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On the impact of excess diesel NO_X emissions upon NO_2 pollution in a compact city

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Abstract

NO_X emissions from diesel light-duty-vehicles (LDV) largely exceed the Euro emission standards in real-world driving conditions. Recent studies have quantified their impact upon air quality and human health primarily based on air quality models at mesoscale and large-scale resolutions. Here, we show that these approaches can significantly underestimate the impact of diesel LDV excess NO_X emissions upon NO_2 pollution in cities, particularly in the more compact and heavily trafficked ones. We compare an air quality mesoscale model at both 4 and 1 km resolution with a street-scale model in Barcelona, a compact city where the EU annual NO_2 limits are repeatedly exceeded and a large share of passenger cars are diesel (65%). We compare consistently two emissions scenarios: a business-as-usual scenario where diesel LDV emit NO_X in excess, and a counterfactual standard limits scenario where emissions are compliant with the Euro emission standards. We first show that in contrast to the mesoscale model, the street scale model is able to largely represent the observed NO_2 concentration gradients between traffic and background stations in the city. In a second step, we find that the mesoscale model strongly underestimates the impact of diesel LDV excess NO_X emissions upon NO_2 pollution both in absolute terms (by 38%–48%) and relative terms (by 10%–35%). We argue that such underestimated impacts should be considered when assessing NO_X excess emissions by LDV in cities. Using the street scale model, we find that diesel LDV excess NO_X emissions are associated with about 20% of NO_2 levels in the city, contributing substantially to an increased number of citizens exposed to high NO_2 pollution in Barcelona.

1. Introduction

In 2017, 16 European Union (EU) countries reported NO_2 exceedances of the annual limit value enforced by the European Air Quality Directive (40 μg m^{-3}), with 86% of them occurring at urban traffic monitoring stations (EEA 2019). Exceedances are mostly due to emissions from vehicles, especially diesel light-duty-vehicles (LDV) (Ntziachristos *et al* 2016, Degraeuwe *et al* 2017) which represent about 40% of the European fleet, and whose emissions fail to meet the Euro emission standards under real-world driving conditions (Weiss *et al* 2011, Thompson *et al* 2014, Lewis *et al* 2015). Hereafter we will refer to diesel emissions above the Euro emission standards as excess diesel NO_X emissions.

Assessments of excess NO_X emissions impacts on air quality and health have relied on air quality models at mesoscale or large-scale resolutions and have mostly focused on $PM_{2.5}$ and O_3. For instance, Barrett *et al* (2015) tackled specifically the impact of Volkswagen diesel LDV equipped with emission defeat devices (Thompson *et al* 2014). Based on estimated $PM_{2.5}$ increases calculated with a chemistry-transport model at 50 km resolution, this study attributed to the so-called 'dieselgate' scandal ~59 premature deaths in the US. Anenberg *et al* (2017) attributed as much as ~38 000 premature deaths to excess diesel NO_X

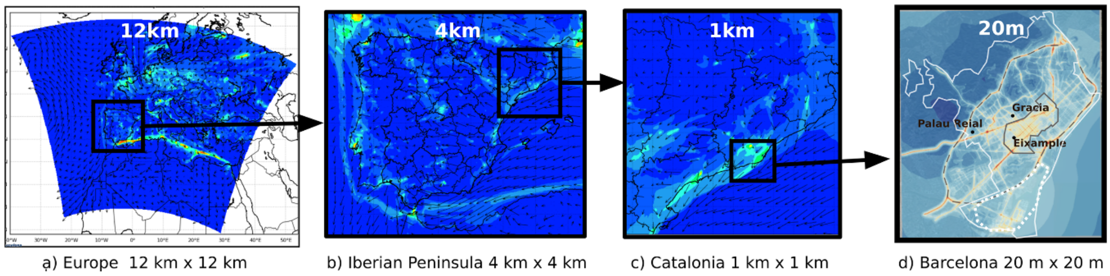

Figure 1. CALIOPE domains for EU, IP, and CAT (a–c), and the CALIOPE-Urban domain over Barcelona city (d). The panels depict NO_2 concentration at surface level. The black dots in (d) represent the location of the monitoring stations used in this work. The lines in (d) depict areas mentioned in this study: solid-white is Barcelona municipality, dashed-white is harbour, dashed-grey are ring roads and solid-grey is Eixample district in the city center.

emissions worldwide via increases in $PM_{2.5}$ and O_3 based on a model at 2° by 2.5° downscaled to 0.1° by 0.1° resolution. Von Schneidemesser *et al* (2017) estimated the impact of excess emissions upon NO_2 concentrations at 16 measurement sites in Berlin based on a model at 1 km resolution and observations, and found that NO_X traffic emissions would be reduced by 30%–55% if diesel LDV would comply with the regulatory standards. Using a chemical transport model at 0.1° by 0.1°, Jonson *et al* (2017) estimated ∼5000 premature deaths from $PM_{2.5}$ and O_3 in the adult population due to excess diesel NO_X emissions in EU28, Norway and Switzerland in 2013. Chossière *et al* (2018) showed that a large fraction of the health impacts from changes in $PM_{2.5}$ and O_3 in Europe are trans-boundary.

