The Evolution and Control of NO_x Emissions from Road Transport in Europe

P.G. Boulter, J. Borken-Kleefeld, and L. Ntziachristos

Abstract Road transport is the largest contributor to NO_x emissions in the EU. This chapter discusses NO_x formation mechanisms, control strategies, trends in emissions and possible future developments. Control strategies include vehicle emission legislation, engine design, exhaust after-treatment, modification of fuel properties, alternative fuels and new powertrain technologies. Calculations show that NO_x emissions from the sector decreased substantially between 1990 and 2010. Such calculations are based on the assumption that the systematic tightening of emission limits has been effective. However, there is evidence that modern diesel vehicles are not delivering the expected reductions in emissions during real-world driving. Moreover, diesel vehicles emit more NO_x than petrol vehicles (with a larger proportion of "primary" NO_2), and their market share has increased in many countries. These factors partly explain the observation that ambient NO_2 concentrations continue to exceed health-based limits in urban areas. Up to 2020 there is a need for a more effective regulation of emissions, and the chapter proposes several measures that can be taken. Beyond 2020 emissions of NO_x from the sector will depend on the market penetration of low-carbon technologies.

Keywords Air quality, Alternative fuels, Emission-control technologies, Transport, Vehicle emissions

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1 Introduction

For many years the presence of nitric oxide (NO) and nitrogen dioxide (NO_2) in the atmosphere has been a cause for concern on account of both the scale of anthropogenic emissions of these compounds and their impacts on health and the environment. There are various atmospheric reactions which cycle NO and NO_2, and it is therefore convenient to think of the two compounds as a group. By convention the sum total of oxides of nitrogen (i.e. NO + NO_2) is termed NO_x and is expressed as NO_2 mass equivalents.

The compound of more interest in relation to local air quality and human health is NO_2. It is an irritant and oxidant gas which has been linked to a range of adverse health effects, including cancer, although it is possible that NO_2 may be acting as an indicator of other traffic-related carcinogens. The most consistent association has been found with respiratory outcomes [1, 2]. NO_2 is also a precursor for a number of harmful secondary air pollutants, such as nitric acid, the nitrate part of secondary inorganic aerosols and photo-oxidants including ozone. In addition, NO_2 absorbs visible solar radiation, thus contributing to impaired atmospheric visibility.

NO_x emissions are implicated in phenomena such as acidification and eutrophication, with their subsequent impacts on the biodiversity of habitats, as well as radiative forcing of climate through the formation of nitrate aerosol and tropospheric ozone, and impacts on the carbon cycle [3].

Because of these adverse effects of NO_x compounds on both human health and the natural environment, several regulatory steps have been introduced to control NO_x emissions from different sources and to limit ambient NO_2 concentrations.

Nitrogen oxides are primarily produced when nitrogen and oxygen combine at high temperatures and pressures. Such conditions are reached in internal combustion engines (ICEs). Consequently, motorised transport modes – and in particular road vehicles – are major contributors to overall NO_x emissions. This chapter deals with

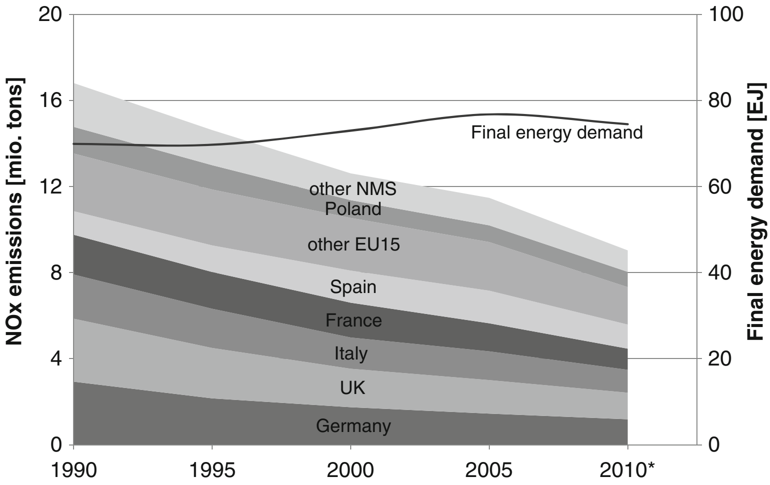

Fig. 1 NO_x emissions in Europe (EU-27) by country versus total final energy demand during the period 1990–2010. The data for 2010 are still provisional [4]

the formation and control of NO_x emissions from road transport, explains historical trends in emissions from the sector and discusses possible future developments.

2 Evolution of NO_x Emissions to date

It can be seen from Fig. 1 that NO_x emissions in the European Union (EU-27) decreased by more than 40% between 1990 and 2010, although total final energy demand grew by almost 10% during the same period [4]. Emissions were reduced in almost all countries, and notably in the larger countries (Germany, UK, Italy, France and Poland) with the exception of Spain, where total NO_x emissions increased between 1990 and 2005. The general decreases resulted from progressively stringent emission controls across all sectors, a restructuring of the power supply, and an overall increase in energy efficiency.

Figure 2 shows sectoral NO_x emissions in the EU-27 countries in 2008 based on submissions to the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP). Almost all of the oxides of nitrogen emitted to air were from combustion sources, and road transport was the single largest contributor (41%) [5]. In urban areas the emissions from road transport are proportionally higher, and the local impacts are exacerbated, due to the density of the road network, the volume of traffic, the close proximity of the population to the emission source, and the larger distances to other relevant sources.

