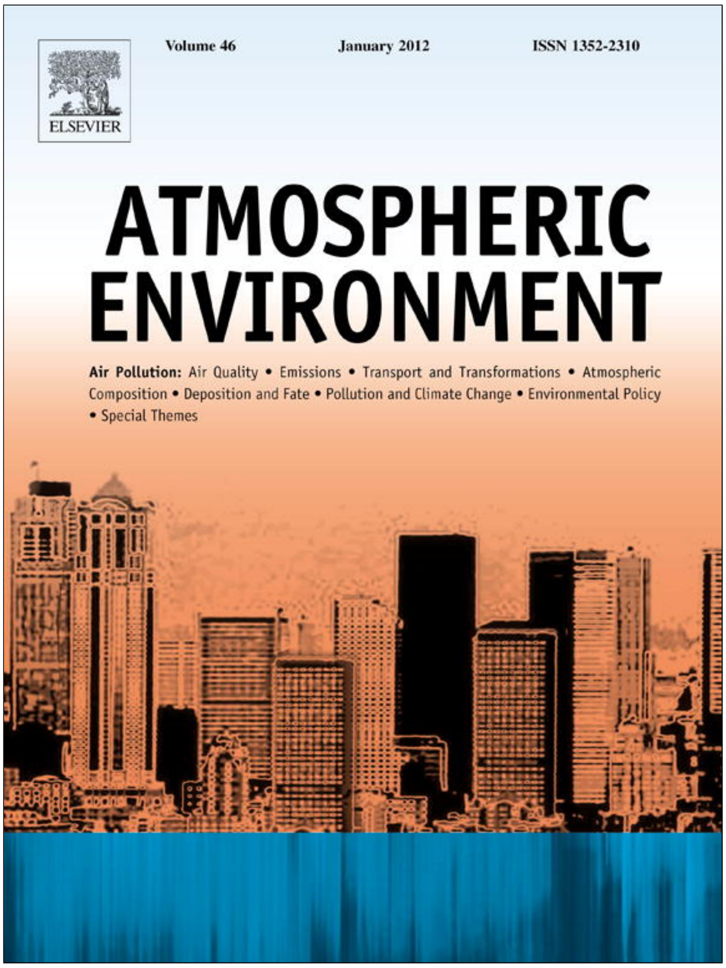

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Trends in primary NO_2 and exhaust PM emissions from road traffic for the period 2000–2020 and implications for air quality and health in the Netherlands

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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 μg 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.

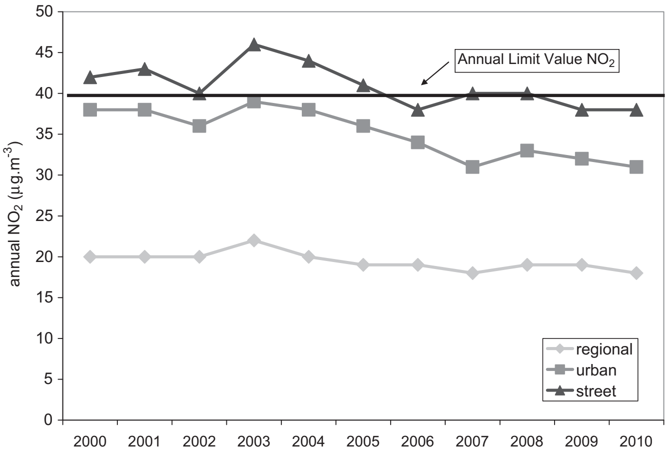

Fig. 1. Trend in annual average NO_2 concentration ($\mu g\ m^{-3}$) at regional ($n = 18$), urban ($n = 8$) and roadside ($n = 16$) locations in the Netherlands for the period 2000–2010 [Source, RIVM].

Trends in the Netherlands are similar to those in Europe (ETC, 2011). In addition, in the Netherlands, regional background concentrations of NO_2 have been more or less constant over the last decade. The decreasing trends of NO_2 at street and urban locations, as presented in Fig. 1, have regression coefficients of −0.6 and −0.8, respectively. This indicates that the decreasing trend at street locations lags behind that at urban background locations. The latter is attributed to increasing contributions of primary NO_2 emissions from road traffic. Fig. 1 illustrates that, since 2006, on *average*, NO_2 concentrations in the Netherlands have been below the annual limit value for NO_2, except near locations with intense road traffic. The total length of streets with NO_2 concentrations that exceeded the limit value in 2009 was estimated in the range of 400–1300 km (Velders et al., 2011b). This illustrates the challenge facing Dutch authorities to meet their European obligation by 2015.