We identify two gaps in previous studies. First, they omit the effect of NO_2 upon health. It has been argued that the relationship between NO_2 and health is not as well-established as for $PM_{2.5}$ (Jonson *et al* 2017). Although the role of NO_2 as a surrogate of other measured or unmeasured pollutants cannot be completely ruled out, a variety of studies are consistent with long- (e.g. Faustini *et al* 2013, Sunyer *et al* 2015, Atkinson *et al* 2018, Achakulwisut *et al* 2019) and short-term (Samoli *et al* 2006, Chiusolo *et al* 2011) NO_2 effects upon health. Given that NO_2 is prominent in populated cities, omitting its effect may substantially bias the estimated health impact of excess NO_X emissions. Second, health effect assessments can be strongly affected by the methods for evaluating exposures; in this sense air quality model-based assessments using models at mesoscale or large-scale resolutions tend to underestimate pollutant levels and the associated health impacts (Li *et al* 2016, Korhonen *et al* 2019) as they do not capture intra-urban and near-roadway exposure gradients (Greco *et al* 2007, Karner *et al* 2010, Borge *et al* 2014). This is particularly important in compact cities for NO_2 and other species within the $PM_{2.5}$ fraction.

Our study focuses on the second gap. Mesoscale air quality models cannot represent NO_2 gradients and underestimate NO_2 levels in compact cities (Borge *et al* 2014, Duyzer *et al* 2015). We provide here a robust quantification of the impact of excess diesel NO_X emissions upon NO_2 levels in a compact city. To the best of our knowledge, this study presents the first such assessment using a street-scale air quality model. We focused on Barcelona city (Spain) (figure 1(d)), a densely populated and trafficked urban area (about 5500 vehicles km^{-2}) where the annual NO_2 limit value set up by the European Air Quality Directive (40 $\mu g\ m^{-3}$) has been exceeded uninterruptedly since year 2000, the majority of passenger cars are diesel (65%) (Barcelona City Council 2017a) and road transport is the main contributor to the chronic NO_2 exceedances. According to the local Public Health Agency about 70% of its 1.6 million citizens were exposed to NO_2 concentration levels above the annual air quality limit value in 2017. These NO_2 exposure levels were associated with about 929 premature deaths in Barcelona city in 2017 (ASPB 2018).

Our underlying hypothesis is that prior modeling studies at mesoscale and large-scale resolutions have substantially underestimated the impact upon NO_2 in cities, particularly in the more compact and heavily trafficked ones. We compare modeled NO_2 concentrations calculated consistently at 4 km, 1 km and street-level resolutions using an air quality multiscale modeling system fed by a state-of-the-art bottom-up emission model for two scenarios: *business-as-usual (BAU)* and *standard limits (SL)* diesel LDV NO_X emissions. The *BAU scenario* represents real-world driving conditions and the *SL scenario* represents diesel LDV emissions complying with Euro emission standards.

2. Methods

2.1. Domain and period of study

Barcelona city covers an area of 101 km^2 (figure 1(d)). The simulation period for the case study is from the 9th to the 25th of November 2017. This period is selected because it is representative of late autumn air quality levels in Barcelona, including both an episode of high NO_2 concentrations and days that are representative of the observed annual mean daily cycle in the city. During this period the NO_2 hourly limit set up by the European Air Quality Directive (200 $\mu g\ m^{-3}$)

was exceeded two times at the Gràcia-Sant Gervasi (i.e. Gràcia) traffic monitoring station in Barcelona. The high concentration levels recorded were due to a strong temperature inversion causing stagnant air.

2.2. Multiscale air quality modeling: from mesoscale to street-level

We used the CALIOPE-Urban street-scale modeling system which consists of the CALIOPE air quality mesoscale modeling system (Baldasano *et al* 2011) coupled with the near road dispersion model R-LINE (Snyder *et al* 2013) adapted to street canyons (Benavides *et al* 2019). CALIOPE consists of the Weather Research and Forecasting model version 3 (WRF; Skamarock and Klemp 2008), combined with the HERMESv3 multi-scale atmospheric emission modeling framework (Guevara *et al* 2019, Guevara *et al* 2020), the Community Multiscale Air Quality Modeling System version 5.0.2 (CMAQ; Byun and Schere 2006) and the mineral Dust REgional Atmospheric Model (BSC-DREAM8b; Basart *et al* 2012). CALIOPE was run over a domain covering Europe at a 12 km by 12 km horizontal resolution (figure 1(a)), Iberian Peninsula at 4 km by 4 km (figure 1(b)), hereafter referred to as IP-4 km, and the Catalonian domain, including Barcelona, at 1 km by 1 km, hereafter referred to as CAT-1 km (figure 1(c)). CALIOPE results have been evaluated in detail elsewhere (e.g. Pay *et al* 2014). For the mesoscale model, simulations were initialised with the ECMWF reanalyses (ERA-Interim), boundary conditions for chemistry come from the CAMS reanalysis of atmospheric composition (Inness *et al* 2019) and pollutant emissions are obtained from HERMESv3. For the European parent domain (EU-12 km) HERMESv3 was run using the TNO-MACIII inventory (Kuenen *et al* 2014) for European countries and the HTAPv2.2 inventory (Janssens-Maenhout *et al* 2015) for countries outside Europe. For the nested mesoscale domains covering the Iberian Peninsula (IP-4 km) and Catalonia (CAT-1 km) the same approach was applied except for Spain, where high resolution detailed emissions were estimated using the bottom-up module of HERMESv3 (Guevara *et al* 2020). The coupling with R-LINE estimates local traffic dispersion driven by channeled street winds and vertical mixing with background air taking into account atmospheric stability and street morphology (e.g. aspect ratio). R-LINE applies the Generic Reaction Set (GRS) to resolve simple NO to NO_2 chemistry (Valencia *et al* 2018). Further information regarding CALIOPE-Urban's methodology and its evaluation using NO_2 observed concentrations can be found in Benavides *et al* (2019). To obtain high-resolution concentration maps for the entire Barcelona city, hereafter referred to as BCN-20 m, we set the domain over Barcelona city as the minimum rectangle where Barcelona municipality is contained and extended it by 250 m buffers that include the highways surrounding the city (figure 1(d)). This domain is covered by a regular receptor grid of 20 m resolution.

