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To cite this article: Erika von Schneidemesser *et al* 2017 *Environ. Res. Lett.* **12** 114025

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# Environmental Research Letters



## LETTER

# Potential reductions in ambient NO<sub>2</sub> concentrations from meeting diesel vehicle emissions standards

### OPEN ACCESS

#### RECEIVED

3 April 2017

#### REVISED

21 August 2017

#### ACCEPTED FOR PUBLICATION

14 September 2017

#### PUBLISHED

8 November 2017

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**Keywords:** urban, nitrogen oxides, vehicle standards, emissions standards, air quality exceedances, WRF-Chem, air pollution

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## Abstract

Exceedances of the concentration limit value for ambient nitrogen dioxide (NO<sub>2</sub>) at roadside sites are an issue in many cities throughout Europe. This is linked to the emissions of light duty diesel vehicles which have on-road emissions that are far greater than the regulatory standards. These exceedances have substantial implications for human health and economic loss. This study explores the possible gains in ambient air quality if light duty diesel vehicles were able to meet the regulatory standards (including both emissions standards from Europe and the United States). We use two independent methods: a measurement-based and a model-based method. The city of Berlin is used as a case study. The measurement-based method used data from 16 monitoring stations throughout the city of Berlin to estimate annual average reductions in roadside NO<sub>2</sub> of 9.0 to 23  $\mu\text{g m}^{-3}$  and in urban background NO<sub>2</sub> concentrations of 1.2 to 2.7  $\mu\text{g m}^{-3}$ . These ranges account for differences in fleet composition assumptions, and the stringency of the regulatory standard. The model simulations showed reductions in urban background NO<sub>2</sub> of 2.0  $\mu\text{g m}^{-3}$ , and at the scale of the greater Berlin area of 1.6 to 2.0  $\mu\text{g m}^{-3}$  depending on the setup of the simulation and resolution of the model. Similar results were found for other European cities. The similarities in results using the measurement- and model-based methods support our ability to draw robust conclusions that are not dependent on the assumptions behind either methodology. The results show the significant potential for NO<sub>2</sub> reductions if regulatory standards for light duty diesel vehicles were to be met under real-world operating conditions. Such reductions could help improve air quality by reducing NO<sub>2</sub> exceedances in urban areas, but also have broader implications for improvements in human health and other benefits.

## 1. Introduction

Air pollution is a pressing environmental issue, and not just in developing countries. More than 85% of the urban population in Europe lives in areas where the World Health Organization (WHO) guidelines are exceeded for air pollutants such as particulate matter (PM<sub>2.5</sub>) or ozone (O<sub>3</sub>) (EEA 2016). Nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) are important contributors to secondary formation of air pollutants, including to concentrations of PM and O<sub>3</sub>. In addition, adverse human health effects such as increases in all-cause mortality, and respiratory and cardiovascular effects have been associated with both short-term and long-term exposure to nitrogen dioxide (NO<sub>2</sub>)

(Faustini *et al* 2014, Mills *et al* 2015). A large number of cities in Europe struggle to meet their NO<sub>2</sub> targets, among those many cities in Germany, including Berlin (BSV 2017). More specifically, more than half of all roadside measurement stations in Germany have not met the annual limit value of 40  $\mu\text{g m}^{-3}$  for the past decade (Minkos *et al* 2017). These exceedances are generally attributed to vehicle emissions, still one of the largest sources of air pollutant emissions, and in this case specifically diesel vehicles (EEA 2016). While the implementation of a defeat device by Volkswagen (VW) during emissions testing was recently discovered by the research of West Virginia University (Thompson *et al* 2014), the inability of diesel vehicles to meet the emission standards in Europe under on-road

conditions is acknowledged (EEA 2016, Fontaras *et al* 2014, Hagman *et al* 2015, ICCT 2014, Rexeis *et al* 2013). The Euro 5 and Euro 6 diesel passenger vehicles have been documented to have real-world  $\text{NO}_x$  emissions up to 5x and 4–20x higher, respectively, than the allowed emission levels (EEA/EMEP 2013, Fontaras *et al* 2014, Hagman *et al* 2015). Additionally, current national reported emissions (which are also used as the basis for emission inventories that are implemented in air quality models) are typically calculated using emission factors (EF) from the Handbook Emission Factors for Road Transport (HBEFA) (Rexeis *et al* 2013), as in Germany, or the COmputer Programme to calculate Emissions from Road Transport (COPERT) (Gkatzoflias *et al* 2012). The EFs in the HBEFA report are more reflective of true on-road emissions than the regulatory targets, and were found to capture the higher emissions from diesel vehicles better than COPERT (Fontaras *et al* 2014). For example, the EF for  $\text{NO}_x$  emissions from Euro 5 light duty diesel vehicles from HBEFA for urban driving conditions is  $0.97 \text{ g km}^{-1}$  (Rexeis *et al* 2013). Reported real-world driving EFs for Euro 5 diesel vehicles from a number of studies ranged from  $0.35 \text{ g km}^{-1}$  to  $1.12 \text{ g km}^{-1}$  under different driving conditions (e.g. urban, motorway, or averages) (Carslaw *et al* 2011, Fontaras *et al* 2014, Weiss *et al* 2012). The EU Euro 5 regulatory standard for  $\text{NO}_x$  from diesel vehicles is  $0.18 \text{ g km}^{-1}$  (EC 2007). The United States standard set by the Environmental Protection Agency (EPA) for light duty diesel vehicles is  $0.043 \text{ g km}^{-1}$ .

