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Low emission zones reduced PM_{10} but not NO_2 concentrations in Berlin and Munich, Germany

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ABSTRACT

Low emission zones (LEZs) aiming at improving the air quality in urban areas have been implemented in many European cities. However, studies are limited in evaluating the effects of LEZ, and most of which used simple methods. In this study, a general additive mixed model was utilized to account for confounders in the atmosphere and validate the effects of LEZ on PM_{10} and NO_2 concentrations in two German cities. In addition, the effects of LEZ on elemental carbon (EC) and total carbon (TC) in Berlin were also evaluated. The LEZ effects were estimated after taking into account air pollutant concentrations at a reference site located in the regional background, and adjusting for hour of the week, public holidays, season, and wind direction. The LEZ in Berlin, and the LEZ in combination with the heavy-duty vehicle (HDV) transit ban in Munich significantly reduced the PM_{10} concentrations, at both traffic sites (TS) and urban background sites (UB). The effects were greater in LEZ stage 3 than in LEZ stages 2 and 1. Moreover, compared with PM_{10}, LEZ was more efficient in reducing EC, a component that is considered more toxic than PM_{10} mass. In contrast, the LEZ had no consistent effect on NO_2 levels: no effects were observed in Berlin; in Munich, the combination of the LEZ and the HDV transit ban reduced NO_2 at UB site in LEZ stage 1, but without further reductions in subsequent stages of the LEZ. Overall, our study indicated that LEZs, which target the major primary air pollution source in the highly populated city center could be an effective way to improve urban air quality such as PM mass concentration and EC level.

1. Introduction

In many parts of the world, air pollution is a major public health risk (Beelen et al., 2014). Major pollutants including particulate matter (PM) and nitrogen dioxide (NO_2) cause adverse health effects (Anenberg et al., 2018; Beelen et al., 2014; Brook et al., 2010; Hoek et al., 2013; Rückerl et al., 2011). In order to protect human health, the European Union (EU) established limit values for several pollutants, including PM_{10}, $PM_{2.5}$ (particulate matter with aerodynamic diameter smaller than 10 μm and 2.5 μm, respectively) and NO_2 (Council of the European Union, 2008). Member States were obliged to implement measures to reduce the regulated pollutants when they exceed the limit values. Despite the continuous improvement of the air quality, the European limit values for PM_{10}, $PM_{2.5}$ and NO_2 are still exceeded in many

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European countries (European Environment Agency, 2018). Moreover, for PM, there is no threshold level, below which no adverse health effects would occur (WHO, 2006, 2013a). Results from a large multicenter European Study, the European Study of Cohorts for Air Pollution Effects (ESCAPE) also showed significant positive associations between PM and adverse health effects even at levels far below the current EU limit values (Beelen et al., 2014; Raaschou-Nielsen et al., 2013). The scientific community also argued that the current European limit values for $PM_{2.5}$ and PM_{10} are too high and provide no incentive for the implementation of those national and local strategies needed to achieve more ambitious goals. It has been advocated that the European Commission should adopt the lower WHO Air Quality Guideline values as limit values for PM in the near future (Brunekreef et al., 2015).

To improve the air quality, air quality action plans such as promotion of public transportation usage, ring road utilization, traffic flow improvements, speed limit reduction, and the low emission zones (LEZ) were implemented. LEZs are areas where access for road vehicles is restricted, usually based on their emission classes. In Europe, there are more than 200 LEZs in operation (Holman et al., 2015; Silva et al., 2014). As in urban areas, traffic is one important source of PM and the major source of NO_2 (Belis et al., 2013; Degraeuwe et al., 2017; Viana et al., 2008), the implementation of LEZ could be an effective measure to reduce traffic related pollution and to improve urban air quality (Sadler, 2011).

LEZ regulations vary between different cities in size, types of vehicles regulated and the ways of control and enforcement. In Germany, LEZs are operated in three different stages, LEZ 1, LEZ 2, and LEZ 3. The individual municipality decides on the implementation of a LEZ, on its area and the stage of the LEZ. In general, LEZ 3 has the most stringent requirements allowing gasoline vehicles with Euro 1 emission standard or Diesel vehicles with Euro 4, Euro 3 with diesel particle filters (DFP) or higher emission standards to enter.