2.2.1. Road traffic emissions

Road traffic emissions were estimated using the bottom-up traffic emission module from HERMESv3 (Guevara *et al* 2020) for the IP-4 km and CAT-1 km mesoscale domains as well as for the BCN-20 m urban domain. This allows to maintain consistency between the different scales and therefore obtain comparable results. Estimated hourly road link-level vehicle emissions were conservatively mapped onto the gridded mesoscale domains (IP-4 km and CAT-1 km) and adapted to the requirements of BCN-20 m. HERMESv3 estimates hot and cold exhaust road transport emissions combining the tier 3 method described in the 2016 EMEP/EEA air pollutant emission inventory guidebook (EMEP/EEA 2016), which is fully incorporated in the COPERT 5 software, with a digitised traffic network. This traffic network contains specific information by road link for daily average traffic, mean speed circulation, temporal profiles and vehicle fleet composition profiles. For the city of Barcelona, this information was obtained from multiple sources, including the local automatic traffic counting network (Barcelona City Council, personal communication), fleet composition from a remote sensing device campaign in 2017 (Barcelona City Council 2017a) and TomTom historical average speed profiles (TomTom 2019) based on GPS data gathered from circulating vehicles between 2015 and 2016. In HERMESv3, we further extended the degradation factors for diesel vehicles taking into account mileage reported by COPERT 5 for gasoline vehicles. For EURO 2 and EURO 3, this extension considers a deterioration of tailpipe NO_X emissions of 22% and 10% respectively, as suggested by Chen and Borken-kleefeld (2016). HERMESv3 disaggregates the calculated NO_X emissions into NO and NO_2 using vehicle-dependent speciation factors that are extracted from EMEP/EEA (2016) (chap. 1.A.3.b.i–iv, table 3.87) and the investigations of Rappenglueck *et al* (2013) and Carslaw *et al* (2016). HERMESv3 also estimates the emissions released from other anthropogenic sources, which are considered in the mesoscale model and their contributions are taken into account in the street-scale model as background concentrations.

2.3. Emission scenarios

We defined two scenarios to quantify the effect of excess diesel LDV NO_X emissions in Barcelona City. The *BAU scenario* is our best estimate of NO_X emissions under real-world driving conditions for Barcelona city in 2017. It incorporates emissions representing real-world driving conditions, which is supported by (a) the use of COPERT 5 real-world adjusted NO_X emission factors for EURO 5 and 6 diesel vehicles, (b) the inclusion of a mileage correction factor for diesel vehicles and (c)

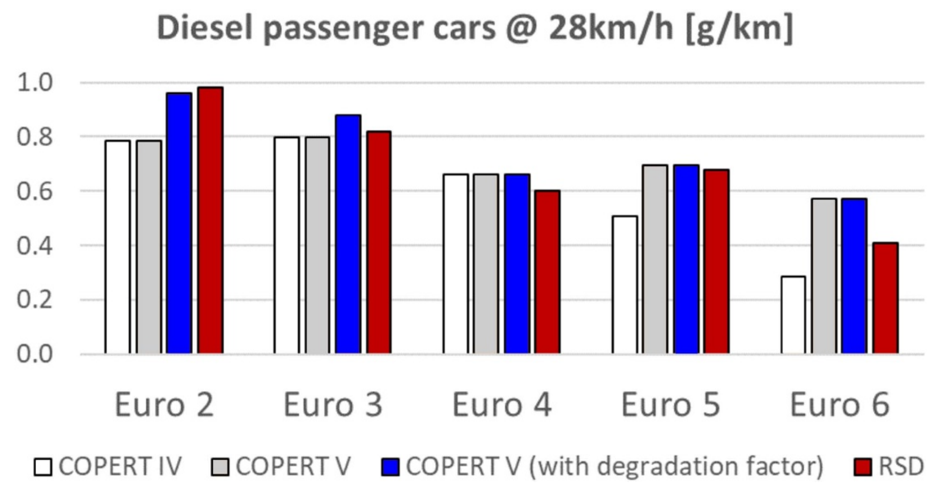

Figure 2. Comparison between diesel passenger cars emission factors ($g\,km^{-1}$) derived from a RSD campaign performed in Barcelona (Barcelona City Council 2017a) and emission factors reported by different versions of the COPERT emission factor model. Emission factors are estimated at an average speed of 28 $km\,h^{-1}$. RSD in red, COPERT 5 using degradation factors in blue, COPERT 5 in grey and COPERT 4 in white.

Table 1. HERMESv3 vehicle fleet for Barcelona city (%). The urban area represents the region within the surrounding ring roads and the ring roads represent the highways surrounding the city. Only the vehicle categories with a percentage higher than 1 are shown.

Vehicle type	Fuel	Euro category	Urban area (%)	Ring roads (%)
Passenger cars	Diesel	EURO 2	2.1	2.2
		EURO 3	8.2	8.5
		EURO 4	13.0	13.2
		EURO 5	13.7	14.0
		EURO 6	9.5	9.7
Light commercial vehicles	Diesel	EURO 2	0.6	0.7
		EURO 3	2.7	2.7
		EURO 4	4.0	4.1
		EURO 5	5.6	5.7
		EURO 6	0.8	0.8
Passenger cars	Petrol	All	19.5	20.0
Light commercial vehicles	Petrol	All	0.3	0.3
Motorbikes	Petrol	All	11	6.5
Mopeds	Petrol	All	2.6	0
Trucks	Diesel	All	1	8
Buses	Diesel	All	2.4	1.2
Buses	Natural gas	All	1.0	0.3
Buses	Hybrid	All	0.5	0.3

