

A typical Euro VI HDV after-treatment system consists of a diesel oxidation catalyst (DOC), a DPF, a SCR and, possibly an NH₃ oxidation catalyst (AOC)⁴. The order in which these components are placed varies. An advantage of placing the DOC and DPF upstream of the SCR is that the DOC oxidises some of the NO to NO₂ which can make the SCR more effective (although this does depend on the catalyst). An AOC can be used after the SCR to reduce the NH₃ emissions, and allow a high dosage of DEF to be used without increasing NH₃ emissions. The advantage of placing the SCR upstream of the DPF is that it will be closer to the engine where the exhaust temperature will be higher, with increased NO_x conversion efficiency, but poorer DPF regeneration. As both the DPF and the SCR require NO₂ there can be competition between these two requirements.

These problems can be potentially overcome by integrating the SCR catalyst into the DPF as one multifunctioning unit. In some after-treatment systems, such as Johnson Matthey's SCRT[®] system the SCR coating is applied directly to the DPF, which enables the SCR to get hotter quicker and increases the SCR volume, both of which result in more efficient NO_x conversion (Görsmann, 2015). There are other advantages including allowing earlier urea injection with improved cold start NO_x reduction, less space needed for after-treatment and less weight. However the substrate must be sufficiently porous to keep the engine backpressure at acceptable levels without incurring an additional fuel penalty and the SCR catalyst loading must be higher than on a separate SCR system. As both components use NO₂ the f-NO₂ needs to be higher than for a separate SCR system, up to approximately 75%. In addition, to achieve high NO_x conversion a second SCR may be required depending on the catalyst formulation in HDV applications (Johansen et al., 2014).

SCR was first introduced to meet the Euro IV HDV emission standards. Engine manufacturers took advantage of the trade-off between NO_x, PM and fuel consumption, to calibrate the engine to have higher engine-out NO_x but lower PM emissions and improved fuel consumption. The SCR was used to control the NO_x emissions, giving overall lower exhaust emissions than Euro III engines, at least under the controlled conditions of the European type approval test.

Engines fitted with SCR systems and tested in the laboratory, using standard EU type approval test procedures, show good NO_x reduction efficiencies. However, this has not been reflected in real driving conditions, especially in urban areas. This is thought to be due to the low exhaust temperature under many urban driving conditions.

The catalyst formulation and its loading as well as the design of the catalyst substrate can all affect SCR performance. For example, the use of high porosity substrates with increased cell density can improve SCR conversion efficiency and potentially allow up to 50% reduction in volume (Görsmann, 2015). Other developments include the use of solid urea or magnesium

⁴ Also known as an ammonia slip catalyst (ASC)

chloride (MgCl_2) as the storage medium. Solid urea needs to be heated in the presence of water vapour to decompose to NH_3 . MgCl_2 stores NH_3 and cartridges can be readily handled, and recharged. The advantage is that both of these storage devices occupy less space and are lighter than a urea tank (Johnson, 2008).

Hydrocarbons can also be used as the reductant in SCR (HC-SCR⁵). Passive systems use hydrocarbons present in the exhaust as the reducing agent whilst active systems use additional fuel added upstream of the catalyst. However, HC-SCR is less efficient at removing NOx than NH_3 -SCR and the active system can give rise to undesirable increases in emissions of carbon monoxide, hydrocarbons, PM and CO_2 . There may also be an increase in the emissions of N_2O .

2.4. Lean NOx trap (LNT)

Lean NOx traps (LNTs) store NOx under lean engine operations and release it under richer conditions. The lean phase may last 30-90 seconds and then there is a brief return to stoichiometric or rich operation, for perhaps 3-5 seconds to remove the stored NOx for reduction to N_2 using a conventional three-way catalyst (TWC) mounted downstream. Storage times are increasing with improved catalyst formulations. This approach is also suitable for use with gasoline direct injection engines which operate under both rich and lean conditions.

According to Johnson (2015) this is the preferred technology for small diesel vehicles meeting Euro 6 emissions. It is less efficient at reducing NOx than SCR, and has a nominal removal efficiency of 70-80% removal. The main advantage of LNT for light duty vehicles is that a DEF storage tank, which takes up significant space on a small vehicle, is not required (Johnson, 2008).