3.2. Trend in primary NO_2 emissions in the Netherlands for 2000–2020

Emission factors of primary NO_2, NO_x and exhaust PM for the Dutch national car fleet are based on a combination of dynamometer driving cycles simulating urban, non-urban and motorway traffic (Velders et al., 2011a; Ntziachristos and Samaras, 2009). In the Netherlands, on-road measurements of exhaust plumes as well as roadside measurements were performed to supplement the regulatory test cycles with real-world emission measurements. Finally, the data were input for the emission model VERSIT+, to derive annual updates of road traffic emission factors for the Dutch traffic composition (Smit et al., 2007). Since 1992, exhaust emissions in Europe have been regulated by emission limit values and testing cycles in accordance to "Euro classes": pre-Euro (previous 1992), Euro 1 (1992), Euro 2 (1996), Euro 3 (2000), Euro 4 (2005), Euro 5 (2009–11) and Euro 6 (2014) (EC, 2007). In Fig. 2, NO_x emission factors are presented for diesel-fuelled passenger cars, based on EU testing cycles and on-road measurements along urban roads and motorways in the Netherlands.

Fig. 2 illustrates reductions by a factor of five in EU limit values for NO_x emissions from diesel-fuelled passenger cars from Euro 1 to Euro 5. The real-world NO_x emissions from vehicles in the Euro 1 class, first introduced in 1992, were actually somewhat higher than those from vehicles in the pre-Euro class. This was due to the shift from indirect injection to direct injection diesel engines (Kousoulidou et al., 2008). Although not shown in Fig. 2, similar trends have been observed for medium- and heavy-duty trucks (EC, 2009). Fig. 2 shows that NO_x emission factors derived for vehicles in the Euro 3 class and onwards, significantly *underestimated* real-world emissions. This is in agreement with a recent study on NO_x emission factors based on remote sensing data of NO_x/CO_2 ratios at roadside locations (Carslaw et al., 2011). It was concluded that NO_x emission factors of diesel-fuelled passenger cars did *not* show a decreasing trend, not even for vehicles from pre-Euro onwards. The study by Carslaw mainly concerned urban driving conditions at mean speeds of 31 km h^{-1} and a mean uphill slope of 0.7°. These conditions may explain the difference with our real-world results, which were based on on-road measurements of vehicles under different driving conditions. NO_x emissions based on regulatory test cycles were not only underestimated for passenger cars and light-duty vehicles, but also for medium- and heavy-duty trucks (Velders et al., 2011b). Therefore, we used in our study emission factors for road traffic in the Netherlands that were based both on dynamometer testing of regulatory and real-world cycles and on-road measurements.

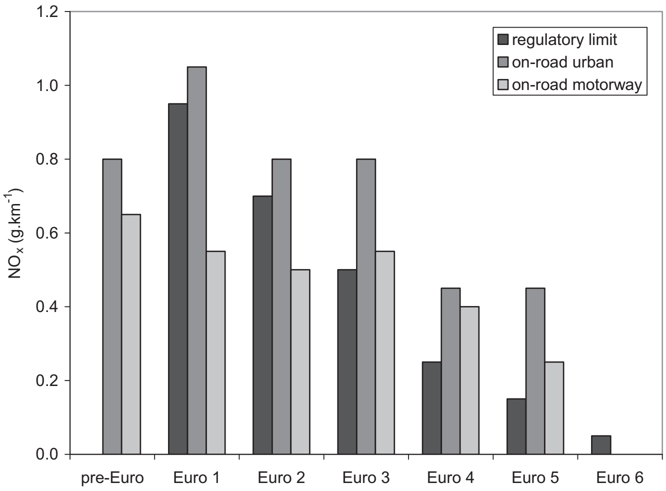

Fig. 2. NO_x emission factors ($g\ km^{-1}$) for diesel-fueled passenger cars in accordance to EU testing cycles EU1 to EU6 and based on actual driving conditions at urban roads and motorways in the Netherlands.