The effects of LEZs have been evaluated in the decision-making stage by emission models or in combination with dispersion models (Emplan, 2010; LfU, 2010; Watkiss et al., 2003). Some studies have been carried out to assess the effects after the LEZ implementation using monitoring data (Boogaard et al., 2012; Cesaroni et al., 2012; Cyrys et al., 2014; Ellison et al., 2013; Fensterer et al., 2014; Jiang et al., 2017; Löschau et al., 2015; Malina and Scheffler, 2015; Morfeld et al., 2014a; Panteliadis et al., 2014; Santos et al., 2019; Tartakovsky et al., 2020). Overall, there is some evidence from Germany that LEZs reduced PM_{10} concentrations by a few percent, but in other countries the evidence is much less clear, as Holman et al. (2015) summarized in their review. This is partly due to the fact that PM emitted from traffic exhaust accounts for a small fraction of ambient PM_{10} concentrations (Belis et al., 2013; Querol et al., 2004; Thorpe and Harrison, 2008). Indeed, elemental carbon (EC) is considered as a better indicator of diesel vehicle emission and more toxic than the regional background PM_{10} fraction (Janssen et al., 2011), and may be more suitable for evaluating the effects of LEZ on reducing traffic related emissions. Besides, LEZs showed weaker effects on NO_2 than PM_{10} (Jiang et al., 2017; Löschau et al., 2016), as NO_2 emission under real driving conditions has not been significantly improved from Euro 4 to Euro 6 (Anenberg et al., 2017; Franco et al., 2014; Lutz, 2018).

Many of the above-mentioned studies evaluated the effect of LEZ by comparing air pollutant concentrations before and after LEZ implementation or between cities with and without LEZs. However, a simple comparison of concentrations neglects several factors other than LEZs that affect the concentrations of air pollutants. These factors include meteorology, variations in the strength of emissions, long-range transport of aerosols and temporal factors such as season, public holidays, day of the week and rush hour times. Therefore, it is advisable to account for the confounding environmental conditions using the air pollutant concentrations at a reference site (Boogaard et al., 2012; Holman et al., 2015). Additionally, long-term measurements should also be used (Cyrys et al., 2014). In order to properly address these confounders, a general additive regression model was developed for validating the effects of LEZ on PM_{10} levels in Munich, Germany (Fensterer et al., 2014).

In the present study, the sophisticated model was applied in validating the effects of all stages of LEZs on PM_{10} and NO_2 in Berlin and Munich, respectively, and at both traffic sites and urban background sites. In addition, long-term monitoring data of EC and total carbon (TC) in Berlin provide a unique opportunity to evaluate the effectiveness of the LEZ on EC and TC in urban air. Overall, the aim of this study is to evaluate the effects of LEZs in reducing PM_{10}, NO_2, EC and TC concentrations in urban air.

2. Methods

2.1. Study area and period

The studies were conducted in the two German cities of Berlin and Munich. The LEZs in both cities are located in and around the city center, as depicted in Fig. 1. A detailed description of German LEZs is provided in the supplementary material part S1. Table 1 provides some information about the two LEZs. The LEZ in Munich has an area of 44 km^2, equivalent of 14% of the urban area. In Berlin, the largest city of Germany, the LEZ covers an area of 88 km^2, i.e. 10% of the urban area. The time of implementation of the LEZ stages were different in the two cities: in Berlin, stage 1 was implemented on January 1, 2008 and stage 3 on January 1, 2010 (note that in Berlin stage 2 LEZ was skipped); in Munich, stage 1 was effective on October 1, 2008, stage 2 on October 1, 2010, and stage 3 on October 1, 2012. There were no further restrictions of LEZ after 2012, although an extension of LEZ by introducing a blue sticker for cleaner diesel cars was argued by Lutz (2018). In addition to the LEZ, a heavy-duty vehicle (HDV) transit ban came into force in Munich on February 1, 2008 (eight months before the implementation of the LEZ): HDVs were not allowed to enter the city area if their final destination is not Munich.

Table 1 gives the time periods of LEZ 1, LEZ 2 and LEZ 3 for the analysis in the two cities. In addition, a period before the LEZ implementation was defined as reference period (LEZ 0): January 1, 2004–December 31, 2006 for Berlin and January 1, 2005–September 30, 2007 for Munich. The data within one year before the LEZ became effective were not included into the analysis. Rather, a buffer of one year between the reference period and the LEZ stage 1 (LEZ 1) was left. This is because the composition of the vehicle fleets in Munich and Berlin started to change rapidly within one year before the LEZ became officially effective. Thus, the introduction of the LEZ may affect the vehicle fleet immediately after the announcement of their implementation, and before the official LEZ launch date (Lutz, 2012).