A typical system comprises a noble metal catalyst, for example platinum, to oxidise NOx and an alkaline and/or alkaline earth component such as barium and/or potassium to trap the NOx as nitrites/nitrates. In simple terms the NOx is oxidised and stored as a nitrite or nitrate on the trap. The engine is switched to rich conditions; the NOx is released and then reduced to form N_2 and O_2 by the TWC. However NH_3 and other reduced compounds are also produced during the process and, if uncontrolled, emitted from the vehicle exhaust.

Over 90% NOx removal has been demonstrated with new LNTs. The main problem has been their sensitivity to fuel and lubricant S which rapidly reduces the NOx conversion efficiency as the LNTs need to be regenerated periodically at high temperatures. The introduction of zero S (<10 ppm) diesel in Europe, improved catalyst formulations which are less S sensitive, and new control strategies that have reduced the desulphurisation temperature are making LNT more effective.

⁵ Also known as lean NOx absorbers or lean De-NOx catalysts

For GDI engines it has been suggested that as NO_x emissions are generally considered to be low at low loads, control strategies could concentrate on reducing emissions when both emissions and exhaust temperature are higher. This could reduce the amount of expensive platinum group metals required for the catalyst making LNTs more economical for use on large GDI engines (Johnson, 2008).

Combined SCR and LNT systems are being developed which combine the advantages of both technologies. In this case a SCR catalyst stores the NH₃ generated during the LNT rich operation, this NH₃ then reacts with the NO_x. This approach increases system efficiency enabling a decrease in the catalyst loading, and hence cost, for the same conversion efficiency (Johnson, 2008).

There have also been improvements in the LNT materials used. Second generation LNTs are able to hold more than twice the NO_x at 130 °C compared to the first generation, and release it at higher temperatures when the downstream SCR is operational (Johnson, 2015).

An example of the combined use of LNT and SCR is the diesel Cold Start Concept (dCSC™) developed by Johnson Matthey. This component traps NO_x from cold start and releases it when the downstream SCR is sufficiently hot to convert the NO_x (Görsmann, 2015).

3. REAL WORLD EMISSIONS

3.1. Introduction

This section assesses the performance of NO_x abatement technologies used on light and heavy duty diesel vehicles under real world driving conditions. It draws largely on work undertaken in London (Carslaw & Rhys-Tyler, 2013) and Leeds (Tate, 2014) using vehicle emissions remote sensing and data from portable emission measurement systems (PEMS). PEMS measure instantaneous emissions as a vehicle is driven and cannot be directly compared to the emissions measured under laboratory conditions.

Data from the most recent Euro standards, nominally Euro 4 and IV (from 2005) to Euro 6 and VI (from 2014 and 2013 respectively) has been used to understand the in-service performance of NO_x abatement devices. These vehicles represent the majority of vehicles on the road today. NO_x, NO₂ and NH₃ emissions are discussed separately.

3.2. Nitrogen oxides (NO_x)

3.2.1. Cars

The remote sensing data in Figure 1 (Tate, 2014) and from Carslaw & Rhys-Tyler (2013) both show that there has been no real reduction in NO_x emissions from diesel cars up to Euro 5. Euro 4 and 5 NO_x emissions were slightly lower than Euro 3, but similar to pre Euro 1. These vehicles use relatively unsophisticated EGR to control NO_x emissions.

The Leeds remote sensing data provides some evidence that NO_x emissions from Euro 6 diesel cars are lower than from Euro 4 and 5 diesel cars (see Figure 1) ⁶.

According to Ligterink et al. (2013) NO_x emissions from Euro 6 diesel cars have a very large variation in NO_x emissions. Depending on the test conditions and vehicle technology the spread is 10% to 1000% of the limit value, when measured under laboratory conditions that simulate real world driving. This wide variation is mainly due to the type, functionality and control strategy of the after-treatment device. The authors concluded that during type approval the control strategies are active and effective, but during real world operation they are not operational all the time.

A meta-analysis of PEMS emissions data of Euro 6 diesel cars (and some equivalent US certified cars) undertaken by the International Council on Clean Transportation (2014b) found that the average on-road emission of NO_x were seven times the emission limit. A sizable share of the emissions occurred during discrete events lasting a few seconds. These were not, in general, due to “untypical” driving but were due to normal transient increases in engine load or during regeneration of the DPF. This suggests that while the Euro 6 emissions may be lower than Euro 5, they are still far in excess of the type approval limit. These high emissions occurred for cars using all three abatement technologies (i.e. EGR, SCR and LNT).