As part of the annual update of emission factors in the Netherlands, primary NO_2/NO_x ratios in vehicle emissions are also measured. Table 1 provides data on this ratio in percentages, for different vehicle classes (i.e., light-, medium- and heavy-duty) and road types (i.e., urban, non-urban and motorways), for the 2000–2020 period (Velders et al., 2011a). The L1 category contains diesel-, petrol- and LPG (Liquid Propane Gas)-fueled passenger cars and light-duty vehicles, L2 contains medium-duty vehicles with a weight of between 3.5 and 20 tons, and L3 contains heavy-duty

Table 1
Average NO_2/NO_x emission ratios (%) per category: L1 (diesel-, petrol-, and LPG-fuelled passenger cars and light-duty vehicles), L2 (medium-duty vehicles) and L3 (heavy-duty vehicles) for the Netherlands, for different road types: urban, non-urban and motorway, for 2000–2020. The averaging was performed over the composition of vehicles on Dutch roads and the number of kilometres driven.

	(% primary NO_2 in NO_x emissions) (urban/non-urban/motorway)		
	L1	L2	L3
2000	5/5/5	7/7/7	7/7/7
2005	17/18/20	7/7/7	7/7/7
2010	22/18/37	6/7/10	5/5/6
2015	31/36/42	4/4/6	4/4/4
2020	31/36/42	3/4/5	3/3/3

vehicles of over 20 tons. Almost all medium- and heavy-duty vehicles use diesel fuel.

It is noted that the ratio of NO_2/NO_x in emissions from diesel-fuelled passenger has increased from 10% (2000) to 55–70% by 2010 (Kousoulidou et al., 2008; Alvarez et al., 2008) but their percentage of vehicle kilometres in the L1 category is "only" 30%–45%, depending on the type of road (i.e., urban, non-urban and motorway). In gasoline-fuelled vehicles the ratio NO_2/NO_x is in the order 3–4% (Kousoulidou et al., 2008). This explains why primary NO_2 emissions in the L1 category of Table 1, for example for motorways, range from 5% in 2000 up to 42% by 2020.

The data in Table 1 also show that the share of primary NO_2 emissions from medium- and heavy-duty vehicles is expected to remain between 3% and 7%, over the 2000–2020 period (EC, 2009). The reason for this is that these types of vehicles do not have oxidation catalysts to reduce exhaust PM emissions but mainly selective catalytic reduction (SCR) equipment (Kousoulidou et al., 2008).

3.3. Trends in primary and secondary NO_2 at a street location (2000–2020)

The impact of increasing NO_2/NO_x emission ratios on ambient air quality was investigated in a typical Dutch street canyon (the *Pleinweg*) in Rotterdam. This street has a width of 47 m, buildings on both sides of the road with an average height of 12 m, and traffic volumes of 37,000 vehicles per 24 h, 2% of which consist of medium- and heavy-duty vehicles. For 2000–2010, urban background concentrations of NO_2 in Rotterdam were based on measurements, and for 2010–2020 on modelling (Velders et al., 2011a). The contribution of traffic emissions to primary and secondary NO_2 concentrations was modelled for the projected evolution of emissions in situations "with" and "without" application of oxidation catalysts. In the latter case, primary NO_2 emissions of L1 vehicle category were kept at 5% of NO_x emissions in any particular year. This simulates the situation before the oxidation catalyst was introduced in 2000, as presented in Table 1. Results are presented in Fig. 3.

Fig. 3 shows that average annual NO_2 emissions at street locations are projected to decrease as a result of large-scale NO_x reduction measures. However, meeting the annual limit value for NO_2 for 2015 may still be difficult along Dutch inner-urban roads