Several studies have been published recently investigating the effect of  $\text{NO}_x$  emissions from diesel vehicles, in some cases due to the VW emissions control defeat device, on air pollution and the implications for human health and social cost in the United States (Barrett *et al* 2015, Holland *et al* 2016, Wang *et al* 2016) and Europe (Brand 2016, Chossiere *et al* 2017), as well as addressing the issues more generally (Rojas-Rueda and Turner 2016). The studies focused on the US estimate that 46 and 59 excess deaths and ca. \$430 or 450 million excess damages result from the excess  $\text{NO}_x$  emissions from the affected diesel vehicles between 2008 or 2009 and 2015, respectively (Barrett *et al* 2015, Holland *et al* 2016). Additionally, one study evaluated the effects for California and found that the additional ‘hidden’  $\text{NO}_x$  emissions (difference between the actual on-road  $\text{NO}_x$  emission factor and the testing emission factor) would result in 12 excess deaths during the same 2009–2015 time period as the US studies, with the majority of the increase in mortality in metropolitan areas (Wang *et al* 2016). In work that parallels the US study by Barrett *et al* (2015), Chossiere *et al* (2017) estimated the cost of excess  $\text{NO}_x$  emissions from VW vehicles in Germany to be 1200 premature deaths (1.9 billion EUR) in Europe. The study by Brand (2016) highlights the potential trade-offs between human health and climate change mitigation in policies related to diesel vehicles in the

UK (not just those affected by the defeat device), showing that the excess  $\text{NO}_x$  emissions from diesel vehicles are significant and that the benefit to air quality in reducing these emissions would be much larger and outweigh any of the few potential carbon disbenefits, based on a comparison of damage costs. Holland *et al* (2016) reached a similar conclusion, finding that the estimated damages (in this case limited to those VW vehicles with defeat devices) greatly outweigh any possible benefits from reduced  $\text{CO}_2$  emissions resulting from increased fuel economy.

This study explores a broader perspective beyond that of the cars affected by the defeat devices and investigates the potential benefits to air quality based on scenarios in which the light duty diesel vehicle fleet meets the regulatory standards under real-world driving conditions. Two independent approaches are used: (1) an estimation based on ambient  $\text{NO}_2$  concentrations from monitoring stations, and (2) a sensitivity study using a chemical transport model. The regional focus is on Berlin, Germany, but includes comparisons to other urban areas in Europe that indicate the potential of such regulation and broader applicability of the results.

## 2. Methods

### 2.1. Emissions

The emission inventory used for both the observation-based calculations and the chemical transport modeling scenarios was the TNO-MACCCIII inventory, an update to the previous version II (Kuenen *et al* 2014). To estimate the total amount of  $\text{NO}_x$  emissions originating from diesel vehicles in Berlin, the following calculations were carried out.

This study specifically focuses on the impact of diesel light-duty vehicles (LDV) on air quality. Therefore, we estimate the total  $\text{NO}_x$  emissions from diesel LDV exhaust in the greater Berlin area based on the TNO-MACCCIII inventory, for which the grid cells over Berlin were extracted. The total amount of  $\text{NO}_x$  emissions for Berlin was 24 598 kT annually, of which 8944 kT are from road transport. 8094 kT are attributed to  $\text{NO}_x$  emissions from diesel vehicle exhaust. The heavy-duty vehicle (HDV) versus light-duty vehicle (LDV) split provided by TNO attributed 43% of diesel vehicles to the LDV category which is based on the national average for Germany (Kuenen 2015). Information from the city of Berlin indicates that 80% of diesel vehicles in the city of Berlin are LDV (BSV 2013). Using these two percentages we estimated that of the 8094 kT  $\text{NO}_x$  emissions attributed to diesel vehicle exhaust (LDV + HDV together), either 3464 kT (national average) or 6446 kT (Berlin urban area average) originate from LDV. Using both estimates of the percent of LDV diesel provides upper and lower estimates to explore the sensitivity of our calculations to such numbers.

**Table 1.** HBEFA emission factors (EF) compared to the regulatory standards for European and US diesel passenger vehicles, shown for the three driving conditions and an overall average. Also presented is the ratio of regulatory standard EF to current HBEFA EF.

	HBEFA EF ( $\text{g km}^{-1}$ )		Regulatory Standards ( $\text{g km}^{-1}$ )			Ratio (reg. Std/HBEFA)	
	Euro 5	Euro 6	Euro 5	Euro 6	US EPA Tier 2 Bin 5	Euro 5	US EPA Std <sup>a</sup>
Urban	0.97	0.26				0.19	0.044
Rural	0.63	0.14				0.28	0.068
Motorway	0.79	0.28				0.23	0.054
Average	<b>0.80</b>	<b>0.23</b>	0.18	0.08	0.043	<b>0.23</b>	<b>0.055</b>

<sup>a</sup> The US EPA Std is implemented as the ‘best-available technology’ scenario.

## 2.2. Emission factors

To evaluate how air quality would be improved if current emission standards for diesel LDV were met under real-world driving conditions, we compare emission factors used for nationally-reported emissions (a proxy for on-road emissions under real-world conditions) and the emission factors dictated by regulations. The emission factors (EFs) currently used for national reporting of emissions in Europe come from the HBEFA report, which includes  $\text{g km}^{-1}$  estimates of  $\text{NO}_x$  emissions for Euro 5 and Euro 6 vehicles (Rexeis *et al* 2013). EF values are provided for urban, rural, and motorway driving conditions. As an example, the HBEFA EF for Euro 5 diesel LDV is  $0.9719 \text{ g km}^{-1}$  for urban driving. The Euro 5 standard is  $0.18 \text{ g km}^{-1}$ . The US EPA Tier 2 Bin 5 standard<sup>3</sup> (hereafter US EPA standard) for diesel LDV is  $0.043 \text{ g km}^{-1}$ . The Euro 6 standard is  $0.08 \text{ g km}^{-1}$ . These emission factors are summarized in table 1. In our calculations we considered cases where diesel LDV emissions under real-world conditions meet the Euro 5, representative of a less ambitious modern standard, and US EPA standard, which is currently the most stringent for diesel LDV emissions, referred to here as the ‘best-available technology’ scenario. Note that the Euro 6 standard is not explicitly considered here, as it represents an intermediate point between the two.

To calculate a simple factor of possible improvement in terms of EFs, we first assume that all diesel LDV are emitting as Euro 5 vehicles at the level reported by the HBEFA EF for the 2011 and 2014 time frames considered in this study. While this assumption does not capture all of the emissions from those vehicles that are Euro 4 or lower, according to data from Berlin, over 70% of all diesel LDV are Euro 4 or better (the reported values unfortunately do not include a category for Euro 5 or better) (BSV 2013), and is therefore not unreasonable. Furthermore, by assuming that the existing fleet is lower-emitting than in reality, we ensure that our results will represent a conservative estimate of the potential benefits of further reductions in the emissions due to meeting the stricter standards. Finally, a calculation was carried out, applying higher EFs for the ca. 27% of the fleet that were Euro 3 or

lower, to assess the sensitivity of this assumption, and the results showed negligible differences.