Legislation and strategies to reduce exhaust emissions from road vehicles have been in place since the early 1970s. Calculations have established that emissions of

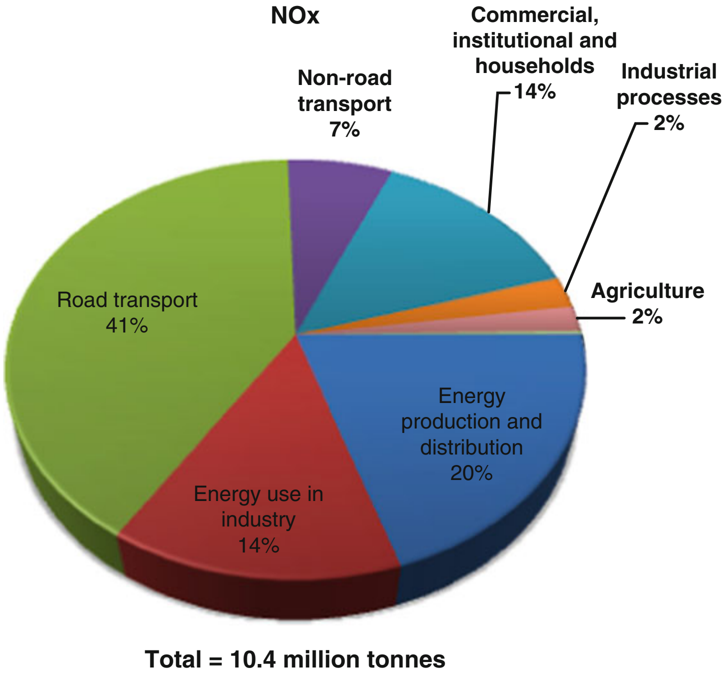

Fig. 2 Sectoral emissions in the EU-27 in 2008 [5]

most regulated pollutants (including NO_x) from road transport in the EU peaked in the early 1990s [6]. NO_x emissions from road vehicles decreased by 45% between 1990 and 2010, while the volumes of passenger transport (in passenger-km) and freight transport (in tonne-km) increased by more than 40% and 80%, respectively (see Fig. 3). This effective decoupling of transport activity from NO_x emissions was achieved through the interplay between legislation and technology. Notably, the mandatory use of the three-way catalytic converter (often referred to as a "three-way catalyst" – TWC) in 1992 reduced emissions from petrol passenger cars rapidly and substantially. With some delay, emission-reduction measures for heavy-duty vehicles also became effective. These two vehicle categories together contribute around 80–90% of all road transport NO_x emissions. However, the reduction in NO_x emissions in Europe has slowed down in recent years due to increasing numbers of diesel cars in the fleet. The emission characteristics of diesel vehicles pose a challenge to any emission-reduction target, as will be explained below.

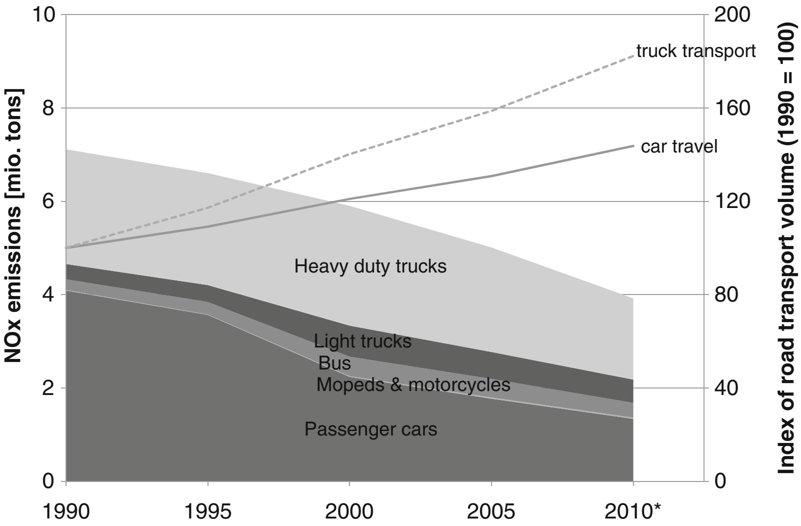

Fig. 3 NO_x emissions by road vehicles in Europe (EU-27) versus transport demand during the period 1990–2010. The 2010 data are still provisional [4, 7]. The calculations have been made at 5-year intervals. The changes in 1995 and 2000 are mainly due to the competing effects of decreasing emission limits and increasing dieselisation

3 NO_x Emissions from Road Vehicles

3.1 Formation Mechanisms

Almost all road vehicles are powered by ICEs. The only notable exceptions include vehicles powered by electricity drawn from an external grid (e.g. trolley buses), from on-board batteries (e.g. electric cars) or, possibly in the future, from fuel cells.

In an ICE energy is derived from the burning of fuel in air, with the main oxidation products being CO_2 and water vapour. However, some of the nitrogen in the combustion air is also oxidised, leading mainly to the formation of NO. NO formation is favoured by high temperatures and pressures found in the combustion chamber, as well as lean (i.e. oxygen rich) fuelling conditions.

NO formation proceeds via two main mechanisms known as "thermal" (or Zel'dovich) and "prompt" [8], with the former being responsible for more than 90% of emissions [9]. "Fuel NO" may also be formed from nitrogen chemically bound in fuels. However, for road vehicles this is responsible for only a small proportion of total NO due to the negligible nitrogen content of fuels.

The thermal mechanism is shown in reactions (1), (2) and (3) [10]:

$$N_2 + O \rightarrow NO + N \tag{1}$$

$$N + O_2 \rightarrow NO + O \tag{2}$$

$$N + OH \rightarrow NO + H \tag{3}$$

Reaction (1) is the rate-determining step, influencing the amount of NO which is formed, and is highly dependent on combustion temperature due to the high activation energy of the reaction (320 kJ/mol). Increasing the temperature from 1,200°C to 2,000°C increases the rate of this reaction by a factor of 10,000 [11].

The prompt-NO mechanism forms NO earlier in the flame than the thermal mechanism and is initiated by reaction (4) [12]. Both N and HCN react rapidly with oxidant to form NO in the flame:

$$CH + N_2 \rightarrow HCN + N \tag{4}$$

Whilst NO is the dominant NO_x species formed during engine combustion, significant amounts of NO_2 can also be produced under certain conditions. The NO_2 which is emitted directly from vehicle exhaust is commonly referred to as "primary NO_2". As will be seen later, the amount of NO_2 emitted from the tailpipe is dependent upon the type of exhaust after-treatment used.