The International Council on Clean Transportation (2014b) suggests that it is difficult to inject the optimum amount of urea in a SCR system, particularly during rapid changes in engine load. For LNT systems there is a fixed NO_x capacity and under high-load situations NO_x can break through. Another possible explanation for the high NO_x emissions for Euro 6 cars is the pressure on manufacturers to reduce CO₂ emissions. Robust control of NO_x emissions is likely to result in a fuel penalty that would affect compliance with the CO₂ standards, thereby creating an incentive for manufacturers to optimize fuel consumption to the detriment of real world NO_x performance. The authors concluded that the vehicle NO_x control strategies are optimised for the current type-approval test procedures, but are not sufficiently robust to give acceptable on-road performance. This approach is currently legal but is unlikely to be when PEMS testing is introduced for EU type-approval of light duty vehicles in the future.

⁶ The remote sensing data cannot be compared to the limit value.

The principle of introducing a ‘real driving emissions’ (RDE) test using PEMS has been agreed for passenger cars; although the details are yet to be finalised. PEMS tests are required from 2015 for reporting and monitoring purposes, with RDE testing scheduled to become an integral part of the type approval process when the Euro 6c standard comes into effect in 2017 for new types and 2018 for all new cars. Negotiations have taken longer than anticipated and there is concern that there could be a delay in implementation.

Of particular importance is the conformity factor. This is the ratio of in-service emissions measured using PEMS to the test cycle emissions limits. This will be determined on the basis of the initial monitoring data. The European Commission has estimated that a conformity factor of 1 would result in compliance with the ambient NO₂ limit value virtually everywhere by 2020, but with a conformity factor of 4 non-compliance in 2020 would be 3 times higher (European Commission, 2013).

PEMS data is not directly comparable with laboratory emissions data as measured on a chassis dynamometer. The Type 1 test in the EU type approval procedure is designed to be as reproducible as possible, and standardises a range of factors that influence emissions under real world driving conditions. These include road gradient, traffic conditions, ambient temperature and driver behaviour. One to one comparison of the laboratory and RDE test results is not possible. The PEMS data will be analysed statistically over periods of similar duration as the Type 1 test.

The proposed RDE procedures include PEMS testing over a 90 to 120 minute long trip, which would include urban, rural and motorway driving on working days. The test would include cold start but data would be excluded from the evaluation until the engine coolant temperature reaches 70 °C or for a maximum of five minutes. However, these details remain subject to final agreement (May, 2015).

3.2.2. *Light duty commercial vehicles (vans)*

There is less information available on the real world emissions of vans. The remote sensing data from London (Carslaw & Rhys-Tyler, 2013), however, suggests that these vehicles behave in a similar way as diesel cars.

3.2.3. *Heavy good vehicles (HGVs)*

Remote sensing data from London (Carslaw & Rhys-Tyler, 2013) shows that there has been little change in real world NO_x emissions of heavy goods vehicles (HGVs) from Euro III to Euro V, although PEMS data from the Netherlands (Vermeulen et al., 2012; 2014) suggests that some Euro IV and V trucks have lower emissions than earlier models, although there is large variability between individual vehicles.

According to Vermeulen et al. (2014) early Euro IV trucks, introduced before the standard was mandated from 2005, did not, on average, result in a reduction in real-world NO_x

emissions compared to vehicles built to previous standards under low speed urban driving conditions. However, the second generation of Euro IV HDVs had, on average, lower emissions at low and intermediate vehicle speeds than the earlier models, but some individual vehicles still performed badly. Early Euro V HDVs had lower average emissions at all vehicle speeds than Euro IV, but again some vehicles performed badly, and not only at low vehicle speeds. The authors concluded that meeting Euro V emissions type approval does not guarantee low emissions. However, the second generation of Euro V vehicles showed significantly lower average NO_x emissions at low speed.

Euro VI HDVs are required to comply with a mandatory RDE test conformity factor of 1.5. Testing of a number of Euro V and VI HDVs (Vermeulen et al., 2014) showed that the in-service NO_x emissions of Euro V HDVs varies significantly, but that 75% achieved a non-mandatory conformity factor of 1.5. On the other hand all the Euro VI vehicles met the mandatory NO_x conformity factor of 1.5, with some vehicles having a factor of well below 1.