To then calculate the change in EFs if the regulatory standards were met, we compared the HBEFA EFs for each of the driving conditions (urban, rural, and motorway) to both the Euro 5 and US EPA standard, and calculated a simple average over the three driving conditions to come up with a coarse reduction factor that would be expected. (The amount of vehicle kilometers traveled by passenger cars in Germany are roughly equal among the three driving conditions (Ehlers *et al* 2016).) If current Euro 5-certified diesel LDVs achieved Euro-5 level  $\text{NO}_x$  emissions (on a per km basis, expressed as an EF) under real-world driving conditions, the reduction in emissions would be 77% (calculated as the ratio Euro 5 to HBEFA emission factors). Similarly, for the best-available technology case (i.e. if the US EPA standard were met), the EF would be 95% lower (table 1). The expected reduction in emissions from the fleet meeting the Euro 5 or US EPA standards would be larger still if Euro 4 and higher-emitting vehicles were included in our emissions estimate for the current fleet composition.

## 2.3. Potential change evaluated based on ambient monitoring data

The factors from table 1 were used to calculate expected total annual  $\text{NO}_x$  emission reductions using Berlin as a case study, as shown in table 2. The reduction in road transport emissions attributed to diesel LDV was calculated by multiplying by the emissions with the reduction factor (‘Ratio’ column in table 1) for both the Euro 5 and the best-available technology case. This calculation was done using the two different estimates of the percent of diesel LDV emissions for Berlin (table 2 and section 2.1). The expected reductions in traffic  $\text{NO}_x$  emissions are shown in table 2, as absolute kT of emissions and as percent reductions. Similarly, a reduction in total urban  $\text{NO}_x$  emissions was calculated. This used the same (absolute) estimated road transport emissions reductions, but compared it to all  $\text{NO}_x$  emission sources in the urban area, rather than just the total road transport  $\text{NO}_x$ , in Berlin. This results in a reduction of 11%–25% in the total  $\text{NO}_x$  emissions for the city (table 2). All of the values and changes are discussed in more detail in the results and discussion section.

Finally, to estimate how the calculated reductions in emissions would affect ambient concentrations, the

<sup>3</sup> Tier 2 refers to cars and light-duty trucks; Bin 5 refers to the certification bins of different stringency, of which Bin 5 represents the fleet average for  $\text{NO}_x$  that has to be met.

**Table 2.** Estimated annual NO<sub>x</sub> emissions for Berlin from diesel LDVs under two different assumptions for the diesel fleet composition (national vs. city) for the current fleet, and for scenarios in which diesel LDVs meet the Euro 5 standard or best-available technology (US EPA standard) during real-world driving. The percent emission reductions relative to the current emissions situation is also shown for both cases. The total road transport NO<sub>x</sub> emissions in Berlin are 8944 kT.

	National fleet (43% LDV)			City fleet (80% LDV)		
	Total NO <sub>x</sub> emissions (kT) <sup>a</sup>	Reduction in traffic emissions (%)	Reduction in total urban emissions (%)	Total NO <sub>x</sub> emissions (kT) <sup>a</sup>	Reduction in traffic emissions (%)	Reduction in total urban emissions (%)
Current road transport from LDV diesel <sup>b</sup>	3464	—	—	6446	—	—
Euro 5	797	30	11	1483	55	20
Best-available technology	190	40	13	355	75	25

<sup>a</sup> Expressed as kT NO<sub>2</sub>.

<sup>b</sup> Based on the TNO-MACCCIII inventory for 2011 (the most recent year available at the time of writing).

percent reductions in road transport and total city NO<sub>x</sub> emissions were used in combination with monitoring data for NO<sub>x</sub> and NO<sub>2</sub> to calculate possible reductions in ambient concentrations at the roadside and for the Berlin urban area more generally. We calculated the expected reduction in NO<sub>x</sub> concentration at the roadside based on the ‘roadside increment’, and the reductions in background NO<sub>x</sub> concentration over the entire urban area based on the ‘urban background increment’. The roadside increment was defined as the roadside concentration minus the urban background concentration. Similarly, the urban background increment was defined as the urban background concentration minus the rural background concentration, using averages of those monitoring stations from the Berlin area. We assume that reductions in traffic emissions lead to proportional reductions in the roadside increment, and that reductions in total urban emissions similarly lead to proportional reductions in the urban background increment. Specifically, for the urban background, we assume in addition that we can estimate the NO<sub>x</sub> attributed to traffic using the traffic NO<sub>x</sub> emissions share of total NO<sub>x</sub> emissions in the urban background. Note that although the emission standards are for NO<sub>x</sub>, the limit values on ambient concentration for Europe are for NO<sub>2</sub>, so we estimate the changes in NO<sub>2</sub> concentrations here.

Given that we consider fractional reductions, the relative amount of change is applied to NO<sub>x</sub>. NO<sub>2</sub>/NO<sub>x</sub> ratios have been shown to depend on a number of factors, including background NO<sub>x</sub>, background O<sub>3</sub>, local emissions, as well as vehicle fleet composition and vehicle speeds (that influence the amount of NO<sub>x</sub> emitted as primary NO<sub>2</sub>), as well as meteorology (Carslaw and Beevers 2005, Carslaw and Carslaw 2007, Clapp and Jenkin 2001). In order to account for this, we calculate the change in NO<sub>2</sub> using the relationship between NO<sub>2</sub> and NO<sub>x</sub>, which for the data used in this study was determined to be linear. For urban background sites, the NO<sub>2</sub>/NO<sub>x</sub> slope is 0.82 ( $r^2 = 0.98$ ), while for roadside stations the slope is 0.48 ( $r^2 = 0.94$ ). The values mentioned here are for July; for a complete list of slopes, for January, July, and the annual

average, see table S4 and figure S1. An analysis of the NO<sub>2</sub>/NO<sub>x</sub> ratios at the urban background sites as well as the roadside monitoring sites, show consistently similar values at each site type across the city, despite differences in traffic intensities at each of these sites. For the chemical transport model simulations, NO<sub>x</sub> chemical cycles are included in the calculations (see section 2.4), meaning that their effects are included in our results at the urban background scale. The data used was hourly NO<sub>x</sub> and NO<sub>2</sub> from 16 monitoring stations in Berlin: 6 roadside, 5 urban background, and 5 rural stations for the year 2014 (Stülpnagel *et al* 2015). The rural stations used here are located on the periphery of Berlin<sup>4</sup>. The increments were calculated based on daily mean values averaged over a year, as well as over two individual months (January and July). The July calculation allowed for a comparison with the chemical transport model results which simulated July only. For the calculations for the individual months, sector-specific time factors were applied to the annual emissions before calculating the difference in total urban emissions. Both annual average and July values are presented in the results section; January values are only included in the SI. Standard deviation was propagated to provide an estimate of temporal and spatial variability within the observation-based estimates.