the good agreement observed between the emission factors estimated by HERMESv3 and measured during the Remote Sensing Device (RSD) campaign performed in Barcelona (Barcelona City Council 2017a) (figure 2). As shown in figure 2, the combination of COPERT 5 emission factors with the degradation factors is in close agreement with the RSD-derived NO_X emission factors for the prevalent EURO categories. COPERT-derived emission factors were calculated considering an average speed of 28 $km\,h^{-1}$, the same speed at which RSD measurements were performed. This speed value is within the range of the common vehicle's average speed within the urban area of Barcelona (Barcelona City Council 2017b). Table 1 shows the two vehicle fleet distributions used in HERMESv3 for Barcelona city. The profiles divide the road network within the city into two main zones, ring roads and urban streets within these ring roads, to account for observed differences in the composition of the circulating vehicles. In Eixample district, the percentage of mopeds and motorbikes is increased up to 25% to better represent the observed fleet (Barcelona City Council 2017a) while the ratio among other vehicle types is kept the same as in the urban streets. Diesel LDV predominate in both the urban fleet (60.2%) and in the ring roads (61.6%) vehicle fleet distributions. The main difference is that diesel trucks are more frequent in the ring roads (8%) than in the inner city (1%), while mopeds are not allowed to circulate in the ring roads (0% versus 2.6%).

The counterfactual scenario is the *SL scenario*, which assumes that diesel LDV emissions comply with the Euro emission standards (European Commission 2007). It therefore considers that emission

Table 2. Diesel LDV real-world emission factors from HERMESv3 estimated at 28 km h^{-1}, Euro emission standards and standard limit scaling factors for NO_X adopted from Anenberg *et al* (2017).

Vehicle type	Fuel	Euro category	Real-world NO_X (g km^{-1})	NO_X emission limit (g km^{-1})	Standard limit scaling factor
LDV	Diesel	EURO 3	0.88	0.50	0.60
		EURO 4	0.66	0.25	0.31
		EURO 5	0.69	0.18	0.23
		EURO 6	0.57	0.08	0.17

Table 3. NO_2 model evaluation statistics calculated at Palau Reial, Eixample, and Gràcia sites (figure 1(d)) during the period of study (9–25th November 2017). Standard statistics are described in Chang and Hanna (2004) and are computed with model hourly results of CALIOPE-Urban, CALIOPE-1 km and CALIOPE-4 km systems. *FAC2* refers to the fraction of model results within a factor of 2 of observations, *MB* is the mean bias, *RMSE* is the root-mean-square error and *r* is the correlation coefficient. Bold numbers represent model results with better performance for each statistic and site.

Site	Method	FAC2	MB	RMSE	*r*
1. Palau Reial	CALIOPE-Urban	0.62	**−5.17**	28.91	0.51
	CALIOPE-1 km	0.54	−18.66	34.11	0.49
	CALIOPE-4 km	**0.65**	−5.53	**27.78**	**0.58**
2. Eixample	CALIOPE-Urban	**0.86**	**4.16**	**31.27**	0.57
	CALIOPE-1 km	0.76	−19.36	33.44	**0.65**
	CALIOPE-4 km	0.75	−21.26	35.97	0.57
3. Gràcia	CALIOPE-Urban	**0.81**	**−16.21**	**36.04**	**0.58**
	CALIOPE-1 km	0.57	−33.04	46.57	0.57
	CALIOPE-4 km	0.57	−34.01	48.81	0.48

limits are not exceeded for light passenger and commercial vehicles under real-world driving conditions following the European Commission regulation 2016/646. To create this scenario we transformed the real-world traffic NO_X emissions using the scaling factors proposed by Anenberg *et al* (2017) for diesel LDV from EURO 3 to EURO 6. These scaling factors are reported by vehicle type and EURO category and are based on an extensive review of previous works measuring real-world vehicle's emissions. The scaling factors are used to estimate standard limit emissions by correcting the real-world NO_X emission factors considered in HERMESv3 for each vehicle type and EURO category. In the present study, emissions from older diesel LDV (i.e. pre-EURO1, EURO1 and EURO2) are not considered in the scaling process due to their low share reported in the vehicle composition profiles (about 3%). We focus on diesel LDV because they are the dominant contributors to excess NO_X health impacts in Europe (Anenberg *et al* 2017) and the dominant vehicle type in Barcelona's circulating fleet (60.2% in the urban area and 61.6% in the ring roads).

3. Results

3.1. Model evaluation

We first provide an evaluation of the *BAU scenario* using hourly NO_2 concentrations reported by the official monitoring network in Catalonia (XVPCA) for the year 2017 in the only two traffic stations available in Barcelona (i.e. Gràcia and Eixample) and in one of the urban background stations (Palau Reial), which is considered to be representative of the models behaviour at this site type, as shown in figure 1(d). The model performance at the other urban background sites can be found in the supplementary material (available online at stacks.iop.org/ERL/16/024024/mmedia) and for a more complete evaluation near traffic in Barcelona we refer to Benavides *et al* (2019). Table 3 shows standard statistics computed using the modeled and measured concentrations and figure 3 depicts the NO_2 average daily cycle. All the simulations tend to slightly underestimate NO_2 and behave similarly well in the urban background station. Yet, IP-4 km and CAT-1 km are strongly biased in the traffic stations. As expected BCN-20 m is able to better reproduce the observed values at the traffic stations. At the Gràcia site, it reduces the underestimation by ∼50% (∼−16 vs ∼−33 $\mu g\ m^{-3}$) and at the Eixample site, by a factor ∼5 (∼4 vs ∼−20 $\mu g\ m^{-3}$). Between the traffic sites, we relate the distinct behaviour at each site to the relative influence of local traffic in each site, to the influence of the main trafficked areas and to the specific micro-meteorological patterns.