The first long distance Euro VI trucks had very low NO_x emissions compared to Euro V vehicles, including at low speeds. The authors conclude that the Euro VI legislation has led to significantly lower real-world NO_x emissions, although not all Euro VI vehicles have low emissions under all driving conditions.

Chassis dynamometer data from the VTT (the Technical Research Centre of Finland) (International Council on Clean Transportation, 2015) of 38 Euro IV to VI HDVs also show that the new Euro VI certification protocols are, in general, effective at reducing NO_x emissions under a wide range of test cycles, including one that simulates urban driving. The changes to the type approval engine tests include cold start and reductions in the average engine load and power during the test cycles. The authors concluded that the good Euro VI NO_x abatement is a result of more efficient catalyst formulations, better thermal management of the catalyst, improved urea dosing strategies and other after-treatment optimisations. The average urea consumption for vehicles tested increased approximately 25% from Euro V to VI, showing that the SCR systems were more often operational. In addition, the Euro VI HDVs showed a much smaller range in NO_x emissions.

Vermeulen et al. (2014), however, have shown using PEMS that there remain issues with some Euro VI distribution trucks and buses. These vehicles have lower emissions than previous generations, but in busy urban driving conditions, with low driving speeds, some vehicles continue to have high NO_x emissions. Under cold start urban driving conditions emissions can be an order of magnitude higher than when the engine is warm. The vehicles tested were all new and little is known of the deterioration in the NO_x abatement over time under real driving conditions.

International Council on Clean Transportation (2015) recommended that other jurisdictions considering adopting EU emission standards should not adopt the Euro V standard, but instead go directly from Euro IV to Euro VI standards to achieve a significant NO_x reduction

and implement strong in-use compliance and enforcement programmes to ensure low in-service emissions.

3.2.4. Buses

The remote sensing data from London (Carslaw & Rhys-Tyler, 2013) shows little reduction in NO_x emissions from Euro III to Euro V buses. Euro III buses show significant differences in emissions by manufacturer, reflecting their different NO_x control strategies. Euro V buses with SCR fitted by the original engine manufacturer (OEM) have no lower emissions than buses using EGR. The lack of improvement in SCR equipped buses is consistent with the PEMS data for urban driving conditions described below.

The Dutch in-service PEMS testing programme included two Euro VI city buses and a 12 t urban distribution truck (Vermeulen et al., 2014). The NO_x emissions of these vehicles were lower than from second generation Euro V HDVs. Two of the three vehicles, however, had significantly higher emissions than the Euro VI long distance trucks. This is thought to be due to the low exhaust temperature rendering the after-treatment less effective under certain operating conditions. The NO_x emissions decreased at higher vehicle speeds and higher payloads when engine temperatures increase. The third vehicle performed well, showing that it is possible to maintain low emissions in urban areas. One of the vehicles that performed poorly under congested traffic conditions had an engine conformity factor of less than 1.5 suggesting that the RDE test requirements do not always identify vehicles with high NO_x emissions in-service.

It is not realistic to expect a vehicle certified to a certain emissions standard to stay below the limits under all driving conditions. Even during the type approval tests emissions will go above the limit as it is an average over the whole test cycle against which it is assessed. To assess the conformity factor the PEMS data is averaged over a period of similar duration as the laboratory engine test, but certain data is excluded, for example when the average engine power is less than 15% of maximum. This means that when a vehicle drives at low speeds, for example, in heavy traffic, data is excluded in the evaluation of RDE conformity. This is a real shortcoming of the RDE procedures for urban buses, and probably also for urban goods distribution vehicles.

Another shortcoming is that the engine manufacturer only has to test three engines from each ‘family’ of engines, and to test these in the type of vehicle in which the engine is most commonly used. As long distance trucks sell more than urban buses manufacturers will tend to test their engines in trucks rather than buses, which have different in-service operating patterns. NO_x emissions from a truck may be low under the RDE tests, while the emissions from the same type of engine in an urban bus could be higher due to the different driving conditions it encounters during its working life.

3.3. Nitrogen dioxide (NO₂) emissions

3.3.1. Introduction

Nitrogen oxides are emitted from combustion sources, such as road vehicles, mainly as nitric oxide (NO), which reacts with ozone (O₃) in the atmosphere to form NO₂. Some NO₂ is directly emitted, but for many years it was generally accepted that this fraction was small, possibly around 5% (Air Quality Expert Group, 2007). There is evidence from both analyses of ambient monitoring data and vehicle measurements that show that this proportion has increased in recent years.