2.2. Air pollution measurement data

Fig. 1 shows the locations of monitoring sites used in the analysis. Hourly PM_{10} and NO_2 data were collected from the official monitoring network. The measurement stations in Munich are operated by the Bavarian Environmental Agency (Bayerisches Landesamt für Umwelt, LfU), whereas the measurement stations in Berlin are operated by the Senate Department for the Environment, Transport and Climate Protection (Senatsverwaltung für Umwelt, Verkehr und Klimaschutz Berlin, SenUVK). As many monitoring sites were included and they were grouped in three categories: traffic sites (TS), urban background sites

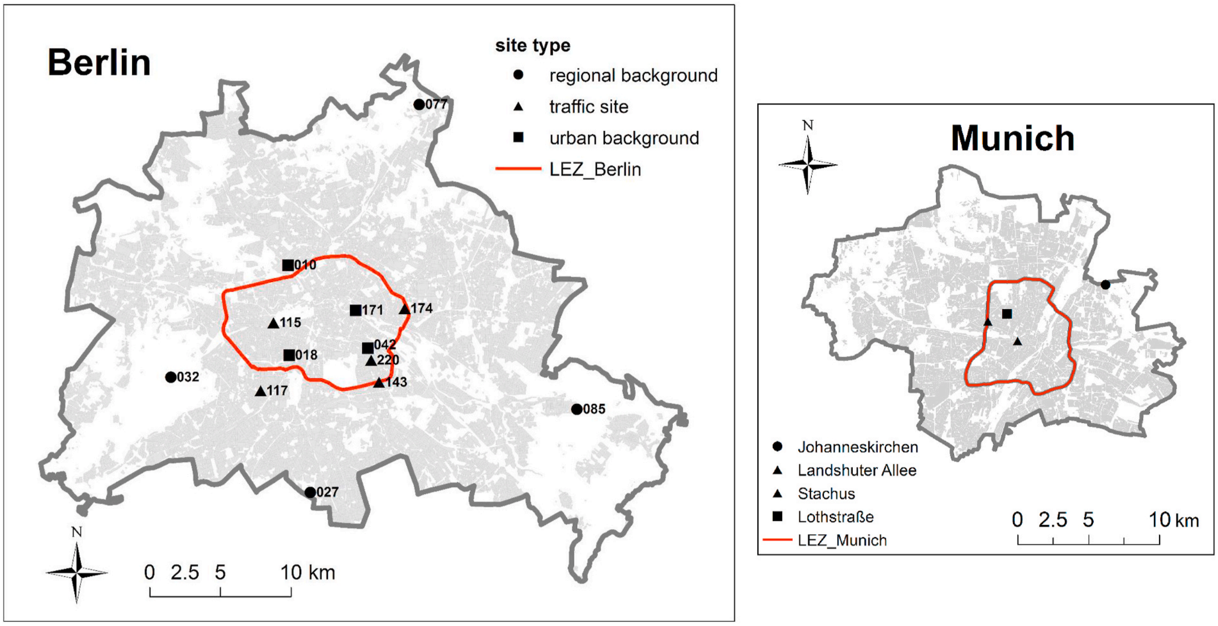

Fig. 1. LEZ area and monitoring sites in Berlin and Munich. The regional background sites are represented by rounds, urban background sites by squares, and traffic sites by triangles. Detailed information on the measurement sites in Berlin are provided in Table S1.

Table 1
Information of low emission zones in Berlin and Munich.

	Berlin	Munich
LEZ Area (% of urban area)	~88 km^2 (10%)	~44 km^2 (14%)
Population within LEZ (%)	~1.000.000 (29%)	~420.000 (32%)
HDV transit ban (start) [a]	No ban	Feb. 1, 2008
Reference period (LEZ 0)	Jan. 1, 2004–Dec. 31, 2006	Jan. 1, 2005–Sep. 30, 2007
Buffer [b]	Jan. 1, 2007–Dec. 31, 2007	Oct. 1, 2007–Sep. 30, 2008
LEZ 1	Jan. 1, 2008–Dec. 31, 2009	Oct. 1, 2008–Sep. 30, 2010
LEZ 2	–	Oct. 1, 2010–Sep. 30, 2012
LEZ 3	Jan. 1, 2010–Dec. 31, 2012	Oct. 1, 2012–Sep. 30, 2014

[a] HDV transit ban in Munich forbade trucks whose final destination is not the city from entering the city area.
[b] Buffer is a one-year period before the implementation of LEZ.