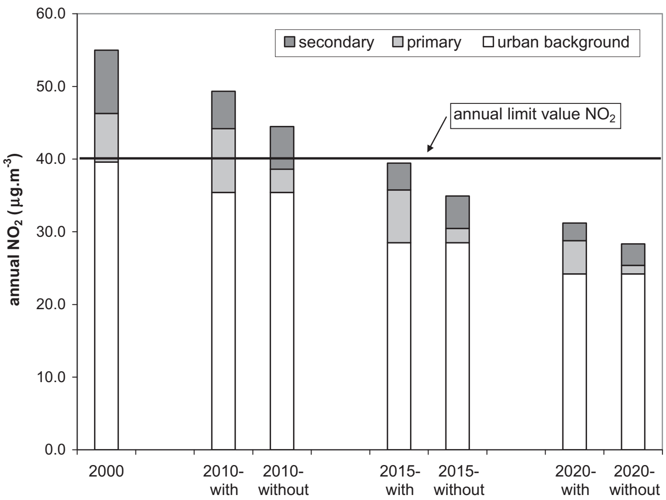

Fig. 3. Annual average NO_2 concentrations in a street canyon (Pleinweg) in Rotterdam for the period 2000–2020 distinguished in urban background, and primary and secondary contribution of traffic emissions "with" and "without" oxidative catalyst.

with high traffic intensities. Firstly, the increasing NO_2/NO_x emission ratios are problematic; their contribution to *local* traffic-related NO_2 increases from 43% (and 57% for secondary NO_2) in 2000 to 65% by 2020, while without the application of oxidation catalysts this would have decreased to 28% by 2020. These results for situations "with catalyst" are in agreement with findings by Anttila et al. (2011) for Finland. Secondly, as concluded by Carslaw et al. (2011), NO_x emissions from passenger vehicles may not decrease by as much as could have been expected based on decreasing emissions following EU regulation, shown in Fig. 2. The latter may be less relevant in the Netherlands where emission factors are established by regulatory testing cycles in combination with on-road measurements (see Section 3.2). Hence, the expected evolution of traffic emissions in the Netherlands seems more realistic than if this were only based on regulatory test cycles.

3.4. Trends in primary NO_2 and exhaust PM emissions from road traffic (2000–2020)

The total number of vehicle kilometres in the Netherlands increased from 114 billion kilometres in 2000 to 130 billion kilometres in 2010, and is expected to further increase to 143 billion kilometres by 2020 (www.cbs.nl). The related emissions from road traffic may be determined from multiplication of emission factors by the number of vehicle kilometres. Accordingly, total primary NO_2 and exhaust PM emissions from all road traffic in the Netherlands were determined for urban roads, non-urban roads and motorways. The results in kilotons per year are presented for the past 2000–2010 period and for the future 2010–2020 period in Fig. 4A ("primary NO_2") and Fig. 4B ("exhaust PM"). Fig. 4A also shows primary NO_2 emissions for a hypothetical situation without the application of oxidation catalysts, i.e., a 5% NO_2/NO_x ratio for passenger cars (i.e., L1 category) over the whole period from 2000 to 2020.

Fig. 4A shows that motorway traffic contributes the most to primary NO_2 emissions from road traffic. The figure also indicates that the maximum amount of primary NO_2 was emitted in 2010 and that – despite a growth in the volume of vehicle kilometres – primary NO_2 emissions by 2020 are expected to be at the level of 2000. This reflects lower projected NO_x emissions from road traffic, especially on motorways after 2010. The dotted lines in Fig. 4A represent estimated primary NO_2 emissions if no oxidation catalysts for diesel-fuelled passenger cars would have been introduced. For example, in 2010, the total annual amount of primary NO_2 emissions from road traffic in the Netherlands would have been 5 kt (without oxidation catalysts) instead of the actual 15 kt (with oxidation catalysts). The implications for NO_2 concentrations at roadside locations are shown in Section 3.5.

Fig. 4B illustrates that road traffic emissions of exhaust PM in 2010 had decreased by a factor of two compared to those of 2000, and are projected to decrease by a factor of two more by the year 2020. This decreasing trend indicates the effective emission reduction in exhaust PM. The dotted lines in Fig. 4B represent estimated exhaust PM in situations without the introduction of oxidation catalysts. Hence, in this hypothetical case, exhaust PM emissions remain at the 2000 level. For example, in 2010, the total annual amount of exhaust PM from road traffic in the Netherlands would have been 7 kt instead of the actual 5 kt. The implications for EC concentrations at roadside locations are shown in Section 3.5.