3.2 Emission-Control Technologies

Various engine and after-treatment technologies have been developed for controlling emissions. These technologies, which are summarised below, are often used in combination to ensure compliance with increasingly stringent legislation. For this reason emission-control systems have become increasingly complex and expensive. Whilst after-treatment technologies are generally fitted during manufacture, retrofitting to older vehicles is also a common pollution-reduction strategy [e.g. in low-emission zones (LEZs)].

3.2.1 Control of Combustion

The quantity of NO formed in a petrol engine or diesel engine depends on the combustion parameters and, to a certain extent, can be controlled by adjustment of the engine operation. For example, NO formation may be limited by retarding the spark timing in petrol engines and the fuel injection in diesel engines [8].

For diesel vehicles reductions in "engine-out" NO_x emissions are harder to achieve, mainly due to the high combustion temperatures and oxygen-rich operational regime of the engine. Moreover, the effects of engine adjustments are limited due to a trade-off between NOx and particulate matter (PM); combustion-related measures which aim to reduce emissions of one pollutant (e.g. NO_x) lead to an increase in emissions of the other (PM), and vice versa. Engine calibration is usually determined by the need to balance emissions of the two pollutants and to maximise fuel efficiency. Alternatively, emissions of either NO_x or PM can be reduced via engine calibration, with the other pollutant being controlled using an after-treatment device.

A relatively recent development has been the gasoline direct injection (GDI) engine, in which fuel injection takes place in the cylinder. This allows better control of the combustion process. Early (1990s) GDI engines operated using the lean-burn principle over their complete range, in which proportionally more air is fed in the cylinder [13]. Such engines benefited from better fuel consumption but the oxygen abundance led to higher NO_x emissions. Exhaust aftertreatment was therefore needed to bring NO_x emissions within the legislative limits [14]. Modern GDI engines can switch between lean-burn (stratified) and stoichiometric combustion depending on the operational mode to achieve low emissions and high fuel efficiency.

3.2.2 Exhaust Gas Recirculation

Exhaust gas recirculation (EGR) is a NO_x-reduction technology which has been used in both petrol and diesel engines for some years (since the mid-1990s in the case of the latter). It works by redirecting a portion of the engine exhaust gas back into the combustion chamber where it is mixed with the fresh fuel-air mixture. The EGR gases act as a diluent, thereby lowering the peak flame temperature and hence the rate of NO formation. Increasing the amount of gas recirculated reduces the rate of NO formation. However, it also reduces the combustion rate, making stable combustion more difficult to achieve [15].

In petrol engines the "internal" EGR concept is often implemented. This involves adjusting the exhaust valve timing so that some of the combusted gas is trapped in the cylinder [16]. This residual gas acts as a diluent for the next combustion cycle, hence lowering the combustion temperature. Internal EGR can only reduce NO_x slightly, because no more than 5–10% of the exhaust gas can be trapped without significantly affecting combustion.

In modern diesel engines the reduction in NO_x emissions required by legislation cannot normally be achieved using internal EGR alone. In this case an "external" EGR loop is the preferred option, whereby some of the exhaust gas is fed back into the cylinder by means of a pump, again reducing the combustion temperature. This EGR configuration has the advantage of higher EGR rates (up to 40–50%) and the possibility of adjusting the quantity of exhaust gas recirculated independently of the valve timing. It also permits the introduction of a heat exchanger between the outlet and inlet pipes (the so-called "intercooler"). This decreases the temperature of the recirculated gas – and hence the combustion temperature – even further. On the other hand, external EGR increases the complexity, size and cost of the system.

3.2.3 Three-Way Catalytic Converter

The TWC has been mandatory on all new petrol cars and vans sold in Europe since the introduction of the Euro 1 emission standard in 1992 and has proved to be a very robust technology for reducing emissions of NO_x, carbon monoxide (CO) and hydrocarbons (HC) (hence the name "three-way").

The TWC is a flow-through device consisting of a ceramic or metal substrate which is coated with an active catalytic layer of precious metals, such as platinum (Pt), palladium (Pd) and rhodium (Rh). The first two oxidise CO and HC, and Rh is used to reduce NO_x to nitrogen. High conversion efficiencies for both reduction and oxidation of pollutants in a TWC can be only achieved through stoichiometric combustion (i.e. maintaining the air-to-fuel ratio at the minimum necessary for complete combustion). A shortage of air (rich fuelling conditions) would make oxidation impossible, whilst air excess (lean fuelling conditions) would inhibit the reduction mechanism. Stoichiometry is maintained by means of closed-loop control, in which the oxygen concentration in the exhaust gas is measured using a so-called “lambda” sensor. The information from the sensor is fed to the engine control unit, and the fuel injection system is adjusted to correct the air-to-fuel ratio. The closed-loop system operates on the basis of fast correction algorithms which achieve oxygen adjustment in real time. Conversion efficiencies in excess of 90% can be achieved with a properly functioning closed-loop TWC.

3.2.4 Diesel Oxidation Catalyst

Whilst NO_x emissions from petrol vehicles can be controlled by catalytic reduction, this is not very effective under the oxygen-rich conditions of diesel combustion. A diesel oxidation catalyst (DOC) is similar to a TWC in terms of structure and configuration but is only capable of oxidation. As the exhaust gases pass through the catalyst CO, unburnt HC and volatile PM are oxidised. The conversion efficiency is a function of cell size, reactive surface, catalyst load and catalyst temperature, although emissions of CO and HC are typically reduced with an efficiency of more that 95%.

DOCs offer no NO_x-reduction capability but can lead to a conversion of NO to NO_2 in the tailpipe, thereby resulting in an increase in primary NO_2 emissions. The extent of the conversion depends on the catalyst specification and the exhaust gas temperature. Typical NO_2/NO_x ratios range from ~10% for diesel vehicles without oxidation after-treatment to more than 50% for DOC- or DPF-equipped vehicles [17].