3.2. Impact of excess diesel LDV emissions upon total NO_X emissions

Table 4 shows the annual total NO, NO_2 and NO_X traffic emissions estimated for Barcelona city for the *BAU* and the *SL scenarios*. NO_X traffic emissions decrease by ∼27% on average when diesel LDV are assumed to comply with EU standard limits. The decrease in primary NO_2 emissions is much stronger (∼49%) than in NO emissions (∼22%). This is

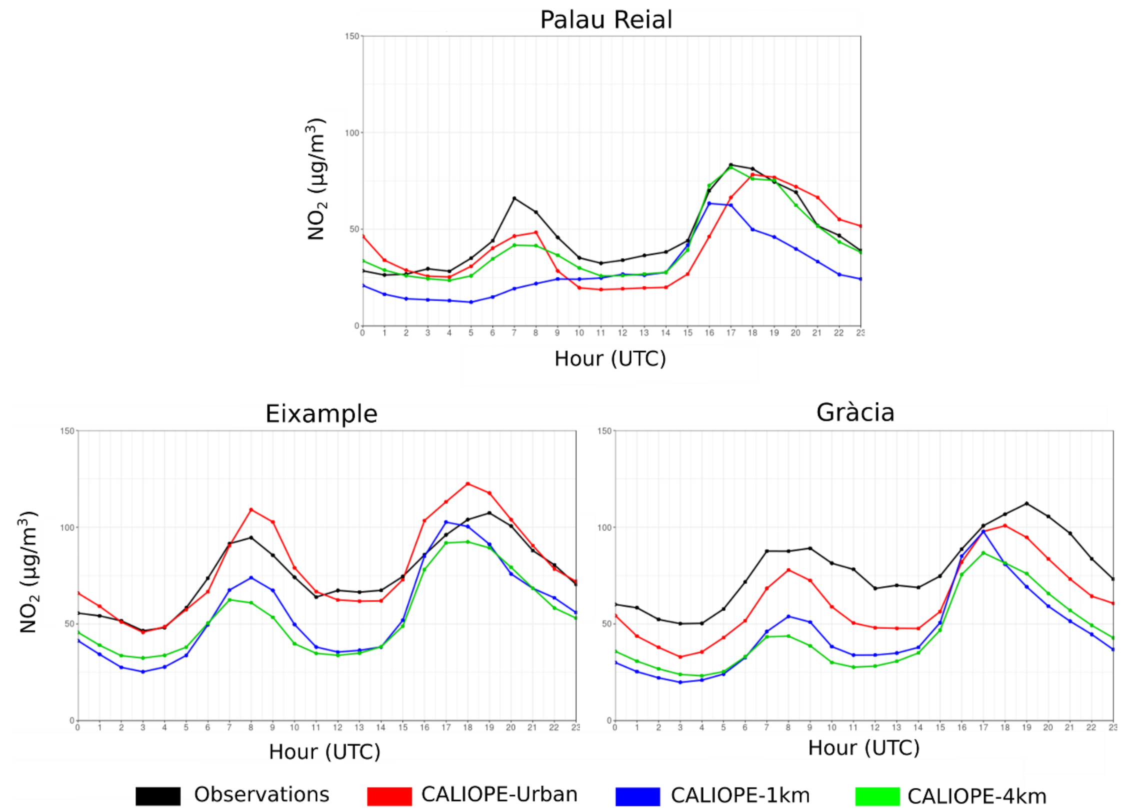

Figure 3. NO_2 average daily cycle at Palau Reial (background), Eixample and Gràcia sites (traffic) (figure 1(d)) during the period of study (9–25th November 2017). Observations are represented in black lines, CALIOPE-Urban in red, CALIOPE-1 km in blue, and CALIOPE-4 km in green lines.

Table 4. Estimated annual NO, NO_2, and NO_X traffic emissions (Tg) for the year 2017 in Barcelona for all the vehicle types at road link-level for the *BAU* and *SL scenarios* and the relative difference in traffic emissions (%) that is calculated by (*BAU*−*SL*)/*BAU*. Reductions are expressed as negative values.

Configuration name	NO	NO_2	NO_X
BAU	5348	1331	6679
SL	4163	681	4844
Reduction in traffic emissions (%)	−22%	−49%	−27%

explained by the larger contribution of NO_2 emissions in diesel LDV compared to other vehicles. For instance, Carslaw *et al* (2016) found the NO_2/NO_X ratio for diesel cars EURO 4, 5 and 6 to range from 0.25 to 0.34 whereas for EURO-equivalent petrol cars it ranged from 0.05 to 0.12.

Table 5 shows the intra-urban variability of NO_X emissions per km^2 at road-link level for both scenarios and the relative difference of these scenarios. The impact upon NO_2 emissions, with relative reductions ranging from −11.4% to −58.1%, is stronger than upon NO (−2.2% to −36.4%).

Figure 4 shows the spatial distribution of NO_X traffic emissions in Barcelona city at road-link level for both scenarios and the relative difference thereof. The relative reductions in NO_X emissions (figure 4(i) range only from 2.2% to 10% in the harbour area (figure 1(d) because the fleet is dominated by heavy duty vehicles, from −20% to −30 % in the very built-up Eixample city center district, and from −30% to −40 % in most of the other districts in the city and the ring roads (figure 1(d)). The lower difference in the centre district may be caused by the greater share of mopeds and motorbikes in that district (25% according to Barcelona City Council 2017a). The impact on NO_2 emissions (figure 4(f)), with a relative difference ranging from 40% to 60%, is greater than it is on NO (figure 4(c)), ranging from 20% to 40%.