Additional calculations were carried out for the Euro 6 standard (EF: 0.08 g km<sup>-1</sup>) and the Euro 6 conformity factor<sup>5</sup> for September 2017 (EF: 0.168 g km<sup>-1</sup>) (EC 2015) and are included in supplemental information (table S3) available at [stacks.iop.org/ERL/12/114025/mmedia](http://stacks.iop.org/ERL/12/114025/mmedia). These results are not detailed in the text as they fall between the Euro 5 and best-available technology results. The relationship is roughly linear between the EFs and the reductions expected. The current Euro 5 standard and the Euro 6 conformity factor results show minimal differences, given the similarities in their EFs.

<sup>4</sup> For more information see [www.stadentwicklung.berlin.de/umwelt/luftqualitaet/luftdaten/index.shtml](http://www.stadentwicklung.berlin.de/umwelt/luftqualitaet/luftdaten/index.shtml).

<sup>5</sup> The conformity factor is the first step in stricter standards, where car manufacturers will need to bring down the discrepancy between ‘real driving emissions’ and the Euro 6 standard.

**Table 3.** Estimated reduction in ambient concentrations of daily mean NO<sub>2</sub> at the roadside and for the urban background of Berlin from the observation-based calculations. Estimates for both the national and city level fraction of LDV diesel are included. Values are monthly average (July) daily mean values and standard deviation, and for comparison, in parentheses annual average daily mean values and standard deviation. Also shown are estimated reductions in monthly average (July) daily mean NO<sub>2</sub> compared to the base case scenario for the European and Berlin simulations. In both model simulations the national (43%) LDV fraction was used for Germany. For the model simulations the 25th and 75th percentiles are indicated by the square brackets. For the European simulations reductions are shown for two chemical mechanisms, RADM2 (MOZART). All units in  $\mu\text{g m}^{-3}$ .

Scenario	Observation-based calculations				Berlin simulation		European simulation		
	National fleet (43% LDV)		City fleet (80% LDV)		National fleet (43% LDV)		National fleet (varies by country)		
	Roadside	Urban background	Roadside	Urban background	Greater Berlin area	Berlin city <sup>a</sup>	Greater Berlin area	Benelux	Paris
<b>Euro 5</b>	10 ± 2.5 (9.0 ± 2.8)	1.3 ± 0.43 (1.2 ± 0.65)	19 ± 4.6 (17 ± 5.2)	2.3 ± 0.80 (2.2 ± 1.2)	—	—	—	—	—
<b>Best-available technology</b>	14 ± 3.3 (12 ± 3.8)	1.6 ± 0.53 (1.5 ± 0.80)	26 ± 6.2 (23 ± 7.0)	2.9 ± 0.99 (2.7 ± 1.5)	1.6 [0.8, 2.1]	2.0 [1.1, 2.8]	1.7 [1.2, 2.0] (2.0 [1.4, 2.4])	2.1 [1.5, 2.5] (2.7 [1.7, 3.3])	4.7 [3.6, 5.2] (5.9 [4.3, 6.7])

<sup>a</sup> The values reported for the Berlin city (model simulation) are most comparable to the urban background estimates (observation-based calculations).

#### 2.4. Potential change evaluated based on chemical transport model simulations

In addition to estimating the effect of emission reductions on NO<sub>2</sub> concentrations using ambient monitoring data (section 2.3), we also implemented emission reduction scenarios in a chemical transport model, the chemistry version of the Weather Research and Forecasting Model (WRF-Chem) (Fast *et al* 2006, Grell *et al* 2005, Skamarock *et al* 2008).

Two setups of the WRF-Chem model have been used, one setup covering Europe (European simulations) and one setup focusing on Berlin (Berlin simulations). Both setups are included in order to assess the changes at a regional level over Europe and for the urban area of Berlin. With each setup, two model simulations using the chemical mechanism RADM2 were done for July 2011, a base case using emissions for 2011 and a simulation assuming the best-available technology. In addition, the European simulations were repeated with a different chemical mechanism (MOZART-4) (see supplementary material). The US EPA emission standard was chosen for our best-available technology emission reduction scenario since it is the most stringent of the emission standards. Thus, the scenarios illustrate the potential for improvements in air quality achievable (at the regional to urban background scale) based on standards that should, in principle, be achievable today.

The emission reductions for the model simulations were calculated analogously to those for Berlin, using the factors in table 1. To calculate the appropriate reduction factor in diesel road transport emissions (the level at which emissions in TNO-MACCI are reported) for each country, we use the fraction of diesel LDV vehicles (i.e. as a fraction of total diesel vehicles, LDV+HDV) based on each country's fraction of diesel LDV (national country estimates) (Kuenen 2015).

Both model setups are described in more detail in the supplemental information as well as in Mar *et al* (2016) (European simulations) and Kuik *et al* (2016) (Berlin simulations), and the main features are summarized in table S1 in the supplementary material.

##### 2.4.1. Modeled reductions in ambient concentrations

From the model results, daily mean concentrations of chemical species were calculated. In order to estimate the reductions in ambient NO<sub>2</sub> concentrations under the best-available technology scenario, the difference between the base case and the EPA case was calculated. From this, the July 2011 monthly mean differences are evaluated. Spatially averaged reductions were then calculated for the greater Berlin area (larger dashed rectangle in figure 2) and Berlin city (inner dashed rectangle in figure 2). In addition, 25th and 75th percentiles of the differences in daily means were calculated for each grid cell. Their averages over the greater Berlin area as well as the Berlin city center are given as an indication for the average temporal variability within the respective region.