Grice et al. (2009) reviewed the literature of vehicle f-NO₂ and made a series of recommendations of typical emissions as shown in Table 1.

Table 1: Recommended Primary NO₂ emission percentages for different vehicle types (f-NO₂) (Grice et al., 2009)

Vehicle type		f-NO ₂ (%)
Gasoline cars	Euro 2 and earlier	4
	Euro 3-6	3
Diesel cars/vans	Euro 2 and earlier	11
	Euro 3	30
	Euro 4-6	55
HGVs	Euro II and earlier	11
	Euro III	14
	Euro IV-VI	10
Buses	Euro II and earlier	11
	Euro III no DPF	14
	Euro III with DPF	35
	Euro IV-VI	10

These f-NO₂ values are similar to those of Kousoulidou et al. (2008) with the exception of HDVs where the f-NO₂ for Euro IV was assumed to be 14%; Euro V 18% and Euro VI 35%.

The f-NO₂ for diesel cars, not fitted with exhaust treatment technology, is more than twice that from gasoline cars. There has been a significant increase from Euro III onward when diesel oxidation catalysts and then DPFs, were fitted. Similar increases occurred for Euro III HDVs fitted with DPFs, although this has reduced with the use of SCR from Euro IV.

Keuken et al. (2011) showed that the use of oxidation catalysts to reduce PM emissions from diesel cars increased from 5% in 2000 to between 55% and 70% in 2010 in the Netherlands, but suggested that overall, despite the increase in NO₂ emissions, there was a health benefit of their introduction due to lower PM emissions. Primary NO₂ emissions from road traffic in the

Netherlands is expected to increase from 8 kt in 2000 to 15 kt by 2015 and subsequently to decrease to 9 kt by 2020.

The retrofitting of many buses in London with catalytically-regenerative DPFs is thought to explain some of the changes in primary NO₂ emissions. The timing of the retrofit scheme fits in with the observations at roadside and kerbside monitoring sites. By 2005 over 90% of Transport for London (TfL) buses were operating with DPFs (Air Quality Expert Group, 2007).

The amount of primary NO₂ has important implications for meeting the EU ambient NO₂ limit values. An assessment of the formation of NO₂ concentrations in areas of Helsinki with heavy traffic (Anttila et al, 2011) found that the proportion of f-NO₂ in the road transport emissions increased from below 10% in the 1990s to approximately 20% in 2009 due to the increase in diesel cars. To achieve the EU ambient NO₂ limit values, the authors concluded that the f-NO₂ emissions need to be reduced alongside the total NO_x emissions.

Grice et al. (2009) also found that in locations where high f-NO₂ from traffic is a key factor contributing to exceedances of the ambient limit values, both NO_x and NO₂ emissions need to be reduced, but elsewhere total NO_x emissions should be reduced. At sites exceeding the ambient NO₂ limit in Greece, for example, their analysis suggests that f-NO₂ is not important and the focus should be on reducing total NO_x emissions from traffic. However, at the time of the study there was an extremely low number of diesel cars in Greece which meant that a large proportion of NO_x emissions were from gasoline vehicles, which have lower f-NO₂.

The diesel proportion of new cars sold in different EU countries varies from over 70% (Luxembourg, Ireland and Portugal) to under 40% (Denmark and Netherlands) (ACEA, 2014). The issue of f-NO₂ will become increasingly important in those countries with very high diesel car populations.

3.3.2. Light duty vehicles

The remote sensing data from London (Carslaw and Rhys-Tyler, 2013) shows that the f-NO₂ from diesel cars has increased from typically 10-15% for pre Euro 3 cars to 25-30% for Euro 4 and 5. For Euro 3 these levels are lower than the estimates of Grice et al. (2009). For Euro 4 and 5 there is a wide range of f-NO₂ from approximately 12% to 55%. The authors found similar f-NO₂ for light goods vehicles.

For Euro 4 and 5 diesel cars with engine capacities less than 2 L the mean f-NO₂ was 27%; but for larger vehicles it was 43%. For Euro 5 cars the average f-NO₂ for one manufacturer was 12%; while for another it was 38%. The authors consider that this wide range is most likely to be due to the engine control strategy utilised which varies with manufacturer and vehicle model, and noted that if all manufacturers followed the control strategies of the one with the lowest f-NO₂ emissions there could be a significant reduction in NO₂ emissions.