(UB), and regional background sites (RB) (details refer to Table S1 in the Supplementary Material).

2.2.1. *Berlin*

Hourly PM_{10} and NO_2 data were obtained from 13 official monitoring stations in Berlin: four RB stations, four urban background stations, and five traffic stations. Berlin is one of the few European cities where TC and EC concentrations were measured biweekly at major roads since the 1990s (Clemen and Kaupp, 2018). TC and EC data were available at four traffic sites and one urban background site. TC and EC were determined according to the method VDI (Verein Deutscher Ingenieure, The Association of German Engineers) 2465 part 2 (VDI 2465, 2016). Filter subsamples were first heated up to 650 °C in helium atmosphere, when the organic compounds were considered evaporated, and later oxidized to carbon dioxide (CO_2) which was determined by Nondispersive infrared (NDIR) sensor. In the second step, the filters were heated up to 700 °C with the presence of oxygen (O_2), the remaining soot were converted to CO_2 and analyzed by NDIR. TC was defined as the sum of organic matter (OM, OM= OC × 1.2) and EC.

2.2.2. *Munich*

Hourly PM_{10} and NO_2 data were used, specifically, from four LfU monitoring sites in Munich including one UB site (Lothstraβe), one traffic site on the border of LEZ (Landshuter Allee), one traffic site in the city center (Stachus), and one regional background site in the outskirts of the city and outside the LEZ (Johanneskirchen) (Regierung von Oberbayern, 2007). Meteorological parameters were obtained from a meteorological station from German Weather Service (Deutscher Wetterdienst, DWD) in Oberschleiβheim located northwest of Munich.

2.3. *Statistical models*

2.3.1. *Models for hourly PM_{10} and NO_2 data (model 1)*

Due to the diverse characteristics of the two LEZs, each LEZ was studied separately, which is different from Malina and Scheffler (2015) and Morfeld et al. (2014b) who combined the LEZs. Separate generalized additive models (GAM) were applied for the hourly PM_{10} and NO_2:

$$\log(P_i) = \beta_0 + \beta_1\log(P_{ref}) + \beta_{LEZ1} I_{LEZ1} + \beta_{LEZ2} I_{LEZ2} + \beta_{LEZ3} I_{LEZ3} + f_{LEZ0}(\text{hour}) I_{LEZ0} + f_{LEZ1}(\text{hour}) I_{LEZ1} + f_{LEZ2}(\text{hour}) I_{LEZ2} + f_{LEZ3}(\text{hour}) I_{LEZ3} + f_{wd}(\text{wind direction}) + \beta_2 \text{ public holiday} + \beta_3 \text{ season} + \beta_{Si} I_{Si} + \varepsilon \quad (1)$$

where P_i represents the concentrations of PM_{10} or NO_2 at the station of interest. Since the data of several stations were analyzed in a single model, an indicator function I_{Si} for the station i was included in the model. P_{ref} denotes the concentrations of PM_{10} or NO_2 at the reference station, i.e., the regional background station. If several reference stations were available, averaged values were used. I_{LEZj} is the indicator function for the stages of the low emission zone ($j = 0, 1, 2, 3$). Confounders include hour of the week, wind direction, public holidays and season. *hour* is the hour of the week; *wind direction* is a variable covering 0–360°. *Hour of the week* and *wind direction* are modeled as cyclic splines whose ends match, up to the second derivative. Thus, the effect of *wind directions* 0° and 360°, as well as *hour* 0 and 168, are the same. *Public holiday* is a dummy variable indicating the German public holidays; and *season* denotes a dummy variable indicating whether the season is summer (April–September) or winter (October–March). ε denotes the model error.

A sensitivity analysis was conducted using a slightly modified model (Model S1). Model S1 used averaged PM_{10} or NO_2 concentrations of the same types of monitoring stations (e.g., averaged hourly PM_{10} concentration from all traffic sites in Berlin). Details of the model S1 are provided in part S3 of the Supplementary Material.

The models were implemented in R, version 3.5.1 with the package "mgcv" (https://cran.r-project.org/web/packages/mgcv/mgcv.pdf).