In epidemiological studies, elevated health effects near traffic locations are associated with traffic-related air pollutants (Brunekreef et al., 2009). It is difficult to separate health effects of NO_2 and exhaust PM, as concentrations of these pollutants are strongly correlated near traffic locations. A change in the composition of these pollutants in ambient air (i.e., the NO_2/EC

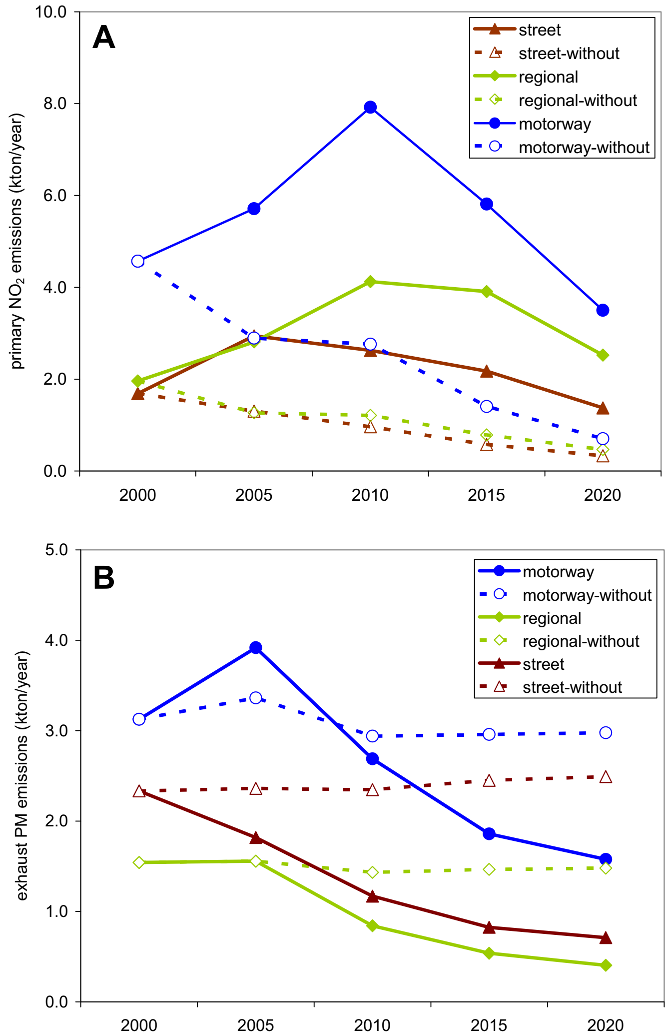

Fig. 4. A: Annual primary NO_2 emission (kt/year) by road traffic on urban roads ("street"), non-urban roads ("regional") and motorways in the Netherlands with established (till 2010) and projected (till 2020) primary NO_2 emissions. Also shown are the NO_2 emissions without oxidation catalyst. B: Annual exhaust-PM emission (kton/year) by road traffic on urban roads ("street"), non-urban roads ("regional") and motorways in the Netherlands with established (till 2010) and projected (till 2020) emissions. Also shown are the exhaust-PM emissions without oxidation catalyst.

concentration ratios) may bring into question whether the formerly established health effects of NO_2 are still valid. This was investigated according to the ratio between primary NO_2 and exhaust PM in traffic emissions, followed by a similar analysis for ambient air at a roadside location, as presented in Section 3.5. Fig. 5 presents the primary NO_2/exhaust PM ratios in emissions from road traffic for various road types in the Netherlands, for the 2000–2020 period. The ratios in Fig. 5 were calculated from the data presented in Fig. 4A (primary NO_2) and 4B (exhaust PM).

Fig. 5 demonstrates that the ratio between primary NO_2 and exhaust PM emissions from road traffic increased over the period from 2000 to 2010 and is expected to further increase up to 2015, followed by a small decrease. The varying changes in ratios for different road types are due to the fact that emission factors change according to driving behaviour and (thus) the road type. Fig. 5 also shows that the ratio between primary NO_2 and exhaust PM emissions would have decreased over time without the introduction of the oxidation catalyst. In Sections 3.5 and 3.6, the impact of the change in traffic emissions on air quality and health near traffic locations is investigated for the 2000–2020 period.