## 3. Results and discussion

### 3.1. Potential concentration changes based on ambient monitoring data

The change in ambient NO<sub>2</sub> concentrations based on calculations using the monitoring station data and the two different assumptions for how much diesel LDV emissions contribute to the total diesel vehicle fleet (national vs. city fleet composition) are shown in table 3. The reduction in the annual average of daily mean NO<sub>2</sub> concentration at the roadside in the best-available technology scenario was estimated to be  $12 \pm 3.8 \mu\text{g m}^{-3}$  or  $23 \pm 7.0 \mu\text{g m}^{-3}$  (mean ± standard

deviation) assuming the national and city fleet LDV percentages, respectively. For context these reductions would be largely affecting the average difference between the observed roadside and urban background concentrations of  $\text{NO}_2$  which was  $26 \mu\text{g m}^{-3}$  and the annual mean roadside  $\text{NO}_2$  concentration of  $51 \mu\text{g m}^{-3}$ .<sup>6</sup> The reduction in the annual average of daily mean urban background  $\text{NO}_2$  concentration was  $1.5 \pm 0.80 \mu\text{g m}^{-3}$  and  $2.7 \pm 1.5 \mu\text{g m}^{-3}$  using the national and city LDV percentages, respectively. Similarly, for context, the average difference between the observed urban background and rural  $\text{NO}_2$  concentrations was  $12 \mu\text{g m}^{-3}$  with an annual mean urban background  $\text{NO}_2$  concentration of  $26 \mu\text{g m}^{-3}$ .

The monthly reductions were similar to the annual values. Specifically, the reductions calculated for July (table 3) were generally somewhat higher than those for the year, while the January values (table S3) were somewhat lower reductions. The differences were largest for roadside concentrations. For example, the monthly average of daily mean  $\text{NO}_2$  concentration if Euro 5 standards were met (national LDV percentage), showed estimated reductions of  $8.3 \pm 2.3 \mu\text{g m}^{-3}$  for January, compared to  $10 \pm 2.5 \mu\text{g m}^{-3}$  for July, and  $9.0 \pm 2.8 \mu\text{g m}^{-3}$  annually. The lesser reductions in January are linked to higher urban background concentrations, corresponding to a reduced roadside increment, as well as larger emissions from energy and non-industrial combustion. Overall, the reductions in January are about 20%–30% lower than in July, or ca. 10%–25% (5%–15%) lower (higher) than the annual reductions for January (July). The reductions calculated for July for the Euro 5 standard scenario were ca. 72% of the reductions calculated for the best-available technology scenario at the roadside, and ca. 80% for the urban background, for both the national fleet and city fleet estimates. In all cases, the standard deviation of the calculated values overlapped between the two scenarios.

Of the six roadside monitoring stations in Berlin providing hourly values, all six exceeded the annual  $\text{NO}_2$  limit value of  $40 \mu\text{g m}^{-3}$  in 2014, with annual average concentrations between  $42$  and  $61 \mu\text{g m}^{-3}$  (Stülpnagel *et al* 2015). The potential reduction in the annual average  $\text{NO}_2$  roadside increment calculated in this study for the Euro 5 standard was  $9.0 \pm 2.8 \mu\text{g m}^{-3}$  (national LDV percentage) and  $17 \pm 5.2 \mu\text{g m}^{-3}$  (city LDV percentage), with even higher reductions possible with stricter standards such as Euro 6 or US EPA standards. Such reductions have significant potential to help achieve the ambient air quality limit values, especially considering the values in this study are likely conservative estimates.

<sup>6</sup> The sum of the difference between the roadside and urban background concentrations ( $26 \mu\text{g m}^{-3}$ ) and the annual mean urban background ( $26 \mu\text{g m}^{-3}$ ) should equal the stated annual mean roadside  $\text{NO}_2$  concentration ( $51 \mu\text{g m}^{-3}$ ). For consistency with significant figures, this is not the case owing to rounding.