2.3.2. Model for biweekly TC and EC data (model 2)

TC and EC concentrations in Berlin from four traffic sites (Nr. 117, 143, 220, 174, see Fig. 1) were included in the analysis, and an urban background site 042 (see Fig. 1) was used as reference site. In order to compare the results of the TC and EC levels with the results of the PM_{10}, biweekly PM_{10} concentrations based on the hourly data were calculated. Because of the biweekly temporal resolution, confounders such as hour of the week and national holidays were no longer relevant in model 2 (in comparison to model 1). The remaining influencing factors were the reference site concentration, LEZ stage, *wind direction*, *season*, and station:

$$log(P_i) = \beta_0 + \beta_1 log(P_{ref}) + \beta_{LEZ1} I_{LEZ1} + \beta_{LEZ2} I_{LEZ2} + \beta_{LEZ3} I_{LEZ3} + f_{wd}(wind\ direction) + \beta_3\ season + \beta_{Si} I_{Si} + \varepsilon \quad (2)$$

where P_i represents the concentration measurements of TC, EC or PM_{10} at the stations of interest. Model 2 was applied to biweekly TC, EC and PM_{10} concentrations in Berlin, respectively. In model 2, P-Splines were used for modelling the effect of the *wind direction*, because the biweekly *wind direction* data range from 79° to 283°.

The goodness of fit of the models to the data is assessed by the adjusted coefficient of determination. The results can be found in part S5 of the Supplemental Material.

2.4. Distinguish the LEZ effects from the natural vehicle renewal

The model analysis based on air quality data alone can barely disentangle the default vehicle fleet renewal from the additional net LEZ impact. In order to identify the sole impact of the LEZ from the normal fleet renewal effect, the changes in vehicle emissions of exhaust particles and NO_x in Berlin for the real-world case with LEZ in operation and for an artificial scenario reflecting the normal fleet turnover without the LEZ in force were used (Lutz, 2013). Briefly, the real-world fleet evolution in 2007 (before the introduction of the LEZ), as well as from 2008 till 2012, was obtained by recording the vehicle number plates at up to 10 of representative traffic spots and retrieving the Euro emission standard of the vehicles from the registration database. The scenario assuming the normal fleet turnover, i.e. the fleet turnover without an operating LEZ, was estimated using the Handbook Emission Factors for Road Transport (HBEFa, https://www.hbefa.net/e/index.html), which is the main tool or database for emission and traffic-related pollution modelling used in Germany and many other EU member states.

Based on the fleet data, the estimates of the annual vehicle emissions for NO_x and PM for the real-world LEZ case and for the without LEZ scenario were obtained. PM and NO_x emission reductions were calculated to compare 2008, 2010 and 2012 to the reference year 2007. For a detailed description refer to Lutz (2013).

The net contributions of the LEZ to the reduction of air pollution due to the renewal of the vehicle fleet were used to adjust the model results. Table 2 shows the reductions in air pollution by natural vehicle renewal and by accelerated vehicle renewal through the introduction of a LEZ, and the percentages of the contributions of LEZ. The percentages of LEZ were used to adjust the GAM model results for both Berlin and Munich (LEZ stages 1 and 3), even though a detailed data of the vehicle fleet composition on the road is missing for Munich. However, information from Munich's vehicle registration data suggests, that the situation in Munich does not differ much from that in Berlin. The share of cars compliant with LEZ stage 3 (green sticker) was more than 90% in both cities (Stadt München, 2012; SenGUV, 2011). The diesel share in Munich was slightly higher in Berlin, resulting in a potentially higher impact of LEZ stage 3. On the other hand, the exemption scheme of Munich was less strict than in Berlin, which could have compensated this bias.

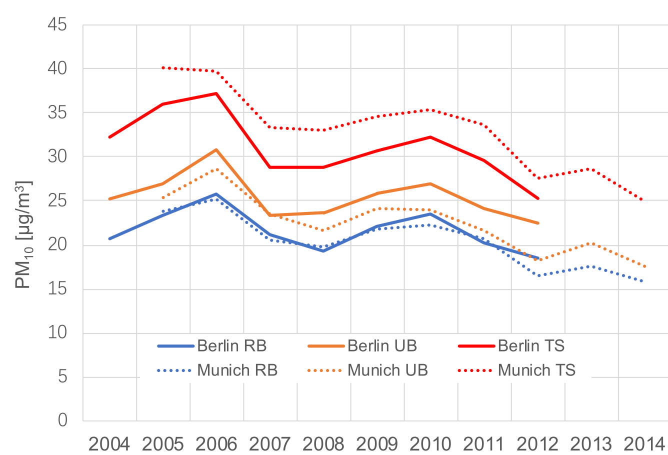

Fig. 2. Yearly mean PM_{10} concentrations in Berlin and Munich. RB: regional background sites; UB: urban background sites; TS: traffic sites.