3.3. Impact of excess diesel LDV emissions upon NO_2 concentration

Figure 5 shows the average NO_2 concentration maps for BCN-20 m, IP-4 km and CAT-1 km. The two mesoscale outputs were interpolated using bilinear interpolation to the 20 m resolution grid for comparison purposes. For the three cases, we show the *BAU* and the *SL scenarios* along with the absolute and relative differences thereof. The spatial detail of BCN-20 m enables characterising the impact of excess diesel LDV NO_X emissions at street-level; the median value of the average *BAU* concentrations across the 20 m receptors is 58.9 μg m^{-3} (interquartile ranges (IQR) from 46.0 to 75.8 μg m^{-3}); the median value of the absolute difference between average BAU and SL concentrations is −10.5 μg m^{-3} (IQR from −6.7 to −14.7 μg m^{-3}); and the median value of the relative difference

Table 5. Estimated annual NO, NO_2, and NO_X traffic emissions (Tg) per km^2 for the year 2017 in Barcelona at road link-level for all the vehicle types for the *BAU* and *SL scenarios* and the relative difference in traffic emissions (%).

	NO			NO_2			NO_X		
Statistic	*BAU*	*SL*	Diff. (%)	*BAU*	*SL*	Diff. (%)	*BAU*	*SL*	Diff. (%)
Average	35.9	27.9	−22.9	8.9	4.6	−47.7	44.8	32.5	−27.9
Median	29.1	22.5	−25.7	7.0	3.8	−51.4	36.9	26.5	−31.2
Max	124.8	116.2	−36.4	32.8	15.7	−58.1	156.0	128.2	−41.9
Min	0.0	0.0	−2.2	0.0	0.0	−11.4	0.0	0.0	−3.1

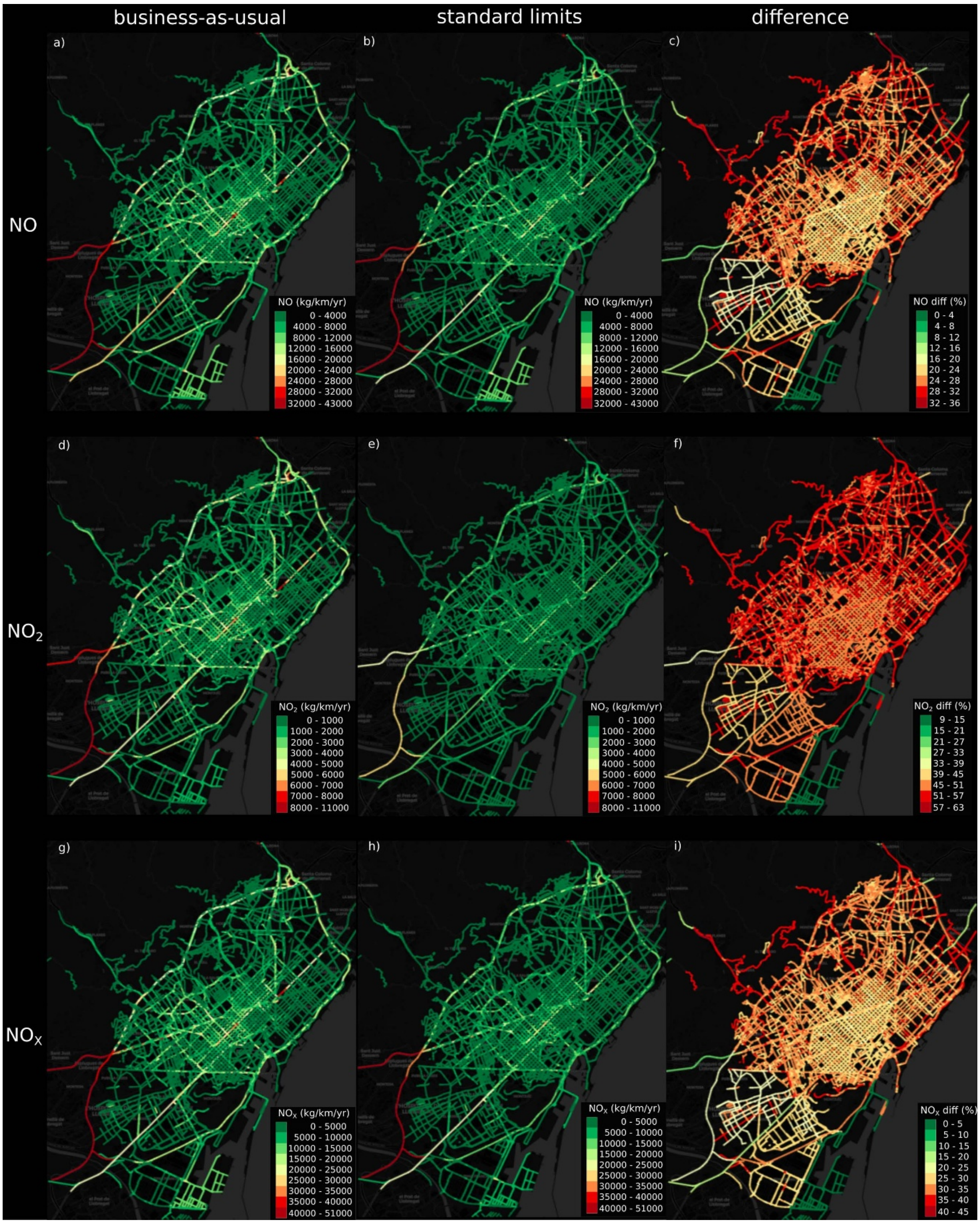

Figure 4. Estimated annual NO (a–c), NO_2 (d–f) and total NO_X (g–i) traffic emissions (kg km^{-1}) for the year 2017 in Barcelona city for the entire vehicle fleet for both *BAU* and *SL scenarios* and the relative percentage difference computed using the *BAU* as reference value.