3.3.3. *Heavy duty vehicles*

The remote sensing data from London (Carslaw & Rhys-Tyler, 2013) for HGVs show that for HDVs there has been a clear reduction in f-NO₂ from approximately 20% for Euro II and III to 5-10% for Euro IV and V. There is also evidence to suggest the smaller HGVs (3.5-12 t) tend to emit higher f-NO₂ ratio than larger HGVs (>12t). These values are also somewhat different to those of Grice et al. (2009).

The f-NO₂ emissions from buses are far more variable than from trucks, ranging from near zero to over 40%. The highest f-NO₂ emissions were observed from vehicles fitted with DPFs, and there was a large variation by manufacturer, with an average f-NO₂ emission of 15-20%, which is lower than in Grice et al. (2009).

Euro IV and V buses with SCR have a f-NO₂ range from close to zero to approximately 30%. For two bus manufacturers almost all the NO_x measured was in the form of NO, suggesting the SRC did not complete the conversion of NO₂ to N₂. Low f-NO₂ for Euro IV SCR buses has also been observed in a PEMS study by Fu et al. (2013). These buses were not fitted with a DPF. As the injection of urea under low temperature (urban) driving conditions is low, the SCR is unlikely to have been operating efficiently, and therefore the low f-NO₂ is likely to be related to the optimisation of the combustion conditions in the engine rather than being related to the SCR system.

Buses that meet the voluntary environmental enhanced vehicle (EEV) emission standards have higher f-NO₂ ratios due to the increased oxidation of the engine gases upstream of the SCR due to the use of a DPF, although the SCR does not appreciably reduce total NO_x. The EEV emission standards are between Euro III and VI depending on the pollutant and test cycle. For NO_x emissions EEVs are required to meet the Euro V steady state and transient limit values. For PM mass emissions these vehicles meet the Euro IV steady state limit, but for the transient test the requirement lies between the Euro V and VI limit values.

Vermeulen et al. (2014) used PEMS to measure f-NO₂ from Euro VI vehicles. They found that the proportion of NO₂ increases from 20% under cold start conditions to 40-80% for trips with warm engines. The range of f-NO₂ emissions is large especially for warm operating conditions, due to the different NO_x control strategies used. The authors noted that under these conditions the NO_x emissions are very low, and therefore imply low accuracy of the measured f-NO₂.

There is some evidence that the f-NO₂ decreases as the DPF ages and there is a reduction in the oxidative potential of the DPF (Carslaw et al., 2015).

3.4. **Ammonia (NH₃) emissions**

There has been concern that the use of urea in SCR will increase exhaust emissions of NH₃, an important precursor to secondary PM. However the remote sensing data from London

(Crawshaw and Rhys-Tyler, 2013) shows that emissions are low from Euro IV and V HDV. When expressed as a ratio to CO₂ emissions Euro IV SCR buses emit considerably less NH₃ than older catalyst-equipped gasoline cars.

4. RETROFITTING BUSES

Transport for London (TfL) has undertaken the largest bus retrofit programme in the world, starting with DPFs and more recently fitting SCR to its Euro III buses. By the end of 2015 nearly 2,000 buses will have been retrofitted with SCR. The remaining Euro III buses in TfL's fleet will be retired and replaced with Euro VI buses (Mayor of London and Transport for London, 2015). The Transport Authority of Barcelona (TMB) has also embarked on a 'low NO₂ SCR' retrofitting programme, with over 400 Euro II and III diesel buses retrofitted.

The results of a TfL commissioned study on the in-service efficacy of retrofitted 'low NO₂ SCR' is described in this section.

The Euro III buses have been fitted with a specially designed 'low-NO₂ SCRT'. The optimised system is designed to work more efficiently under urban driving conditions than the OEM fitted SCR (Carslaw & Rhys-Tyler, 2013) to minimise the emissions of both NO_x (see section 3.2.4) and NO₂. It combines a DPF to reduce PM and a SCR to reduce NO_x emissions. The SCR is close coupled and thermally managed so the catalyst activity is maximised. The calibration increases the urea injection when operating conditions allow. The SCR catalyst is three times the volume of those used in a standard SCR.

