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Recent trends and projections of primary NO_2 emissions in Europe

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A B S T R A C T

An assessment of recent trends in primary NO_2 emissions has been carried out for ten case study locations across the European Union. Estimates of the percentage of NO_x from road traffic emitted as primary NO_2 (f-NO_2) have been derived for 1995, 2000 and 2005 by combining the results of a literature survey of primary NO_2 emission factors for different vehicle types and technologies with an emission inventory. Estimates of f-NO_2 have also been derived from ambient monitoring data at roadside sites in each case study location using a model.

The results of the analysis of trends show that f-NO_2 has increased in recent years and that the rate of increase has been greatest since 2000. f-NO_2 has increased from 8.6% in 2000 to 12.4% in 2004 as an average across the monitoring sites and from an average of 6.3% in 2000 to 10.6% in 2005 as an average of the emission inventory based calculations for the case study countries. f-NO_2 is predicted to increase further to an average of 19.6% in 2010 and 32.0% in 2020 as a result of the further penetration of exhaust after treatment technologies for diesel vehicles in the fleets. This increase is expected to be offset by the large reduction in NO_x emissions over this period, resulting in an increase in NO_2 emissions from road traffic to 2015, followed by a decline to close to 2004 levels by 2020. Estimates of future ambient NO_2 concentrations have also been calculated for the roadside monitoring sites included in the study. At 29 out of 45 of these sites the annual mean NO_2 limit value is predicted to be exceeded in 2010. At 22 of these sites, the annual mean concentration is expected to remain above the limit value until 2020 and beyond.

1. Introduction

1.1. Previous work and context

Current European legislative standards controlling the pollutants released in vehicle exhaust gases, the Euro Standards (for example Directives 98/69/EC and 1999/96/EC), are intended to reduce the total emissions of NO_x from vehicle exhausts and do not differentiate between NO and NO_2 fractions. In contrast, the Framework Directive (Directive 96/62/EC) and First Daughter Directive (Directive 1999/30/EC) have been designed to control the concentrations specifically of NO_2 in ambient air to which the public is exposed. Two limit values have been specified for NO_2 in the First Daughter Directive: an annual mean value of 40 $\mu g\ m^{-3}$ and an hourly value of 200 $\mu g\ m^{-3}$ with 18 permitted exceedences each year. Both limit values enter into force on 01/01/2010.

The two different approaches to controlling oxides of nitrogen in air have resulted in a legislation gap where vehicle manufacturers have reduced NO_x emissions in compliance with the Euro standards and other directives without yielding a corresponding reduction in ambient NO_2 concentrations to below the NO_2 limit values in many locations.

One possible reason for this gap relates to the proportion of NO_x emitted directly as NO_2 from vehicle exhausts (this is the primary NO_2 fraction, f-NO_2, often expressed as a percentage). f-NO_2 in many locations may be rising as a result of changes in the composition of national vehicle fleets across Europe and the introduction of new exhaust technologies that have been brought in to meet the emission limits for various pollutants. For petrol-fuelled vehicles f-NO_2 is less than 5%, whereas f-NO_2 in diesel vehicles not fitted with new exhaust treatment technology is higher at around 10–12%. The continuing increase in the proportion of diesel-engine vehicles in national fleets will therefore have a significant impact on the ambient NO_2 concentrations, particularly in roadside environments. Furthermore, the pressure to fit diesel vehicles with after exhaust treatment technology such as particulate traps and oxidation catalysts is likely to further increase f-NO_2. Some catalyst-based particulate filters achieve the catalytic action by oxidising a portion of the NO in the exhaust to NO_2 in order to promote the oxidation of soot collected in the filter and so potentially emit a higher proportion of NO_x as NO_2.

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Early indications of a possible impact of an increase in f-NO_2 on ambient NO_2 concentrations at the roadside in London, UK, were presented by Carslaw and Beevers (2004, 2005a) and Carslaw (2005). Carslaw and Beevers (2005b) also developed an inventory for road traffic emissions of NO_2 for London and Carslaw et al. (2007) developed methods for the assessment of ambient concentrations further to predict the impact of changes in f-NO_2 on future ambient concentrations. Jenkin (2004) also noted the impact of changes in primary NO_2 emissions in London.

An analysis of monitoring data for Baden Wurtemberg in Germany has been presented by (Kessler et al., 2006) who found a significant increase in f-NO_2 over the period from 1995 to 2005. This has been linked with an increase in the number of modern diesel cars fitted with oxidation catalysts. Hueglin et al. (2006) have examined data from a roadside monitoring site situated on a motorway in Switzerland and they also found evidence of an increase in f-NO_2 between 1992 and 2004.

1.2. Aim

There are two aims of the work presented here. The first is to investigate time-series trends in f-NO_2 at a selection of case study locations across Europe using a consistent approach in all locations. This analysis includes examining how f-NO_2 has changed over the period between 1995 and 2005 and how it is likely to change up to and including 2020. The second aim is to produce predictions of ambient NO_2 concentrations at these locations for 2010, 2015 and 2020 taking the predicted changes in f-NO_2 into account.