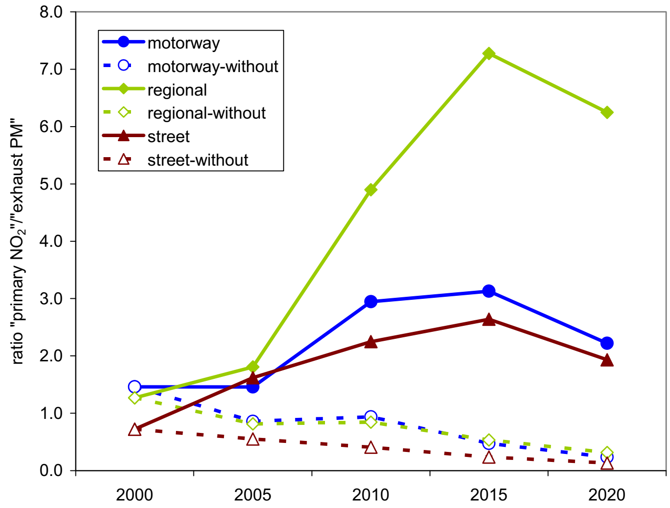

Fig. 5. Ratio of primary NO_2 and exhaust PM emissions by road traffic in the Netherlands for 2000–2020 on urban roads ("street"), non-urban roads ("regional") and motorways. Shown are curves "with" and "without" the application of oxidation catalysts.

3.5. *Trend in NO_2/EC concentration ratios near road traffic locations (2000–2010)*

The impact of the increasing NO_2/NOx fraction on the ratio between NO_2 and EC (as an indicator of exhaust PM) emission concentrations in ambient air is demonstrated by measurements carried out within the Dutch National Air Quality Monitoring Network (www.lml.rivm.nl). Routine measurements of EC have started only recently in the Netherlands, but EC concentrations for previous years may be derived from black smoke data (Keuken et al., 2011). At four regional locations and one street location (i.e., the *Florestraat* in Rotterdam) parallel measurements of NO_2 and black smoke were performed during the 2000–2010 period. The ratio between average annual NO_2 ($\mu g\ m^{-3}$) and EC ($\mu g\ m^{-3}$) were computed for these locations and presented in Fig. 6.

Fig. 6 illustrates that the NO_2/EC concentration ratios in ambient air both at the regional background locations and the street location are in the range of 25–30. Ratios did not change significantly over 2000–2010 (although an increasing trend since 2007 was observed

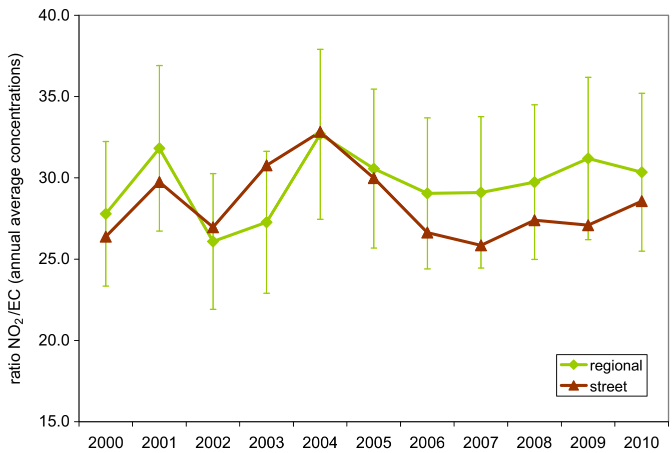

Fig. 6. The ratio in annual average concentrations of NO_2 and EC ($\mu g\ m^{-3}$) at regional locations ($N = 4$) and one street location in the Netherlands for the period 2000–2010. The vertical lines shows the standard deviation in the annual average NO_2/EC at the regional locations.

for the street location) despite the increasing ratio between NO_2 and exhaust PM in traffic emissions, shown in Fig. 5. The elevated ratios in 2003 and 2004, as shown in Fig. 6, can be attributed to meteorological conditions (i.e., exceptional dry and warm years) favouring photochemical NO_2 production.