is −20% (IQR from −15% to −21%). These differences are consistent with the daily results as seen in section S2 figure S2 in the supplementary material. The largest differences are generally found in the areas with highest NO_X traffic emissions (figure 4). In fact, the relative differences in NO_2 levels between

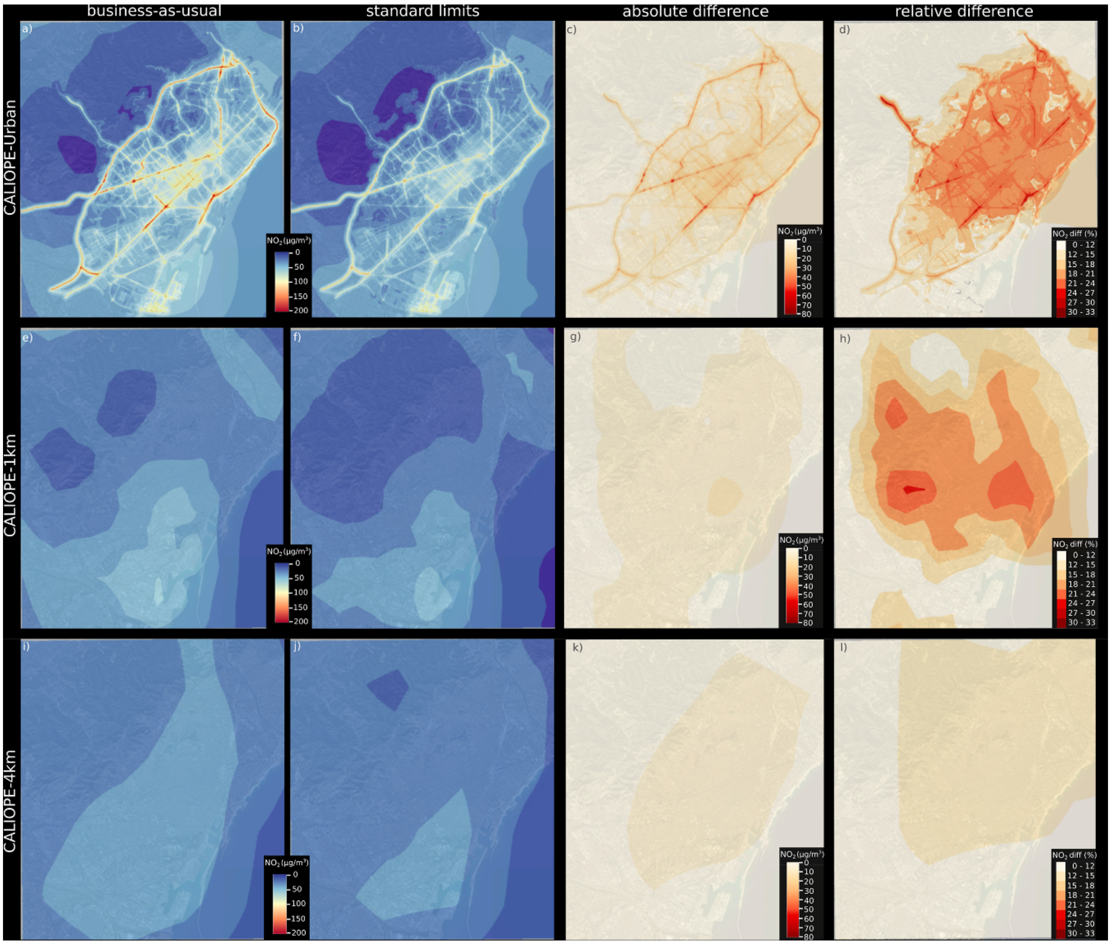

Figure 5. Average NO_2 concentrations during the period of study (9–25th November 2017) for *BAU scenario*, *SL scenario* and their absolute and relative differences for CALIOPE-Urban (a)–(d), CALIOPE-1 km (e)–(h) and CALIOPE-4 km (i)–(l).

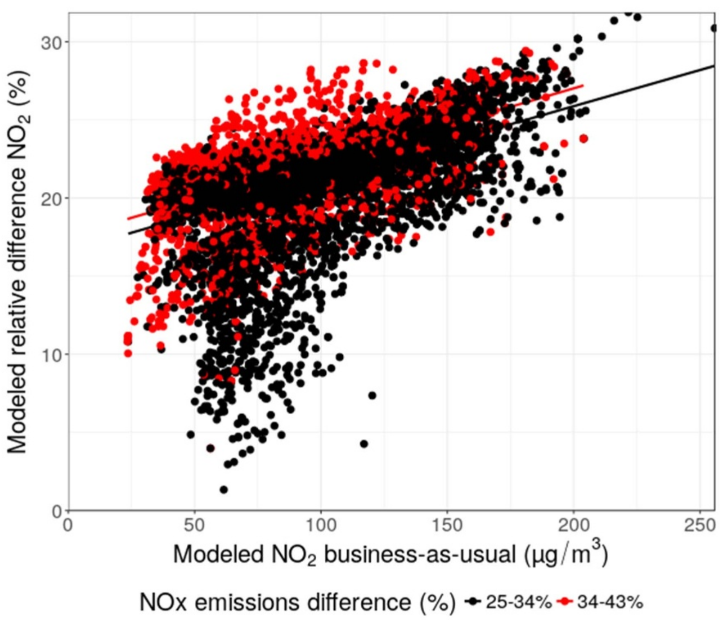

Figure 6. Scatter plot showing the relation of NO_2 mean concentrations for the *BAU scenario* and the relative difference between scenarios during the period of study (9–25th November 2017) using CALIOPE-Urban results. The dots represent values at 12 389 receptors within 10 m radial distance from road-link mid-points for road-links with NO_X emissions difference between scenarios above 25% (a 93% of links) in 2017 in Barcelona municipality. Colours represent relative difference in NO_X emissions.

scenarios broadly scale with the average NO_2 concentration (figure 6), a feature that is not reproduced in IP-4 km and CAT-1 km (not shown). Diesel LDV NO_X emissions strongly contribute to NO_2 concentrations in the ring roads and some major streets that act as the main entrance routes towards the city center. Additionally, some districts such as the Eixample are more affected to excess diesel emissions than others mainly due to the combination of the aspect ratio of the streets and the traffic intensity. In contrast, in areas where NO_2 concentrations are predominantly affected by other sources, the difference between scenarios is comparably low. For instance, in the area surrounding the harbour, the impact of LDV is lower than in other parts of the city due to the high share of diesel high-duty-vehicles operating there (around 45% of the total circulating vehicles). This area is represented by the points showing relative differences below 10% in figure 6.