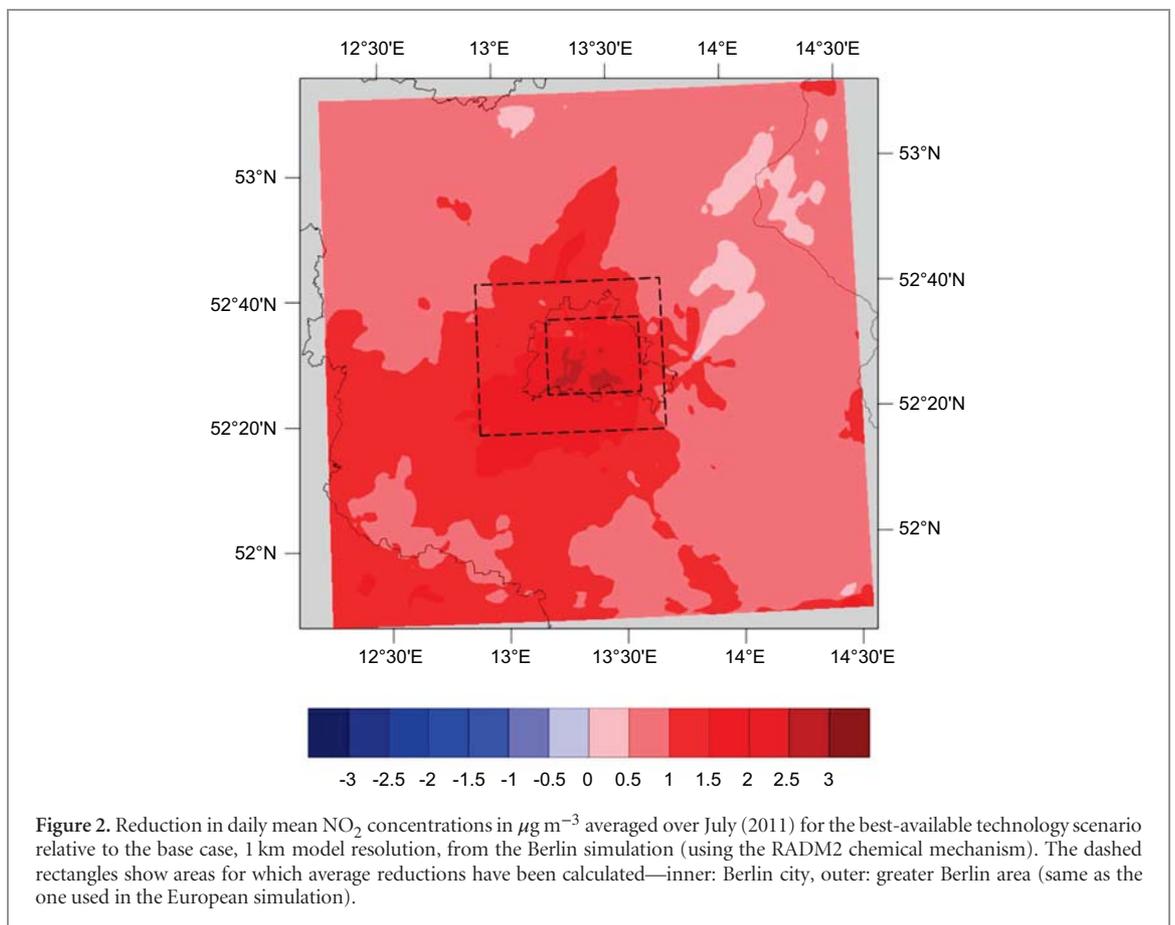
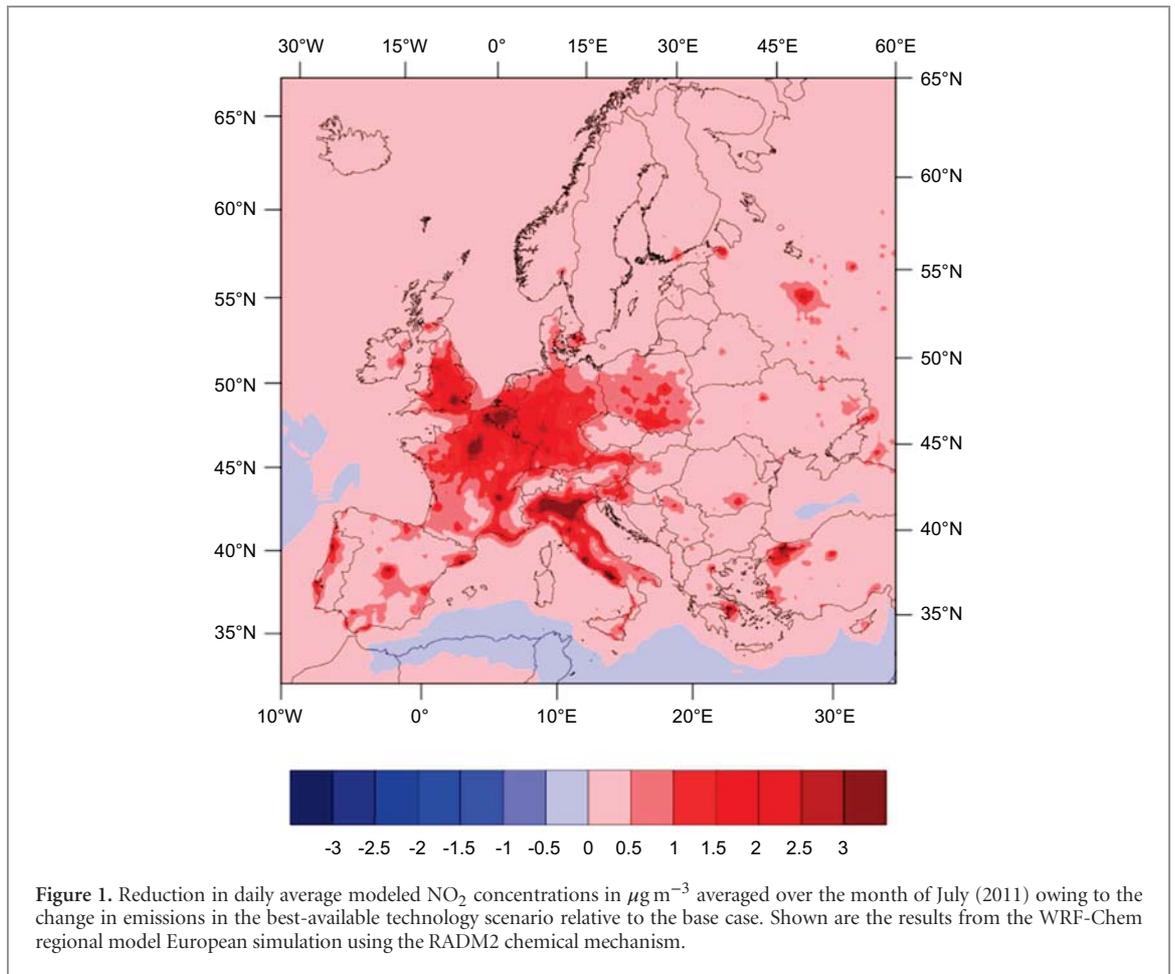
### 3.2. Model simulations using WRF-Chem

The model results showing the change in  $\text{NO}_2$  concentrations from the European simulation and the Berlin simulation are shown in figures 1 and 2, respectively. Both model setups calculated the change in  $\text{NO}_2$  over the greater Berlin area (larger dashed rectangle in figure 2), for which the daily mean  $\text{NO}_2$  averaged over the month of July showed reductions of  $1.7 \mu\text{g m}^{-3}$  and  $1.6 \mu\text{g m}^{-3}$ , respectively. The modeled reductions over the greater Berlin area agreed well between the European and Berlin simulations. Furthermore, a slightly larger reduction of  $2.0 \mu\text{g m}^{-3}$  was found for the city of Berlin in the high resolution simulation (inner dashed rectangle in figure 2). The European simulation was also carried out with the MOZART-4 chemical mechanism, in addition to the RADM2 mechanism and found to produce similar results (table 3).

As a further comparison, the modeled change in average  $\text{NO}_2$  concentrations for the BeNeLux region and Paris, both urban centers with significant vehicular emissions, were also calculated from the regional model results. These urban agglomerations showed larger reductions of  $2.1 \mu\text{g m}^{-3}$  and  $4.7 \mu\text{g m}^{-3}$ , respectively, relative to the change observed over the greater Berlin area. This is reasonable as the populations of these regions are much higher than those for Berlin.