2. Materials and methods

2.1. Tools available

Two independent methods have been used to achieve these aims:

- **An emissions inventory based approach to calculate f-NO_2.** The basis of this approach is that f-NO_2 from different road vehicles obtained from a review of the literature can be combined with a road transport NO_x emissions inventory to derive a road transport NO_2 emissions inventory. This can then be used to calculate a national urban roads average f-NO_2.
- **A model that enables f-NO_2 to be calculated from ambient monitoring data, including at locations where ozone has not been measured.** A model is required because f-NO_2 cannot be derived directly from ambient monitoring data because the ratio of ambient NO_2 to ambient NO_x concentration is determined by the overall equilibrium between ozone, NO and NO_2 concentrations, of which the primary NO_2 emission fraction, f-NO_2 is only one of the drivers. The model can also be used to calculate ambient NO_2 concentrations for a given f-NO_2 estimate.

A description of these two methods is presented below.

2.1.1. Emissions inventory approach

2.1.1.1. NO_x emissions. NO_x emissions from road transport for countries considered in this analysis have been estimated using the European TREMOVE model version 2.44 (TREMOVE, 2007). This is a policy assessment model, developed by Transport Mobility Leuven, designed to study the effects of different transport and environmental policies on emissions from the transport sector. TREMOVE models both passenger and freight transport and covers the period 1995–2020 at 5-year intervals. However, due to the emission limits for Euro 5 and 6 light duty vehicles (LDVs) not being approved by the time version 2.44 of the TREMOVE model was completed, the model in this analysis only includes LDV vehicles up to Euro 4 standard. In this study we have included emission estimates from these more advanced vehicles obtained by utilising figures calculated for the UK's Air Quality Strategy (AQS) work (Grice et al., 2006). This data provided the relative change in NO_x emission factors between Euro 4 and Euro 5/6 for light duty vehicles and gave information on the penetration of the fleet by these new vehicle types. Whilst the latter information is UK specific, it is thought that this data would be representative of the European fleets studied. This information is presented in Tables 1 and 2.

Table 1
The reduction in NO_x emissions expected with the introduction of Euro 5 and 6 relative to a Euro 4 vehicle.

	Euro 5		Euro 6	
	Entry into service	Emissions relative to Euro 4 (%)	Entry into service	Emissions relative to Euro 4
Petrol cars	Jan 2010	66	–	–
Petrol LGVs	Jan 2011	66	–	–
Diesel cars	Jan 2010	72	Jan 2015	28
Diesel LGVs	Jan 2011	72	Jan 2016	25

Note: The table shows the expected NO_x emissions relative to a Euro 4 vehicle. For example, NO_x emissions from Euro 5 diesel cars are expected to be approximately 72% of a Euro 4 diesel car.

Table 2
The fraction of the fleet that originally would have conformed to Euro 4 that is now expected to conform to Euro 5 and Euro 6.

	2010	2015	2020
Petrol LDV, Euro 4	0.83	0.35	0.10
Petrol LDV, Euro 5	0.17	0.65	0.90
Diesel LDV, Euro 4	0.87	0.30	0.09
Diesel LDV, Euro 5	0.13	0.62	0.31
Diesel LDV, Euro 6	0	0.07	0.61

Note: Of the petrol light duty vehicles (cars + light goods vehicles) that would have conformed to Euro 4 in 2015, 65% of these are now expected to conform to Euro 5 standards; 35% will remain as Euro 4. For diesel LDVs in 2015, of those that would have conformed to Euro 4 standards, 62% of them are now expected to conform to Euro 5 and 7% to Euro 6.

2.1.1.2. Vehicle specific f-NO_2 estimates. Measurements of the f-NO_2 from different road vehicles have not been undertaken within this study but relevant information has been summarised. Details of this information are given in Table 3.

From the data reviewed, it is clear that the previously accepted assumption of a 5% f-NO_2 rate is a systematic underestimate for diesel vehicles. Furthermore, the data strongly indicates that no single value of f-NO_2 is appropriate for all vehicle types. Rather, it is dependent on:

- Vehicle type (passenger car, heavy goods vehicle, bus, etc.)
- Emission standard
- Any exhaust after treatment fitted
- The average speed of the drive cycle

On the basis of these data, the f-NO_2 values used in this study are listed in Table 4. This table has been compiled with the further tenet that selective catalytic reduction (SCR) will be the dominant exhaust after-treatment system fitted to Euro IV heavy-duty vehicles and that f-NO_2 for SCR is 10% (although it is acknowledged that this is based on only a little experimental data).

The data for petrol fuelled vehicles shows f-NO_2 has remained around 3–4% for all technologies and emissions standards. For older diesel vehicles f-NO_2 is around 11%. However, this value changes with changes in vehicle technology. In particular oxidation