Carslaw et al. (2015) tested emissions from London buses retrofitted with this device using remote sensing under normal driving conditions and on a test track. The on-road tests show that the 'low-NO₂ SCRT' on average reduces NO_x by 45% and NO₂ by 61% compared with similar buses fitted only with a DPF. There are times, however, when even the 'low NO_x SCRT' system fails to reduce NO_x, at other times the NO_x reduction is greater than 90%. It seems that the system cannot reduce NO_x when the engine is cold or following a period of driving through congested traffic, when the exhaust temperature is low.

Under test track conditions NO_x reductions of 77% were observed with the 'low NO₂ SCRT' system compared to the 'base' bus fitted only with a DPF. However, the bus fitted with a SCR but no DPF reduced NO_x emissions by 90% compared to the 'base bus'. The higher efficiency of the SCR compared to the 'low NO₂ SCRT' was related to the higher inlet temperature resulting from the engine exhaust not having to pass through a DPF first. When inlet temperatures were greater than 200 °C the NO_x removal from the 'low NO₂ SCRT' was approximately 90%.

The test track results show that it took approximately 20 minutes for the 'low NO₂ SCRT' to warm up at a mean speed of approximately 30 km h⁻¹.

Although the tests showed that the absolute emissions of NO_x were lower with the ‘low NO₂ SCRT’, the absolute emissions of NO₂ were higher compared to the ‘base bus’. The original Euro III engine would have had no after-treatment such as a DPF and therefore would have had lower absolute emissions of NO₂ compared with the retrofitted bus.

The reduction in primary NO₂ emissions compared to buses fitted with DPFs are important as buses can make a significant contribution to roadside NO₂ concentrations. According to Carslaw et al., (2015) urban buses account for one third of the road transport primary NO₂ emissions in central London, and on individual road links this can be even higher. These road links are where there are exceedances of the EU ambient NO₂ limit values. Retrofitting urban buses with a ‘low-NO₂ SCRT’ system could therefore make an important contribution to the reduction in ambient NO₂ concentrations and achievement of the EU limit value. Buses tend to have a long service life and therefore a slow fleet turnover, making them particularly suitable for a retrofit programme.

5. RECOMMENDATIONS

5.1. Light duty vehicles

A new type approval test cycle for light duty vehicles is to be introduced for Euro 6c from 2017 for new types and 2018 for all new cars. This test cycle, the World Harmonized Light duty test Procedure (WHLP), has been developed from measurements of real world driving conditions, and is considered to be more representative of European driving conditions than the existing test cycle. It will include steeper transient accelerations as well as higher speeds and loads. Other changes to the test procedure include tightening the specifications for warming the engine prior to testing and lowering the test temperature. It is anticipated that adoption of the WHLP will require advanced control for cold start and will make NO_x abatement more challenging (Forzatti et al., 2015).

Real driving emissions (RDE) tests are also to be introduced for passenger cars with the introduction of the Euro 6c standards. This measure is expected to ensure that real world emissions of NO_x from diesel cars and vans are low.

Progress has been made with the development of the RDE requirements, but agreement between the motor manufacturers and the legislators on the compliance factor (the ratio of the in-service emissions to the laboratory Type 1 test emissions) is urgently needed and at the lowest reasonable level.

The RDE data should be made publically available to inform consumer choice.

5.2. Heavy duty vehicles

For HDVs the Euro VI emission standard introduced ‘not to exceed’ limits to control off-cycle emissions; that is emissions that occur under driving conditions not included in the test cycles, and RDE tests. Other changes include an emission limit for NH₃. An emission limit for NO₂ may be defined in implementing regulations. There is evidence that these measures together have resulted in Euro VI long distance trucks having low NO_x emissions.

To control in-service emissions from all HDVs the legislation needs to ensure that the RDE tests include urban buses and distribution trucks. The different operating cycles of these vehicles need to be explicitly recognised, so that the issue of poor NO_x control during and following periods of idling in congested traffic is addressed.

More information is needed on urban bus and distribution vehicle emissions during operation. For example understanding how quickly NO_x emissions increase under idling conditions. This is important because these vehicles operate where public exposure to NO₂ is the greatest.

5.3. NO₂ emissions

There is evidence that SCR can reduce NO₂ as well as NO_x emissions significantly, but this needs to be kept under review as the technology develops, and appropriate emission limits legislated if necessary.

5.4. Emission factors

New emission factors need to be developed for use in inventories that explicitly consider the in-service performance of abatement technology used in Euro 6 and VI vehicles, and its effectiveness under different during conditions.