3.2.5 Selective Catalytic Reduction

Selective catalytic reduction (SCR) is currently the main technology for enabling diesel vehicles to comply with the latest NO_x emission standards. SCR systems became standard in Euro V heavy-duty vehicles (launched in Oct. 2010), and their use is gradually being extended to light-duty vehicles. The method involves the introduction of ammonia (NH_3) into the exhaust stream to chemically reduce NO_x to nitrogen. Typically, an aqueous solution of urea ($CO(NH_2)_2$) is used as the reagent, with the ammonia being generated via thermolysis. The urea is fed into the system in defined doses upstream of the SCR catalyst. An oxidation catalyst downstream of the SCR catalyst may be used to eliminate the possibility of “ammonia slip”. The following equations describe the different reactions in these systems:

$$CO(NH_2)_2 \rightarrow NH_3 + HNCO \text{ (thermolysis of urea)} \quad (5)$$

$$HNCO + H_2O \rightarrow NH_3 + CO_2 \text{ (hydrolysis)} \quad (6)$$

$$NO + NO_2 + 2NH_3 \rightarrow 2N_2 + 3H_2O \text{ (reduction)} \quad (7)$$

Typical SCR systems may achieve on-road NO_x conversion efficiencies of 60–70% [18]. However, the thermolysis of urea is an endothermic reaction that is favoured at high temperatures; SCR is inefficient at temperatures below around 200°C [19]. Hence, NO_x emissions from SCR-equipped vehicles can often increase during urban driving where traffic conditions result in low exhaust temperatures [20]. Supplementary systems, such as EGR, are required to maintain acceptable emission-control performance under such conditions.

3.2.6 Lean NO_x Trap

A lean NO_x trap (LNT) (or NO_x adsorber) is similar to a three-way catalyst. However, part of the catalyst contains some sorbent components which can store NO_x. Unlike catalysts, which involve continuous conversion, a trap stores NO and (primarily) NO_2 under lean exhaust conditions and releases and catalytically reduces them to nitrogen under rich conditions. The shift from lean to rich combustion, and vice versa, is achieved by a dedicated fuel control strategy. Typical sorbents include barium and rare earth metals (e.g. yttrium). An LNT does not require a separate reagent (urea) for NO_x reduction and hence has an advantage over SCR. However, the urea infrastructure has now developed in Europe and USA, and SCR has become the system of choice for diesel vehicles because of its easier control and better long-term performance compared with LNT. NO_x adsorbers have, however, found application in GDI engines where lower NO_x-reduction efficiencies are required, and the switch between the lean and rich modes for regeneration is easier to achieve.

3.2.7 Diesel Particulate Filter

Diesel particle filters are a very efficient means of reducing PM mass emissions from diesel vehicles, but do not directly target NO_x. However, most diesel particulate filter (DPF) systems contain catalytic materials which assist in the DPF “regeneration” (the combustion of PM accumulated on the filter to clean the DPF and prevent blockage). Such catalytic materials can have a similar impact to DOCs

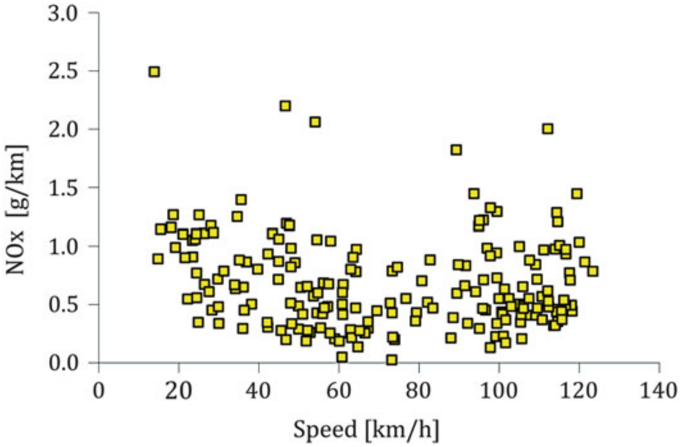

Fig. 4 NO_x emission factors for a Euro 5 diesel passenger car [23]

(i.e. the conversion of NO to NO_2 as exhaust gas flows through the filter). In some systems the NO oxidation is intentional, as NO_2 has been shown to be an effective agent for oxidising PM [21]. Again, the conversion results in higher primary NO_2 emissions and may be one of the reasons for the observed increase in the NO_2/NO_x ratio in ambient air at roadside locations in cities (e.g. [22]).

3.3 Factors Affecting On-Road NO_x Emissions

The main technological factors which influence on-road exhaust emissions are the vehicle type (e.g. passenger car, heavy goods vehicle), the fuel type (e.g. petrol, diesel) and the vehicle technology. The latter usually refers to either a specific type of engine or exhaust after-treatment or, more generally, compliance with a particular emission standard. Other considerations include the age and condition of a vehicle's engine and exhaust after-treatment system. High emission rates are often a result of component ageing, component failure, or generally poor maintenance.

Important operational factors include vehicle weight, road gradient, vehicle load and the use of auxiliary equipment such as air conditioning, the thermal state of the engine and exhaust emission-control system, and the way in which a vehicle is driven (e.g. speed, or the so-called "dynamics" of driving).

Much attention has focussed on driving behaviour, as this has a large impact on the emission level. Whilst "calm" and steady-speed driving reduces pollutant emissions, transient operation and frequent speed changes have the opposite effect. Such differences in vehicle operation account for part of the (high) variability of real-world vehicle emissions. For example, NO_x emissions from a single vehicle can vary by more than an order of magnitude, depending on the driver behaviour. Figure 4 shows a typical graph of emission factors (in g/km) for a Euro 5 diesel passenger car, plotted as a function of mean travelling speed. Road vehicle emission models in Europe, such as COPERT, HBEFA and VERSIT+, take driving dynamics into account to predict emission factors [24].

4 Reducing NO_x Emissions from Road Transport

Various technical and non-technical measures have been introduced by different authorities to reduce air pollution from road transport, and these can be grouped according to two general philosophies: "prevention" and "mitigation". Prevention measures are designed to reduce or eliminate emissions at the source, whereas mitigation measures are designed to remove pollutants which have already been emitted, to convert them to more benign compounds, or to modify their dispersion. As this chapter is concerned with emissions, the emphasis here is on prevention rather than mitigation. Mitigation measures typically involve the use of physical barriers, vegetation or some form of air treatment, and further information is available from the literature (e.g. [25–28]).