The data in Figs. 5 and 6 indicate that the increasing ratio between primary NO_2 and exhaust PM in traffic emissions had no measurable impact on the NO_2/EC concentration ratios in ambient air near locations with intense road traffic up to 2010. This is attributed to the relatively high urban background concentration which dominates the NO_2 concentrations, even at locations with intense traffic in the Netherlands (Fig. 3), and the relatively small change in the ratio between NO_2 and exhaust PM in traffic emissions at street locations (Fig. 5). It is noted that the data on street locations in Fig. 6 are based on one monitoring station. More parallel measurements of NO_2 and EC at street and urban locations are required, in the coming years, to validate the aforementioned conclusion. Based on the available data in Fig. 6, it is concluded that the health effects of long-term exposure to NO_2 concentrations, as established in epidemiological studies are still valid (Brunekreef et al., 2009).

3.6. *Health effects of increasing primary NO_2 and lower EC near traffic (2000–2020)*

Finally, the health impact of the increasing share of primary NO_2 emissions was weighted against decreasing exhaust PM emissions. EC concentrations in ambient air were applied to quantify the health impact of these decreasing exhaust PM emissions. To study the impact on health resulting from the introduction of the oxidation catalyst, the contribution of traffic emissions to NO_2 and EC concentrations in the street canyon of *Pleinweg* in Rotterdam was modelled for situations "with" and "without" application of oxidation catalysts. For these two cases, the contribution of traffic emissions to NO_2 and EC concentrations at the Pleinweg location, for 2000–2020, are presented in Fig. 7.

Fig. 7 illustrates that contributions of local traffic emissions to NO_2 are projected to decrease from 16 μg m^{-3} in 2000 to 8 μg m^{-3} by 2020. However, without the introduction of the oxidation catalyst, this could have decreased to 4 μg m^{-3}. As a result of this introduction, the contribution of traffic emissions to EC is projected to decrease from 2.2 μg m^{-3} in 2000 to 0.3 μg m^{-3} by 2020, instead of remaining at the 2000 level.

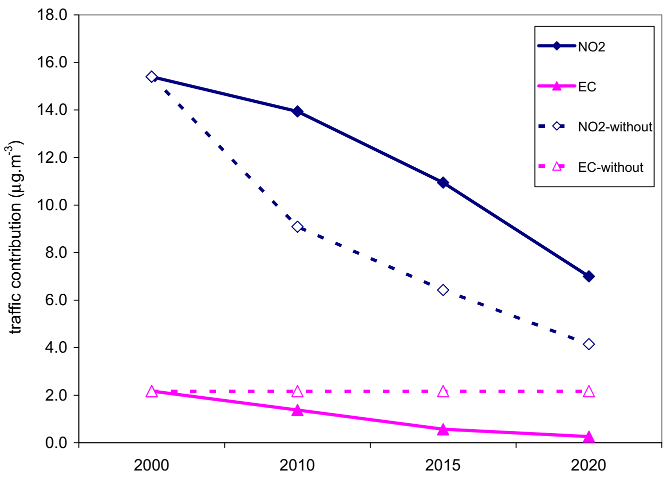

Fig. 7. Contribution of traffic emissions to annual average NO_2 and EC concentrations (μg m^{-3}) in ambient air at the Pleinweg in Rotterdam for the period 2000–2020 "with catalyst" and "without catalyst".

The modelled data for 2010 (Fig. 7) were compared with measurements taken from a street location (*Pleinweg*) and an urban background location in Rotterdam. The differences in the measured concentrations of NO_2 and EC at the street location and the urban background location are considered the "measured" traffic contributions. The measured and modelled average annual traffic contributions were 14.6 and 13.9 μg m^{-3}, respectively, for NO_2, and 1.1 and 1.4 μg m^{-3} for EC. Hence, measured and modelled contributions of traffic emissions are in good agreement for 2010.