The modeled results for the daily mean  $\text{NO}_2$  concentrations from five urban and four suburban/rural sites in Berlin are shown in figure 3 for the base case and the best-available technology case, with a comparison to observations. The difference between the base case and best-available technology case shows substantial reductions in the daily mean values, with the largest differences often occurring when  $\text{NO}_2$  concentrations peak. This further indicates the potential of such improvements to reduce  $\text{NO}_2$  exceedances.

The comparison between modeled and observed  $\text{NO}_2$  concentrations further shows that the model underestimates observed  $\text{NO}_2$  concentrations (figure 3 and table S2 in the SI). While the model bias at suburban and rural background stations ranges between  $-5\%$  and  $+6\%$  (July 2011 average), the model consistently underestimates  $\text{NO}_2$  concentrations in the urban background between  $-25\%$  and  $-31\%$  ( $-3.4$  to  $-6.5 \mu\text{g m}^{-3}$ ). The difference in model performance for suburban and urban areas shows that the negative bias for urban background sites might be related to a general underestimation of traffic emissions in the emission inventory, which might for example be due to an underestimation of emission factors or the amount of congestion within the city, but might also be due to model limitations in representing the chemistry or boundary layer height (e.g. Giordano *et al* 2015, Karl *et al* 2017, Knote *et al* 2015). As the difference between base run and best-available technology case is generally largest when (modeled) concentrations are higher, the underestimation in simulated concentrations might lead to an underestimation of the effect of emission reductions.



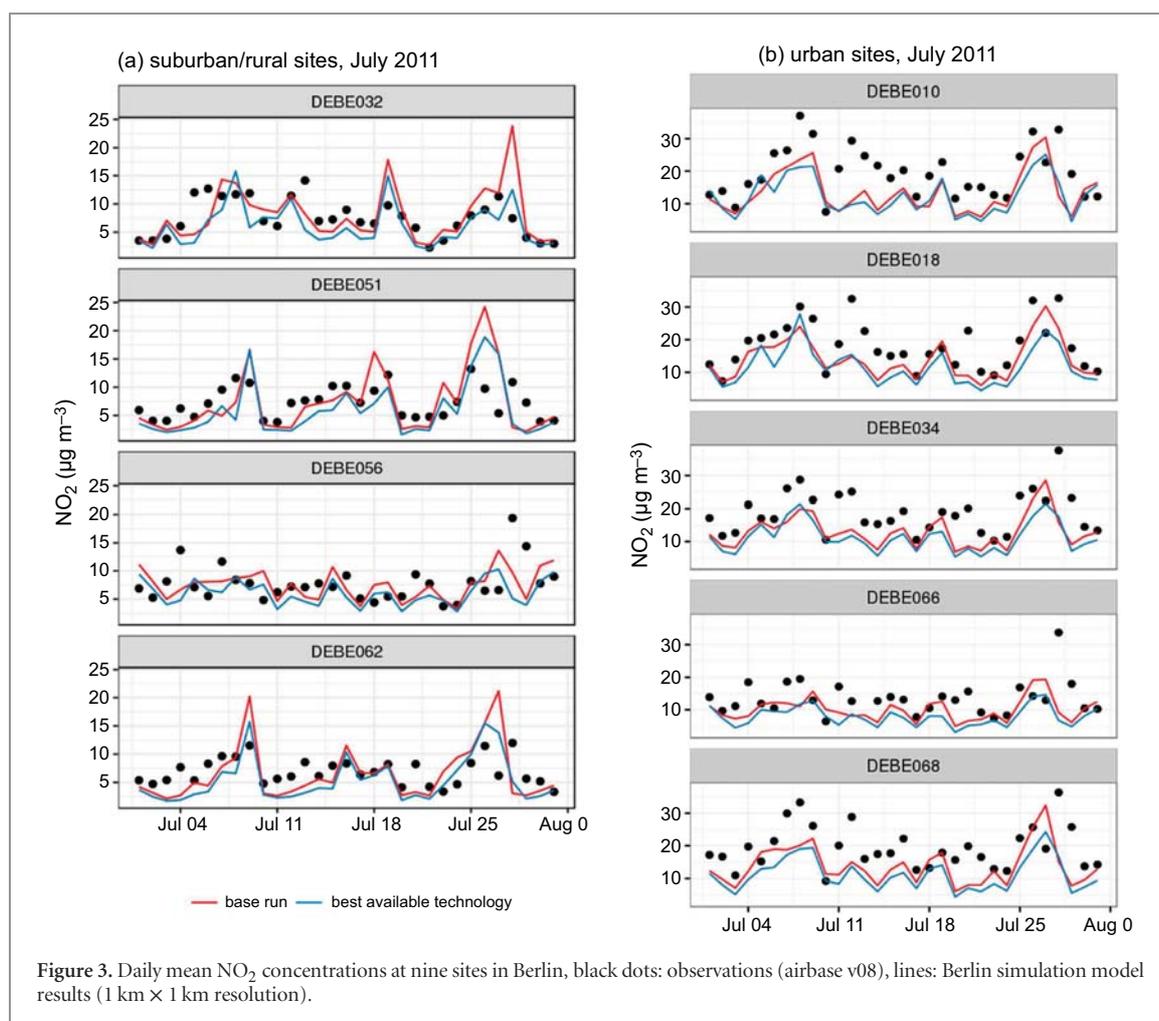


Figure 3. Daily mean  $\text{NO}_2$  concentrations at nine sites in Berlin, black dots: observations (airbase v08), lines: Berlin simulation model results ( $1 \text{ km} \times 1 \text{ km}$  resolution).

### 3.3. Comparison between model simulations and observation-based estimates

Overall, the  $\text{NO}_2$  reductions observed in both the European simulation and the Berlin simulation paint a consistent picture of the expected changes in  $\text{NO}_2$  concentrations were emission standards to be achieved under real-world driving conditions. The reductions estimated for Berlin city in the Berlin simulation ( $2.0 [1.1, 2.8] \mu\text{g m}^{-3}$ ) are also in line with the urban background increment from the observation-based calculations ( $1.6 \pm 0.53 \mu\text{g m}^{-3}$ ) (table 3). A comparison to the roadside reductions calculated from the monitoring data cannot be made to the models, as the resolutions are too coarse to capture street-level phenomena. However, the consistency of the results obtained with the different methods for different scales suggests that these estimates can give a good indication of the possible decrease of regional background, urban background and roadside  $\text{NO}_2$  concentrations if the Euro 5 emission standard or best-available technology case (reflecting the US EPA emission standard) were met by diesel LDVs in Europe.