Some examples of prevention (i.e. emission-reduction) measures are described in the following paragraphs. It is worth noting that an important tool is air quality legislation, but this does not fall conveniently into the prevention or mitigation categories. Although air quality legislation acts as a driver for the development of pollution-reduction policies and technologies, and can thus be considered as "prevention", compliance with the legislation (and the need for further action) can only be determined from historical data. Moreover, air quality legislation does not specifically target road transport.

4.1 Vehicle and Engine Type Approval

The primary tool for combating air pollution from road transport is vehicle emission legislation. In the EU emission tests are required for the type approval of all new passenger car and light-duty vehicle models, and for the engines used in heavy-duty vehicles. Emission limits have been applied to vehicles and engines at the type approval stage since the early 1970s. The exhaust pollutants which are regulated are CO, HC, NO_x, PM, and, recently, particle number (PN). Whilst emissions of NO_x from vehicle exhaust are regulated at type approval, NO_2 emissions per se are not. The limits have been reduced in stages since they were first introduced (through progressive "Euro" standards), and changes have been made to the test methods to make them more realistic and effective. Emission-control technologies have developed accordingly.

For cars and light-duty vehicles the current and future test procedures and limit values have been consolidated in the Euro 5 and Euro 6 legislation [Regulation (EC) No. 692/2008]. In the exhaust emission test a production vehicle is placed on a power-absorbing chassis dynamometer. The driver must follow a driving cycle and the vehicle's emissions are collected and analysed. Emissions are measured over the New European Driving Cycle (NEDC), which is composed of low-speed "urban" segments and one high-speed "extra-urban" segment. The vehicle exhaust gases are diluted with filtered air to prevent condensation or reactions between the exhaust gas components. In addition to the regulated pollutants, carbon dioxide is measured to allow fuel consumption to be calculated using the carbon balance method. For diesel vehicles up to and including Euro 4, PM was collected separately from the other pollutants on a filter. For Euro 5 and Euro 6 vehicles, PM mass and PN are measured using a new procedure. The PN limit is designed to prevent the possibility of the PM mass limit being met using technologies that would enable a high number of ultrafine particles (<0.1 μm diameter) to be emitted. The emission limits are stated in grammes of pollutant (or number of particles) per kilometre. The NO_x and PM limits for cars are shown in Table 1, with the dates shown corresponding to new models. The EC Directives also specify a second date – usually 1 year later – which applies to first registration of existing, previously type-approved vehicle models.

The emission standards for heavy-duty vehicles apply to all vehicles with a maximum laden mass of more than 3,500 kg. The responsibility for compliance is borne by the engine manufacturer, and it is therefore the engine that is subject to type approval. The engine is operated on a test bed, with the exhaust emission limits being expressed in g/kWh (Table 2). The legislation is consolidated in the Euro V/VI standards [Regulation (EC) No. 595/2009]. In addition to introducing more stringent emission limits, the Euro V/VI regulation includes a concentration limit of 10 ppm for ammonia (NH_3), which can be emitted due to the use of additive-based control systems. A particle number limit is also planned in addition to the mass-based limit, and a maximum limit for the NO_2 component of NO_x emissions may also be defined.

The type approval of the engine rather than the complete vehicle introduces difficulties for current and future heavy-duty vehicles, as they are equipped with advanced after-treatment systems. In such cases the complete engine, after-treatment system and electronic control system have to be set-up in the laboratory for type approval to take place. To avoid this complication, type approval of the complete vehicle at the Euro VI level can be performed on the road using portable emission measurement systems (PEMS). Regulation 582/2011 specifies the technical details of the measurement.

The type approval regulations also lay down rules for in-service conformity, durability of pollution-control devices, on-board diagnostic (OBD) systems, measurement of fuel consumption, and accessibility of vehicle repair and maintenance information.

Emissions from in-use vehicles are controlled by legislation relating to periodic technical inspection (PTI) (Directives 2009/40/EC and 2010/48/EC). Here, compliance is the responsibility of the vehicle owner and, given the number of vehicles on the road, PTI tests are by necessity much simpler, shorter and cheaper than type approval tests. However, at present the PTI legislation does not cover NO_x or PM

Table 1 Type approval limits for NO_x and PM from cars

		Limit value							
		Diesel				Petrol			
Stage	Date	HC + NO_x (g/km)	NO_x (g/km)	PM (g/km)	PN (#/km)	NO_x (g/km)	HC + NO_x (g/km)	PM (g/km)	PN (#/km)
Euro 1	1992.07	0.97	–	0.14	–	–	0.97	–	–
Euro 2 IDI	1996.01	0.7	–	0.08	–	–	0.50	–	–
Euro 2 DI	1996.01	0.9	–	0.10	–	–	–	–	–
Euro 3	2000.01	0.56	0.50	0.050	–	0.15	–	–	–
Euro 4	2005.01	0.30	0.25	0.025	–	0.08	–	–	–
Euro 5	2009.09	0.23	0.18	0.005[a]	6×10^{11}[b]	0.06	–	0.005[a,c]	–
Euro 6	2014.09	0.17	0.08	0.005[a]	6×10^{11}	0.06	–	0.005[a,c]	6×10^{11}[d]

[a] 0.0045 g/km using PMP measurement procedure
[b] Added on 2011.09
[c] Direct injection only
[d] 6×10^{12} per km within first 3 years from Euro 6 effective date

Table 2 Type approval limits for NO_x, PM and smoke from heavy-duty engines

Stage	Date	Limit value[a]		
		NO_x (g/kWh)	PM (g/kWh)	Smoke (m^{-1})
Euro I	1992	8.0	0.612[b]	–
Euro II	1996.10	7.0	0.25/0.15[c]	–
Euro III	1999.10[d]	2.0	0.02	0.15
	2000.10	5.0	0.10[e]	0.8
Euro IV	2005.10	3.5	0.02	0.5
Euro V	2008.10	2.0	0.02	0.5
Euro VI	2013.01	0.4	0.01	–

[a]European Stationary Cycle. Smoke is measured over European Load Response test
[b]0.36 g/km for engines >85 kW
[c]New limit introduced in October 1998
[d]For "enhanced environmentally friendly vehicles" (EEVs) only
[e]0.13 g/km for smaller low-speed engines

mass emissions, largely because of the difficulties associated with measuring these pollutants in a simple, low-cost test.