Table 2
Reductions in exhaust particle emissions and NO_x (tons/year) in comparison to the reference year 2007 in Berlin from traffic originating from the natural vehicle fleet renewal and from real-world reduction (the natural fleet renewal and the LEZ).

	Year (LEZ stage)	Reduction by natural vehicle renewal [t/a]	Real-world reduction with LEZ [t/a]	Reduction solely due to LEZ [t/a]	Percentage of LEZ in total reduction	Percentage of LEZ (by stage)[a]
Particle	2008 (LEZ 1)	25	112	87	78%	73%
	2009 (LEZ 1)	51	154	103	67%	
	2010 (LEZ 3)	76	247	171	69%	63%
	2012 (LEZ 3)	123	281	158	56%	
NO_x	2008 (LEZ 1)	361	1609	1248	76%	72%
	2009 (LEZ 1)	743	2166	1453	67%	
	2010 (LEZ 3)	1103	2620	1517	58%	51%
	2012 (LEZ 3)	1736	3055	1319	43%	

[a] It is calculated by averaging the percentages of LEZ of two years, specifically, mean of 2008 and 2009 for LEZ stage 1, and mean of 2010 and 2012 for LEZ stage 3.

Table 3
Mean and median hourly PM_{10} concentrations (μg/m^3) before the LEZ (reference period, LEZ 0) and during different LEZ stages in Berlin and Munich.

City	Site type[a]	Variable	LEZ 0	LEZ 1	LEZ 2	LEZ 3
Berlin	RB	mean ±	24.4 ±	20.7 ±		21.0 ±
		std[b]	18.0	12.4		16.6
		median	19.9	17.9		16.3
	UB	mean ±	28.8 ±	24.6 ±		24.9 ±
		std	22.3	16.0		19.8
		median	23.6	21.6		19.8
		ratio[c]	1.18	1.19		1.19
		diff[d]	4.4	4.0		4.0
	TS	mean ±	36.5 ±	29.8 ±		29.0 ±
		std	32.0	31.6		22.5
		median	31.2	26.5		23.9
		ratio	1.50	1.44		1.38
		diff	12.2	9.2		8.0
Munich	RB	mean ±	22.9 ±	22.0 ±	19.5 ±	16.4 ±
		std	19.6	21.8	15.0	14.9
		median	18.0	17.5	15.5	13.0
	UB	mean ±	26 ±	24.5 ±	20.7 ±	18.6 ±
		std	22.2	23.2	14.4	15.5
		median	21.0	19.5	17.0	15.0
		ratio	1.14	1.11	1.07	1.14
		diff	3.1	2.4	1.3	2.2
	TS	mean ±	36.7 ±	35.2 ±	31.5 ±	26.7 ±
		std	26.0	25.1	20.4	21.0
		median	32.0	30.5	27.0	22.8
		ratio	1.61	1.6	1.62	1.63
		diff	13.9	13.2	12.0	10.3

[a] RB: regional background site; UB: urban background site; TS: traffic sites.
[b] Std: standard deviation.
[c] The ratios of the mean PM_{10} between UB or TS sites and the RB sites.
[d] The differences of the mean PM_{10} between UB or TS and the RB sites.

Because of the limitations in the estimation of the net LEZ contributions, and extra uncertainties when adjusting the Munich results using percentages from Berlin, the adjusted net LEZ effects were presented as an addition to the main GAM model results.

3. Results

3.1. PM_{10}

3.1.1. Average PM_{10} levels before and after LEZ implementation

Fig. 2 shows the time series of PM_{10} yearly mean concentrations in two cities from 2004 to 2014. PM_{10} was highest at traffic sites, followed by urban and regional background sites. The differences between traffic and background sites (RB and UB) were larger in Munich than in Berlin.