### 3.4. Implications for ozone concentrations

Finally, the change in ozone concentrations in response to the reduction of  $\text{NO}_x$  emissions in the best-available technology scenario was also examined in the European

and Berlin simulations. The areas of Europe showing  $\text{NO}_x$ -sensitive and  $\text{NO}_x$ -saturated regimes are consistent for both the RADM2 and MOZART chemical mechanisms and with the findings of Mar *et al* 2016 (where a 30% increase in emissions was applied to  $\text{NO}_x$  for all sectors). Comparing the best-available technology and base case scenarios, most of the European domain is observed to be  $\text{NO}_x$ -sensitive, with a decrease in traffic  $\text{NO}_x$  in the best-available technology scenario leading to reductions in daily average  $\text{O}_3$  concentrations, with decreases in up to a few  $\mu\text{g m}^{-3}$  seen in southern Europe and the Mediterranean. When the maximum daily 8 hr mean ozone (MDA8, the metric corresponding to regulatory targets) is considered, a similar pattern is seen, with reductions in MDA8  $0.5\text{--}1.0 \mu\text{g m}^{-3}$  greater than reductions in daily average  $\text{O}_3$ . The UK, Benelux, northern France, and north-west Germany, as well as some more isolated urban centers, show  $\text{NO}_x$ -saturated behavior, in which the decreased  $\text{NO}_x$  emissions led to increases in daily mean  $\text{O}_3$  with a magnitude up to about  $2 \mu\text{g m}^{-3}$  ( $1 \mu\text{g m}^{-3}$  for MDA8), due to a reduced daytime sink of  $\text{O}_3$  via reaction with  $\text{NO}$ . For Berlin, the change in  $\text{O}_3$  concentrations was smaller than for other major European urban areas. The European model simulations showed  $\text{NO}_x$ -saturated behavior for the Berlin center area. However, the results from the high-resolution

model simulation for Berlin show a decrease in  $O_3$  with decreasing  $NO_x$  concentrations in most of the Berlin urban areas.  $NO_x$  titration, wherein the loss reaction of  $O_3$  with  $NO$  dominates in areas of high  $NO$  emissions, tends to reduce high  $O_3$  concentrations in urban centers (Sillman 1999). Because reductions in  $NO_x$  emissions can then lead to higher  $O_3$  concentrations, we see in our simulations that it will take greater reductions in  $NO_x$  emissions than in our best-available technology scenario (and/or reductions in NMVOC emissions) to significantly reduce urban ozone concentrations.

#### 4. Conclusions

The expected reductions in  $NO_2$  concentrations in a scenario where all diesel LDVs were assumed to meet the Euro 5 and US EPA Tier 2 Bin 5 (best-available technology) standards under real-world driving conditions was evaluated using monitoring data and in two chemical transport model simulations (which considered the best-available technology only). The reductions in  $NO_2$  for the Euro 5 standard were smaller than those for the best-available technology scenario (reductions in the annual average roadside increment:  $9.0 \pm 2.8 \mu\text{g m}^{-3}$  (national LDV) and  $17 \pm 5.2 \mu\text{g m}^{-3}$  (city LDV)), but still substantial. Greater reductions from on the observation-based calculations were estimated for July than for January. Furthermore, while the Euro 6 standard ( $0.08 \text{ g km}^{-1}$ ) is not as strict as the US EPA standard ( $0.043 \text{ g km}^{-1}$ ), substantial reductions would still be expected. Additional analysis (see SI) indicated that meeting the Euro 6 conformity factor for September 2017, would yield estimated reductions similar to all diesel LDVs meeting the Euro 5 standard. Model simulations yield reductions in urban background  $NO_2$  concentration similar to the observation-based approach, indicating robustness of the findings. These results indicate that stricter  $NO_x$  emissions standards—if implemented, appropriately controlled, and met—could have a significant impact on the  $NO_2$  concentrations in cities and especially at roadside locations. Owing to the  $NO_x$ -saturated environment in many of the urban areas of Europe, it will take even greater reductions in  $NO_x$  (and/or NMVOC reductions) to significantly reduce urban ozone concentrations. These results are however an idealized scenario in that they assume that the standards will be strictly enforced and that under real-world driving conditions the emission factors would still be met, and not allowed to exceed the standard as is currently the case. Based on the assumptions made in this study, the reductions calculated here are likely conservative estimates of what would be possible. Furthermore, any changes in technology that may influence the amount of  $NO_x$  emitted as primary  $NO_2$  were not considered. Further studies would be needed to investigate the role of potential technologies in  $NO_x$  reductions and the implications for primary  $NO_2$  emissions and subsequent chemistry effects.

It has become clear that emissions testing procedures are no longer adequate and that the promised improvements in air quality expected from the increasingly stringent standards are not being realized. This study and others, such as Brand (2016), Holland *et al* (2016), and Wang *et al* (2016), are an indication of the scope of the issue and potential benefits that could be realized were policies enacted that would effectively address diesel vehicle emissions standards. Furthermore, if policies addressed urban transport systems in a more comprehensive way, considering the bigger picture and the myriad linkages rather than individual elements in isolation, significant benefits could result, not only for air quality and/or climate change, but also for e.g. safety, noise pollution, levels of physical activity—even reducing social and health inequalities in the population (Rojas-Rueda and Turner 2016).

#### Acknowledgments

This work was hosted by IASS Potsdam, with financial support provided by the Federal Ministry of Education and Research of Germany (BMBF) and the Ministry for Science, Research and Culture of the State of Brandenburg (MWFK). The authors would like to thank the Berlin Senate Department for the Environment, Transport and Climate Protection and those in charge of the BLUME network for the availability of the monitoring data for the city of Berlin; Hugo Denier van der Gon and Jeroen Kuenen (TNO) for providing information pertaining to the TNO-MACCI3 inventory; colleagues at the IASS for discussions that helped shape the manuscript.

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