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Trends in primary NO_2 and exhaust PM emissions from road traffic for the period 2000–2020 and implications for air quality and health in the Netherlands

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A B S T R A C T

Application of an oxidation catalyst mainly by diesel-fuelled passenger cars reduces harmful exhaust emissions of particulate matter (PM). As a side effect, the primary NO_2/NO_x emission ratio by these vehicles increased from 10% in 2000 (before the introduction of the oxidation catalyst) to between 55% and 70% in 2010. The impact of this evolution in traffic emissions was studied from both a health and a regulatory perspective. Primary NO_2 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. Meanwhile, exhaust PM emissions from road traffic in the Netherlands will decrease from 7 kt in 2000 to 3 kt by 2020. The impact of exhaust PM on air quality and health was assessed according to the mass concentrations of elemental carbon (EC) in ambient air, as EC is a more sensitive indicator than PM. Monitoring data on the NO_2/EC concentration ratios near road traffic between 2000 and 2010 indicate no significant change in ambient air quality. This indicates that health effects in epidemiological studies associated with long-term exposure to NO_2 concentrations are still valid. The health impact from the introduction of the oxidation catalyst was assessed by comparing the relatively higher NO_2 ("cost") and lower EC ("benefit") concentrations at street locations. "Relative" refers to traffic emissions in situations "with" and "without" the oxidation catalyst being introduced. The cost–benefit ratio in 2010 was in balance, but benefits are expected to outweigh costs by 2015 and 2020. It is concluded that the application of oxidation catalysts is beneficial from a health perspective, but from a regulatory perspective it complicates compliance with the average annual limit value of NO_2. This indicates that additional local measures may be required in order to meet air quality standards at locations with high traffic intensities.

1. Introduction

Ambient NO_2 concentrations in Europe in 2009, especially those at street locations, widely exceeded the existing 40 $\mu g\ m^{-3}$ average annual limit value (ETC, 2011). Similar observations were found for each year of the period between 2005 and 2009. Especially at traffic locations, NO_2 concentrations decreased at a slower rate than NO_x concentrations (ETC, 2011; Carslaw et al., 2011). This is attributed to increasing *primary* NO_2 emissions from diesel-fuelled passenger cars (Carslaw, 2005) and the non-linear, photochemical reaction of traffic-emitted NO to NO_2 (Keuken et al., 2009). In modern diesel-fuelled passenger cars, the NO_2/NO_x emission ratio is in the range of 55–70% (Kousoulidou et al., 2008; Alvarez et al., 2008), while this was typically in the order of 10% before the introduction of oxidation catalysts (Kousoulidou et al., 2008). This increased NO_2/NO_x ratio is caused by equipment used in the after-treatment of exhaust emissions, in particular the oxidation catalyst (Alvarez et al., 2008). This trend started in 2000 with the introduction of Euro 3 standards for passenger cars and retrofitted Continuously Regenerating Particulate Traps (CRT) on urban buses. Exhaust treatment equipment has been introduced to reduce emissions of carbon monoxide, hydrocarbons and particulate matter (Alvarez et al., 2008). These emissions contribute to the formation of tropospheric ozone (e.g., Seinfeld, 1986) and have adverse health effects, especially in the case of exhaust particulate matter ("exhaust PM") (Janssen et al., 2011). Hence, from a health and regulatory perspective to comply with NO_2 limit values, the question is: What is the trade-off between reducing harmful exhaust PM emissions and increasing the NO_2/NO_x ratio in traffic emissions? The contribution of exhaust PM to ambient PM mass concentrations – even near heavy traffic locations – is limited as a result of the relatively high regional background concentrations in the

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Netherlands (Boogaard et al., 2011). The mass concentration of elemental carbon (EC) is a more sensitive indicator of the impact of exhaust PM emissions on air quality and health (Janssen et al., 2011). Therefore, EC was used in our study as an indicator for exhaust PM in ambient air.

A study in Finland (Anttila et al., 2011) concluded that, in the 1994–2009 period (and especially during the last five years), the contribution of *secondary* NO_2 (i.e., NO photochemically converted to NO_2) to total NO_2 concentrations at roadside locations decreased from 53% to 43%, while the contribution of primary NO_2 emissions increased from 32% to 44%. These traffic contributions were in addition to the regional background concentrations, which remained around 15% of total NO_2 concentrations in Finland. It is expected that NO2/NOx ratios in urban traffic emissions will continue to increase in Europe (Grice et al., 2009; EEA, 2009). In the Netherlands, for example, the percentage of vehicle kilometres driven by diesel-fuelled passenger cars increased from 25% in 2000 to 32% in 2009 (Statistics Netherlands (CBS): www.cbs.nl).