IP-4 km and CAT-1 km show very different spatial patterns; the steep spatial gradients appearing in BCN-20 m are smoothed out and concentrations in the *BAU scenario* only reach a median value of 41.7 μg m^{-3} (IQR from 39.5 to 44.7 μg m^{-3}) and 38.6 μg m^{-3} (IQR from 35.0 to 48.4 μg m^{-3}), respectively, a 29% (IQR from 14% to 41%) and 34% (IQR from 24% to 36%) less compared to BCN-20 m.

The absolute differences between scenarios are -5.5 μg m^{-3} (IQR from -5.0 to -6.0 μg m^{-3}) for IP-4 km and -6.5 μg m^{-3} (IQR from -5.6 to -8.1 μg m^{-3}) for CAT-1 km, which is 38%–48% less than in BCN-20 m. The median relative differences are -13% (IQR from -13% to -14%) for IP-4 km and -18% (IQR from -15 to -19%) for CAT-1 km, which is 10%–35% less than in BCN-20 m.

4. Discussion and conclusions

To quantify the impact of excess NO_X diesel LDV emissions upon NO_2 concentrations over Barcelona city we compared simulations with two different emission scenarios: a *business-as-usual (BAU) scenario* representing NO_X diesel LDV emissions under real-world driving conditions and a counterfactual *standard limits (SL) scenario*, which represents NO_X diesel LDV emissions compliant with the Euro emission standards.

We first showed that, in contrast to the mesoscale model, the street-scale model is able to reproduce the NO_2 concentration gradients observed in the city between open areas and trafficked zones. We found a decrease on the order of -30% in total NO_X and -50% in total primary NO_2 emissions, consistent with other studies (e.g. Von Schneidemesser *et al* 2017). The differences are not homogeneous across the city; we estimated higher NO_X emissions in the very built-up city center and near the ring roads. Overall this translated into a reduction of median NO_2 concentrations and interquartile ranges in Barcelona municipality of -10.5 μg m^{-3} (-6.7 to -14.7 μg m^{-3}), -6.5 μg m^{-3} (-5.6 to -8.1 μg m^{-3}), and -5.5 μg m^{-3} (-5.0 to -6.0 μg m^{-3}) in absolute terms and -20% (from -15% to -21%), -18% (from -15% to -19%), -13% (from -13% to -14%) in relative terms using BCN-20 m, CAT-1 km and IP-4 km, respectively. In other words, the mesoscale simulations underestimated the absolute reductions in NO_2 concentrations by ~38% and ~48% and the relative reductions by 10% and 35% compared to the street-scale model when NO_X diesel LDV emissions were assumed to comply with Euro emission standards.

Other prior studies (e.g. Anenberg *et al* 2017, Jonson *et al* 2017) have used low resolution models to characterise the impacts upon $PM_{2.5}$ and O_3. While our study focuses only on NO_2, it is likely that $PM_{2.5}$ or at least a fraction of it is largely underestimated in compact cities at mesoscale resolutions. Stronger underestimations on both the absolute and relative reductions are to be expected as model resolution decreases. Thus, street models are strongly recommended to avoid underestimations in impact assessments of alternative emission scenarios in compact cities.

The largest differences in NO_2 between scenarios were found in the areas with the highest NO_X traffic emissions. Using observations, Von Schneidemesser *et al* (2017) estimated a potential NO_2 reduction in traffic sites across Berlin ranging from -9 to -23 μg m^{-3} if diesel LDV would comply with the EURO standards. Similarly, we found an average reduction ranging from -13 to -18 μg m^{-3} at traffic sites during the period of study using the street-scale model. Despite the large improvements, these sites would still not meet the EU and WHO annual limit values assuming that the relative differences between scenarios during our study are maintained during the whole year (40.6 μg m^{-3} in Gràcia and 45.6 μg m^{-3} in Eixample sites).

Our results imply an increase in the number of citizens exposed to unhealthy NO_2 levels when comparing the BAU with the SL scenario. We estimate that 90.2% of citizens would be exposed to NO_2 levels above 40 μg m^{-3} under the BAU scenario, a percentage that would decrease to 76.6% within the SL scenario during the study period (see section S3 and figure S3 in supplementary material). Assuming that the relative difference is maintained throughout the year 2017, the percentage of Barcelona citizens exposed to higher concentrations than the annual limit would have been reduced from 70% (ASPB 2018) to 59%. Future studies should further investigate the associated health implications in a city where exposure to NO_2 was associated with about 929 premature deaths in 2017 (ASPB 2018).

Based on our findings, diesel LDV should actually meet the EURO standards under real-world driving conditions to substantially reduce NO_2 concentration levels in cities with a high proportion of diesel LDV and chronic NO_2 exceedances. Moving towards cleaner LDV in these cities is likely not enough to meet the EU and WHO annual limit values for NO_2 in trafficked streets and other policies targeting the overall reduction of LDV circulating should be encouraged.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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