4.2 *Emission-Control Technology*

As a result of the type approval legislation manufacturers have been required to develop increasingly effective emission-control technologies, and these were described earlier in this chapter. Table 3 provides an overview of the different devices which are typically required for light-duty diesel vehicles in each Euro category. In modern vehicles various elements are used in combination, and these have different effects on the properties and composition of the exhaust gas.

The two major steps for diesel cars were the introduction of oxidation catalysts at the Euro 2 level and the effective mandatory introduction of DPFs at the Euro 5 level. For NO_x, EGR has been the main tool to control emissions up to Euro 5. SCR systems for light-duty vehicles are starting to appear for passenger cars at the Euro 6 level.

In the case of petrol light-duty vehicles the strict control of combustion, together with improvements in TWC efficiency, has proven sufficient for compliance with the emission limits up to Euro 6.

The technology for controlling emissions from heavy-duty vehicles has, until recently, focussed on in-cylinder measures such as direct injection (DI) and high-pressure injection (HPI, >150 MPa). However, at current and future emission levels SCR has become the NO_x emission-control technology of choice, whilst DPFs will effectively become mandatory at the Euro VI level (Table 4).

In addition to engine optimisation measures, Euro V NO_x control has been achieved by implementation of two alternative configurations – either the use of EGR or the use of SCR. Ligterink et al. [20] described the different performance of

Table 3 Typical exhaust after-treatment for diesel light-duty vehicles

Emission standard		Emission control			
Stage	Date	EGR	DOC	DPF	SCR[a]
Euro 1 and earlier		–	–	–	–
Euro 2	1996	(✔)	✔	–	–
Euro 3	2000	✔	✔	(✔)	–
Euro 4	2005	✔	✔	(✔)	–
Euro 5	2009	✔	✔	✔	–
Euro 6	2014	✔	✔	✔	(✔)

Brackets correspond to application to some vehicles of the particular emission standard only
[a]SCR or NO_x trap

Table 4 Typical exhaust after-treatment for diesel heavy-duty trucks

Emission standard		Emission control					
Stage	Date	DI[a]	HPI[b]	EGR[c]	DOC[c]	DPF	SCR
Pre-Euro I		–	–	–	–	–	
Euro I	1993	✔	–	–	–	–	
Euro II	1996	✔	✔	–	–	–	
Euro III	2000	✔	✔	–	(✔)	–	
Euro IV	2005	✔	✔	(✔)	(✔)	–	(✔)
Euro V	2009	✔	✔	(✔)	✔	–	(✔)
Euro VI	2014	✔	✔	✔	✔	✔	✔

Brackets correspond to application to some vehicles of the particular emission standard only
[a]Direct injection
[b]High-pressure injection
[c]EGR and DOCs were used in buses earlier than stated in the table, and in some cases as early as the Euro I stage

the two systems in terms of NO_x control efficiency as a function of exhaust temperature. Figure 5 shows an example of the emission behaviour of vehicles equipped with the two systems. EGR performs much better at low speeds, whereas SCR becomes more efficient at high speeds as exhaust gas temperature increases. The actual performance of the two systems in real-world terms will depend on the operational patterns of vehicles, such as the frequency of use in urban or highway situations.

4.3 *Fuel Quality and Alternative Fuels*

Engine and vehicle technologies normally achieve their best emissions performance with high quality fuels. One property on which a great deal of attention has focussed is the sulphur content, partly because of the need to reduce PM and SO_2 emissions and partly because fuel sulphur has an adverse effect on certain types of engine and exhaust after-treatment technology. In Europe the controls on fuel

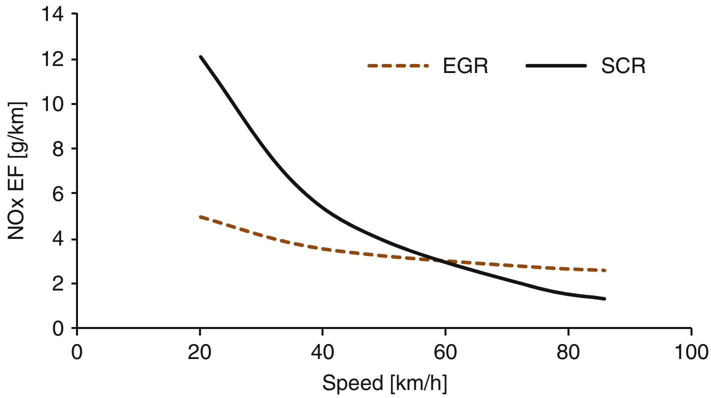

Fig. 5 NO_x emission factors for Euro V trucks equipped with SCR and EGR systems. The example refers to articulated trucks in the 34–40 t gross vehicle weight range. Source: COPERT [29], original data derived from Hausberger et al. [30]

sulphur content have been tightened in a stepwise fashion, and in the latest step Directive 2003/17/EC required a full transition to "sulphur-free" petrol and diesel (having less than 10 ppm of sulphur) by 2009. This should enable advanced technologies – such as lean-burn engines, particle traps and regenerative NO_x storage systems – to meet stringent exhaust emission limits.

In an effort to tackle climate change, biofuels such as fatty-acid methyl esters (FAME or biodiesel), paraffinic fuels from biomass-to-liquid procedures (BTL) or hydrotreated vegetable oil (HVO) and bioethanol have also been introduced into the energy mix, mostly in blends with conventional fuels. Current European regulations (Directive 2009/30/EC) permit up to 10% vol. and 7% vol. bioethanol and biodiesel mixing in fossil fuels, respectively. Higher bioethanol blends (up to 85%) require dedicated vehicle technology (so-called "flexi-fuel" vehicles) due to the need to adjust combustion parameters to the new oxygenated fuel and to use materials that can withstand the corrosive character of ethanol.