Fig. 7 shows that, at the *Pleinweg* location, traffic emissions in situations "with" application of oxidation catalysts, compared to those without, result in *higher* average annual NO_2 concentrations: +5 μg m^{-3} for 2010, +4 μg m^{-3} for 2015, and +3 μg m^{-3} for 2020, and in *lower* average annual EC concentrations: −0.5 μg m^{-3} for 2010, −1 μg m^{-3} for 2015, and −1.5 μg m^{-3} for 2020. The health impact of the "costs" (i.e., relatively higher NO_2 concentrations) versus the "benefits" (i.e., relatively lower EC concentrations) can be assessed by comparing the relative risks of both pollutants. The relative risk indicates the percentage of an adverse health effect (e.g., mortality rate in the population) that may be attributed to exposure to air pollution (Miller and Hurley, 2003). The relative risk of "all-cause mortality" due to long-term exposure to NO_2 and EC concentrations is ten times larger per μg m^{-3} for EC than for NO_2 (Brunekreef et al., 2009; Janssen et al., 2011). The relative risk for NO_2 per μg m^{-3} is 1.007 (95% confidence interval 1.002–1.011) and for EC per μg m^{-3} is 1.06 (95% confidence interval 1.02–1.10). This implies that the costs and benefits of health effects from the introduction of the oxidation catalyst, at the *Pleinweg*, were in balance for 2010, i.e., the ratio between relatively higher NO_2 and relatively lower EC concentrations was ten, but the relative risk related to EC was ten times larger than for NO_2. For 2015 and 2020, however, the benefits are expected to outweigh the costs, as the ratio between relatively higher NO_2 and relatively lower EC will be four (4/1) and two (3/1.5), respectively (Fig. 7).

4. Conclusions and discussion

Due to the introduction of the oxidation catalyst, the share of primary NO_2 in NO_x emissions from diesel-fuelled passenger cars increased from 5% in 2000 to between 55% and 70% in 2010. As a result of more stringent European emission limit values, it is expected that NO_x emissions from road traffic will have decreased since 2010 and will continue to do so. However, actual NO_x emissions may be significantly higher than those based on regulatory emission test cycles. In the Netherlands, road traffic emission factors have been derived from a combination of regulatory test cycles, real-world driving cycles and on-road measurements. Based on these emission factors, it is estimated that the maximum amount of primary NO_2 from road traffic in the Netherlands was emitted in 2010, and that – despite a growth in the volume of vehicle kilometres – primary NO_2 emissions by 2020 will be at the level of 2000. Meanwhile, exhaust PM emissions from road traffic in the Netherlands will decrease from 7 kt in 2000 to 3 kt by 2020.

The impact on air quality of this change in the composition of exhaust emissions was assessed according to the ratio between NO_2 and EC concentrations in ambient air. Compared to PM mass concentrations, EC is a more sensitive indicator of exhaust PM. The monitoring data for 2000–2010 in the Netherlands showed that secondary NO_2 formation still dominated the *total* NO_2 concentrations, even at street locations. Since 2007, the trend for the street location indicates an increasing NO_2/EC ratio, as expected from increasing primary NO_2 and decreasing exhaust PM emissions. However, continued monitoring of NO_2 and EC at a greater number of street locations, in the coming years, is required for more conclusive conclusions. Hence, there are no indications that the correlation between the various traffic-related air pollutants in *ambient air* has changed significantly because of the introduction of the oxidation catalyst. It is concluded that the health effects of NO_2 from traffic emissions as established in epidemiological studies are still valid.

The health impact of the relatively higher NO_2 and lower EC concentrations, as compared to a situation without the application of oxidation catalysts, was estimated for a street location in the Dutch city of Rotterdam, for the period from 2000 to 2020. The ratio between the relatively higher NO_2 and lower EC concentrations was a factor of ten in 2010, and is expected to be a factor of four by 2015 and a factor of two by 2020. The adverse health effects per ug m^{-3} are ten times larger for EC than for NO_2. This indicates that the costs and benefits of the introduction of the oxidation catalyst were balanced in 2010, but that the benefits will become larger in the period from 2010 to 2020. A complication of this analysis is the high correlation between NO_2 and EC concentrations, as shown in epidemiological studies. This hampers the determination of the relative risks related to the individual components. Therefore, the cost–benefit analysis is only valid if we assume that NO_2 and EC are equally, and in time consistently, related to the causal agent(s) for the associated health effects near traffic locations.

The results from this study are probably relevant for other locations with high traffic intensities of diesel-fuelled passenger vehicles. From a health perspective, it is concluded that, at these locations, the introduction of the oxidation catalyst is beneficial, but that from a regulatory perspective, increasing primary NO_2 complicates compliance with the average annual limit value for NO_2. This indicates that additional measures to reduce traffic emissions (e.g., speed management, car-free zones) might be necessary to meet air quality standards for NO_2 at street locations.

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