The introduction of NO_x after-treatment is relatively recent and there is insufficient knowledge of in-service deterioration in the efficiency of these devices. Measurements need to be undertaken on older vehicles to ensure that robust emission data is available for use in inventories.

5.5. Retrofitting

Retrofitting pre Euro VI urban buses and distribution trucks with ‘low NO₂ SCRT’ devices may offer a relatively cost-effective way of achieving the ambient NO₂ limit value, whilst also reducing PM emissions, where exceedences are close to busy roads with a high percentage of these vehicles.

6. SUMMARY AND CONCLUSIONS

There is little robust evidence that real world NO_x emissions from diesel vehicles declined from when the Euro emission standards were first introduced in the early 1990s until the Euro 6 (light duty) and VI (heavy duty) standards were introduced. In addition, PM abatement has increased the proportion of NO₂ in the NO_x emitted from vehicles. Both these factors, together with the increasing proportion of diesel cars in the vehicle fleet, have contributed to the difficulty in achieving the EU ambient NO₂ limit values in many cities across Europe.

There is some in-service evidence that NO_x emissions for Euro 6 cars are typically lower than from earlier generations, but they remain on average many times the type approval emission limit. Portable emissions measurement system (PEMS) testing shows that there is a very large variation in emissions depending on the test conditions and vehicle. There is concern that Euro 6 cars using exhaust gas recirculation (EGR) on its own will have high NO_x emissions during motorway driving. The introduction of a new test cycle and real driving emissions (RDE) tests with the Euro 6c legislation are likely to address this issue.

The same picture can be seen with NO_x emissions from early generations of HDVs fitted with selective catalytic reduction (SCR). These devices were not very successful at reducing real world NO_x emissions especially, but not exclusively, at low speeds. Although some Euro IV and V HDVs have been shown to have low real world NO_x emissions under all driving conditions, good abatement of the emissions from these vehicles is not universal.

Controlling real world emissions of NO_x from diesel vehicles is not easy. Huge progress has been made with the efficiency of NO_x abatement using SCR and LNT over the last decade or so, but further development is needed to reduce emissions when the exhaust temperature is low during cold start and in congested driving conditions.

Future developments in vehicle emission legislation are likely to focus on closing the gap between the NO_x emissions measured during the laboratory type approval tests and real driving conditions for both heavy and light duty vehicles.

The introduction of the ‘not to exceed’ emission standards, designed to capture emissions under conditions excluded from the test cycles, and RDE tests has resulted in Euro VI NO_x emissions being low, at least for long distance trucks. There remain problems, however, with emission from urban buses and distribution vehicles. The SCR does not operate when the engine is cold, and it can take many minutes to warm up, and following periods in congestion the engine-out temperature can also fall to below the SCR operating temperature.

Combined NO_x abatement systems that exploit the advantages of different abatement systems are likely to be used increasingly in the future. Sophisticated EGR combined with SCR enables lower emissions under low loads. Alternatively, low NO_x traps (LNTs) combined with SCR allow the NO_x to be trapped under cold conditions and released when the SCR is

warm. Systems that combine SCR with diesel particle filters (DPFs) are also commercially available.

For light duty vehicles the new more stringent driving cycle and test procedures and RDE tests to be introduced with the Euro 6C standard from 2017/2018 are likely to reduce in-service NO_x emissions. It is important that the introduction of these measures is not delayed, and that the agreed conformity factor is as low as reasonable. As these standards will only apply to new vehicles it will be a number of years before the benefits are observed at ambient monitoring stations.

There have been important changes to the heavy duty engine emissions legislation over recent years, with the introduction on 'not to exceed' emission limits and RDE tests. These seem to be effective at reducing NO_x emissions from long distance trucks. The RDE requirements, however, need to be modified to ensure that NO_x emissions from urban buses and distribution vehicles are low under all driving conditions.

Retrofitting urban buses and distribution vehicles offers a cost effective method to significantly reduce NO_x and NO₂ emissions and could have an important role to play in achieving the ambient EU limit values in locations where these vehicles are an important source.

New emission factors for use in inventories are needed. These need to take account of the impact of different technologies under different driving conditions, and it may no longer be sufficient to have emission factors based solely on the Euro standard the vehicle or engine was built to meet.

Well controlled NO_x and NO₂ emissions from all diesel vehicles are essential to enable the ambient NO₂ limit values to be achieved as soon as possible.

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