The level of NO_x emissions from road traffic may decrease substantially because of the more stringent NO_x limit values in Europe; for passenger cars due to Euro 5/6 in 2011–2014 and for light- and heavy-duty vehicles due to Euro V/VI in 2010–2015 (EC, 2007; EC, 2009). However, it is reported (ETC, 2011, pp. 29) that “In 2009, in nearly all countries of the EU Member States exceedances of the annual average limit value of NO_2 are observed at one or more stations and at 47% of the traffic stations”. Consequently, it is expected that the implementation of these Euro emission limit values may have come too late for many European urban areas to comply with the annual NO_2 limit value for 2010. This even applies to the Netherlands which has a five-year derogation period but may not meet its obligation to comply with the NO_2 limit value for 2015 (Velders and Diederen, 2009).

Acidification, eutrophication and tropospheric ozone formation are valid reasons for reducing NO_x emissions. Animal toxicological studies suggest that long-term exposure to NO_2 at concentrations above current ambient concentrations has adverse health effects (WHO, 2006). However, as pointed out by Williams and Carslaw (2011), the World Health Organization also concluded that “*it is unclear to what extent health effects observed in epidemiological studies are attributable to nitrogen dioxide itself or to other highly correlated combustion pollutants*” (WHO, 2006). In case of the latter, health effects attributed to NO_2 may not occur when the ratio between NO_2 and other combustion pollutants changes over time. In relation to the aforementioned issues, the following questions are relevant:

- What is the contribution of primary NO_2 emissions from road traffic to NO_2 concentrations in ambient air?
- What is the trend in the NO_2:EC ratio in ambient air at various locations?
- What will be the future relevance of NO_2 as a health impact indicator?

To provide answers to these questions, this paper presents the results from research in the Netherlands. Their outcomes are relevant for many European urban areas in view of the widespread dieselization of passenger cars in Europe.

2. Methodology

In the approach for our study we chose to investigate the three following issues for the Netherlands, for the period 2000–2020:

1. The trend in the ratio between NO_2 and exhaust PM emissions from road traffic;
2. The effect of this trend on NO_2/EC concentration ratios in ambient air;
3. The health impacts at locations close to intense road traffic.

We compared the projected evolution of traffic emissions for situations “with” and “without” the introduction of the oxidation catalyst. We based our study on measured data from national and regional monitoring networks, emission data from the national emissions inventory, and a street canyon model (Beelen et al., 2010) to compute the contribution of traffic emissions to concentration levels at roadside locations.

Air quality in the Netherlands is measured by the Dutch National Air Quality Monitoring Network of the RIVM at eighteen regional, eight urban and sixteen traffic locations (www.lml.rivm.nl). In this network, hourly measurements of NO_2 and NO_x are carried out by automatic chemiluminescence-based analyzers (Model 42C, Thermo Environmental Instruments Inc.). In addition, black smoke is monitored at five regional, one urban and four traffic locations, by automatic monitoring using reflectometry (ETL-SX200, ETL Systems) based on OECD’s black smoke method (ISO 9835, 1993). In the Dutch region of Rotterdam, air quality is monitored by the regional environmental protection agency (DCMR) at urban and street locations. DCMR applies multi-angle absorption photometers of the MAAP model 5012 (Thermo Scientific) for measuring black carbon. The MAAP operates by detecting both the transmittance and reflectance of light (670 nm) by particulate matter that is collected on filter tape (Petzold and Schönlinner, 2004). Black smoke and black carbon measurements may be converted to elemental carbon (EC) concentrations in ambient air (Keuken et al., 2011).

In the Netherlands, emission inventories in combination with dispersion modelling results together deliver an annual update of background concentrations at a spatial resolution of $1*1\ km^2$ (Velders and Diederen, 2009; Velders et al., 2011a). Emission factors of road traffic for NO_2, NO_x and exhaust PM, which are input for the dispersion models related to traffic emissions, are based on dynamometer testing, on-road measurements and composition of the national car fleet in the Netherlands. These emission factors are updated annually for past and future years (Velders et al., 2011a). For EC, emission factors were derived from an EU database with information on EC presented as a fraction of exhaust PM emissions (Ntziachristos and Samaras, 2009). Information from this database was combined with exhaust PM emission factors for the Netherlands to compute EC emission factors.

The outline of this paper is as follows. Section 3.1 presents the measured trend in NO_2 concentrations for 2000–2010 in the Netherlands. Section 3.2 investigates the trend in the NO_2/NO_x ratio in traffic emissions, for 2000–2020. Section 3.3 shows modelling results for the contribution of primary and secondary NO_2 to total NO_2 concentrations at a street location in Rotterdam, for 2000–2020. Section 3.4 elaborates on the trend in primary NO_2 and exhaust PM emissions from road traffic in the Netherlands, followed by details on the impact of this trend on NO_2 and EC concentrations in ambient air, as described in Section 3.5. Finally, in Section 3.6, the health impact of the relatively higher NO_2 emissions by road traffic is weighted against the relatively lower EC emissions in 2010, 2015 and 2020. Section 4 presents the conclusions and discussion of these findings.

3. Results

3.1. NO_2 trend in the Netherlands for 2000–2010

The average annual concentrations of NO_2 in the Netherlands for 2000–2010 are presented in Fig. 1.