The impacts of biofuels on NO_x emissions are variable, and depend heavily on vehicle technology, operational conditions, blending ratio and biodiesel feedstock. On average, low biodiesel blends (up to 10%) do not affect NO_x emissions, or lead to a slight increase (less than 5%), but they do reduce PM emissions [31–33]. Effects at the individual vehicle level may be higher [34]. All these studies propose several possible explanations for the biodiesel impact on emissions, including the effects of higher density and viscosity, and lower compressibility and heating value, on fuel metering. Biodiesel also contains more oxygenated molecules than fossil diesel, which may also affect the combustion chemistry. It has been shown that HVO fuels can lead to reductions in both NO_x (~10%) and PM (~30%) due to their 100% paraffinic composition, and the absence of aromatics and other trace elements [35]. However, due to the unique character of these fuels (high cetane number, low density) engine recalibration may be needed to maximise their benefits [36].

The effects of bioethanol use on NO_x emissions are not consistent between different vehicles and studies. Larsen et al. [37] provided an overview of available studies, and concluded that bioethanol use can lead to either increases or decreases in NO_x emissions during tests, depending on the experimental conditions. This is consistent with the non-linear behaviour of the emission-control system in petrol vehicles, where the smallest deviations from stoichiometry greatly affect NO_x emissions. Leaner mixtures lead to an increase in NO_x emissions, and richer mixtures lead to a decrease in NO_x emissions. This erratic behaviour suggests that any direct fuel effects are masked by the ability of the fuelling system to maintain stoichiometry when changing from petrol to bioethanol blends in the different vehicles tested.

4.4 New Powertrain Technologies

Several new powertrain technologies are currently reaching niche markets in Europe. These include vehicles that use electricity for propulsion, either neat (such as battery-electric vehicles) or in combination with ICEs (such as hybrid or plug-in hybrid systems). These technologies are designed to reduce fuel consumption and greenhouse gas emissions, although they also offer significant benefits in terms of air pollutant reduction. For example, Fontaras et al. [38] showed that emissions of NO_x (and other pollutants) from two hybrid cars were distinctly lower than emissions from conventional vehicles complying with the same emission standards. The benefit comes as a result of the much more efficient use of the ICE. In the case of a battery-electric vehicle air pollutant emissions at the point of use are zero; pollutants are only emitted during electricity production, and depend on the energy mix used in each country. Assuming that power generation occurs away from urban areas, electric vehicles offer the potential for significant air quality improvements in city centres, but may also increase pollution over wider areas [39].

4.5 Eco-Driving

"Eco-driving" has been widely publicised as a means of reducing the fuel consumption and emissions of road vehicles. It is aimed at both private motorists and fleet operators, and typically involves either a simple set of rules to be followed or a programme of training. The advice or training varies considerably in terms of the level of detail, but it generally features a number of common actions, including keeping the tyres at the correct pressure, reducing the vehicle weight, avoiding sharp acceleration and heavy braking, driving in the highest gear, and avoiding unnecessary engine idling. Average overall reductions in fuel consumption of around 5–10% are typically reported for eco-driving. However, it should be noted that some adverse effects of eco-driving have been observed, such as an increase in NO_x emissions from diesel cars during urban driving [40].

4.6 *Traffic Management*

Numerous forms of traffic management offer the possibility of reducing emissions from road vehicles. Local restrictions to the access and/or circulation of traffic have been introduced in many cities. Such restrictions can take a range of forms, including tolls, congestion charges, alternate number plate schemes and weight restrictions. Cities which have implemented these types of scheme include London, Stockholm, Athens, Budapest and Prague. LEZs are one of the more effective types of restriction. Entry to a LEZ is usually conditional on a vehicle meeting specified standards. These standards can be set in various different ways, but are typically based on emission legislation, the use of specific exhaust after-treatment (e.g. DPF), or vehicle age.

The potential benefits of such measures can be illustrated by reference to a trial road charging scheme introduced in Stockholm city centre in 2006. It was estimated that the scheme resulted in a 15% reduction in total road use within the charging zone. Emissions of NO_x and PM_{10} from road traffic in the zone fell by 8.5% and 13%, respectively [41].

Various technologies and concepts involving the use of information and communication technology (ICT) and intelligent transport systems (ITS) are currently under development. Such systems include dynamic on-trip routing, "green" routing, adaptive traffic control with vehicle-to-infrastructure communication, and others. The main aims are to optimise traffic conditions and maximise mean travel speed, which are both likely to reduce fuel consumption and CO_2 emissions. However, emissions of regulated pollutants, including NO_x, may also be affected (e.g. [42]). The actual impacts of these technologies will depend on their real-world implementation and penetration.

5 Future Evolution of Emissions and Remaining Challenges

In order to protect health and the environment, vehicle exhaust emission standards will continue to be tightened in the EU, and increasing numbers of vehicles will be fitted with the latest exhaust emission-control technologies. By 2020 about one quarter of the total mileage in EU-27 is likely to be covered by cars and trucks certified to Euro 5/V, and more than half by vehicles certified to Euro 6/VI, according to scenarios examined in the LIFE + EC4MACS project (www.ec4macs.eu).

If the latest technologies are effective under real-world driving conditions, then a reduction in NO_x emissions of almost 60% over the period 2010–2020 is projected. However, there is evidence that Euro 5 diesel cars are not delivering the expected reductions in NO_x emissions during real-world driving (see discussion below). The future emission reduction therefore crucially depends on the performance and rate of introduction of Euro 6/VI technologies. So far, only a few tests on prototype Euro

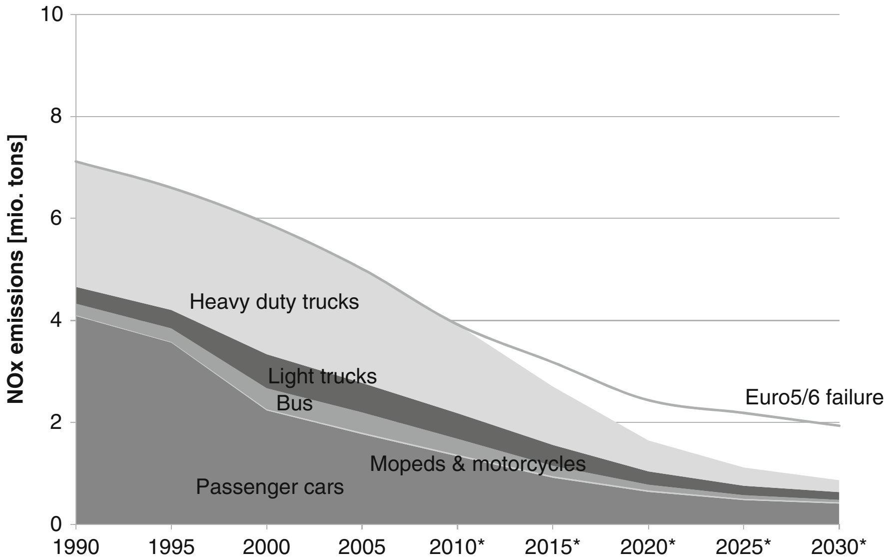

Fig. 6 Projection of NO_x emissions from road vehicles in Europe (EU-27). Together with a business-as-usual scenario, a trend scenario up to 2030 – in which Euro 5 and Euro 6 emission limits do not reduce car NO_x emissions during real-world driving below the Euro 4 level – has been included (Euro 5/6 failure line). Source: Amann et al. [44]

6 technologies equipped with SCR have been conducted [43]. The vehicles achieved high overall NO_x-reduction efficiencies, even over real-world driving cycles. However, these results were based on a non-representative sample of pre-production vehicles. If the Euro 6 emission controls for diesel passenger cars also prove to be ineffective (for example, due to poor SCR performance at low-speed conditions – as noted earlier for heavy-duty vehicles), then the actual overall reductions may not even reach 40%. Such a failure scenario is shown in Fig. 6.

The emphasis on real-world driving follows on from evidence suggesting that diesel vehicles, and in particular Euro 5 cars, have failed to deliver the expected NO_x reductions. Hausberger [45] showed that Euro 5 diesel cars may have similar emissions to Euro 3 cars. The same study showed that none of the Euro 2 to Euro 4 emission steps actually delivered the expected emission reductions. Hence, although the NO_x emission limit for a Euro 5 car is around 5 times lower than that for a Euro 1 car, Hausberger [45] showed that over real-world conditions the former may actually emit more than the latter. This unexpected behaviour is confirmed by PEMS tests performed by Kousoulidou et al. [23]. The discrepancy between type approval and real-world emissions arises from the selective optimisation of vehicle emissions over the type approval test. When vehicles operate outside the rather limited range of conditions at type approval, emissions can be largely uncontrolled. This has led to significant implications for the total NO_x emissions reported by European countries [46].

For these reasons, during the last 20 years NO_x emissions have decreased less than would have been expected given the systematic tightening of the emission limits. A direct implication of this is that NO_2 concentrations still frequently exceed health-based limits in many urban areas. According to the European Environment Agency, in 2004 more than 20% of the European urban population were exposed to ambient NO_2 concentrations above the annual mean limit value of 40 $\mu g/m^3$ [47]. Furthermore, NO_2 concentrations at many monitoring sites are not decreasing [15, 47, 48]. Although NO_2 is only a fraction of the total NO_x emitted from vehicles, analyses have indicated that a significant proportion of ambient NO_2 is actually primary exhaust from vehicles, and that the road traffic contribution to ambient NO_2 has increased in recent years [22, 49–52]. Two contributing factors have been cited:

1. Diesel vehicles emit more NO_x than petrol vehicles, and with a larger proportion of primary NO_2. In parallel, the market share of diesel vehicles has increased in many European countries. For example, the share of first registrations of diesel passenger cars in Finland increased from 17% in 2005 to 52% in 2008 [53].
2. The average proportion of primary NO_2 in diesel exhaust is increasing with changes in technology. This appears to be linked to the growth in the use of specific after-treatment technologies in modern diesel vehicles which involve in situ generation of NO_2, such as catalytically regenerative DPFs and DOCs [22, 45].

This increase in primary NO_2 emissions is compounded by atmospheric processes; background concentrations of ozone are also increasing [54], which increases the amount of atmospheric NO converted to NO_2.

It is clear that effectively addressing the NO_x emission problem involves not only more stringent emission limits over the current type approval procedure, but also a more effective regulatory policy as a whole. Measures that can be taken include:

- A revised type approval procedure that introduces a more realistic driving cycle covering a wider area of engine operation.
- The selection of random engine modes during type approval emission testing, and ensuring that emission limits are not exceeded (the "not-to-exceed" limit approach).
- The direct regulation of NO_2 emissions, independently of NO_x.
- Revised and advanced in-use compliance testing/inspection and maintenance schemes, involving OBD checks to control emissions over the lifetime of the vehicle.
- Extension of PEMS-based regulation in all vehicle categories, so as to effectively measure emissions in the field.

Several of these items are already being discussed within the UNECE working group on a Worldwide Harmonized Light vehicle Test Procedure.

Beyond the 2020–2030 horizon, reductions in NO_x will depend heavily on more effective regulations and actions, as well as the rate of introduction of low-carbon technologies such as electric and hydrogen vehicles. Vehicles operating on concepts other than ICEs have the potential for zero emissions at the point of use. However, the wide penetration of such technologies is expected to continue to be hindered by performance issues, such as limited operational range and drivability concerns. For these reasons, but also due to the relatively high cost, hybrid and electric vehicles are still considered a niche market despite the relatively wide range of models offered. Overcoming technological and infrastructural barriers remains a key future challenge for the wider penetration of such vehicles into the fleet.

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