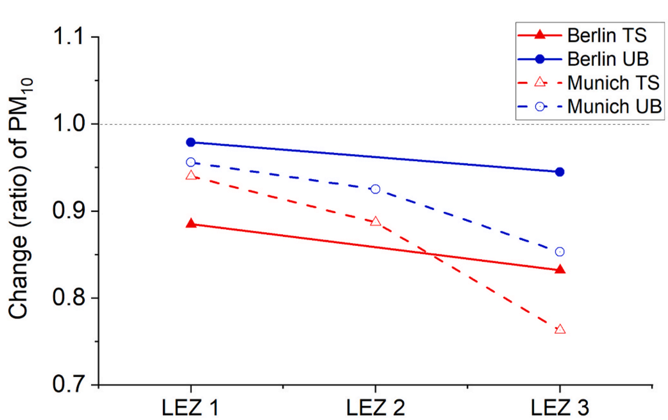

Fig. 3. Changes of PM_{10} concentrations in Berlin and Munich in LEZ stage 1–3 compared with the period before the introduction of LEZ. The results for TS and UB were obtained from model 1, and RB is used as reference.

There is an overall decreasing trend of PM_{10} concentrations in Berlin (2004–2012) and Munich (2005–2014), although inter-annual variations exist.

Table 3 shows the PM_{10} concentrations before and during the operation of LEZs (LEZ 0, LEZ 1, LEZ 2 and LEZ 3) at all three types of measurement sites in Berlin and Munich. The ratios and differences were calculated against RB concentrations, and used as indicators of absolute and relative deviations in PM_{10} between TS and RB, as well as UB and RB.

In Berlin, PM_{10} concentrations decreased (statistically significantly using Mann-Whitney U test) from LEZ 0 to LEZ 1 for all three types of monitoring sites, but barely from LEZ 1 to LEZ 3. The ratios and differences of PM_{10} were very close between LEZ 0 and LEZ 3 at UB, but decreased at TS. In Munich, PM_{10} decreased significantly and consistently from LEZ 0 to LEZ 3. At TS, the ratios of PM_{10} to the RB concentrations were very similar among LEZ 0, 1, 2 and 3; by contrast, the differences between TS and RB decreased from LEZ 0 to LEZ 3. At UB sites of Munich, the ratios and differences of PM_{10} to RB decreased from LEZ 0 to LEZ 2, and increased from LEZ 2 to LEZ 3.

3.1.2. Effects of LEZ - hourly PM_{10} (model 1)

Fig. 3 shows the effects of LEZ (LEZ with HDV transit ban in Munich) on PM_{10} at TS and UB sites in Berlin and Munich. Detailed results are presented in Table 4. It should be noted that RB site is not shown since it is used as reference for the adjustment of UB and TS.

In Berlin, significant reductions of PM_{10} concentrations of 2.1% (UB) and 11.5% (TS) during LEZ 1, and 5.5% (UB) and 16.8% (TS) during LEZ 3, were observed compared to LEZ 0. The reductions were greater at TS than at UB. In Munich, significant and gradually stronger decreases of PM_{10} concentrations were observed in parallel to the introduction of LEZ 1, LEZ 2 and finally LEZ 3: in comparison to LEZ 0, the PM_{10} concentrations were reduced by 4.4% (UB) and 6.0% (TS) during LEZ 1, by 7.5% (UB) and 11.3% (TS) during LEZ 2, and by 14.7% (UB) and 23.7% (TS) during LEZ 3. In LEZ 1, there were very small differences in the reduction effects between UB and TS. In LEZ 2 the difference became larger, while in LEZ 3, a very pronounced difference was observed between UB and TS. Table 4 also shows the net effects on PM_{10} which are adjusted for the renewal of the vehicle fleet using the percentages from Table 2, which were 73%% and 63% of the GAM model results for LEZ stage 1 and 3, respectively. The highest net LEZ effects for PM_{10} in Berlin (10.5%) and Munich (14.8%) were at traffic sites at LEZ stage 3.

Table 4
Changes of PM_{10} concentrations in Berlin and Munich in LEZ stage 1–3 compared with period before the introduction of LEZ. The results for TS and UB were obtained from model 1, and RB is used as reference.

City	Site type[a]	Effect	95% CI	p-value	Net LEZ effect[b]
Berlin		LEZ 1			
	UB	−2.1%	(-3.0%, −1.3%)	<0.001	−1.5%
	TS	−11.5%	(-12.5%, −10.5%)	<0.001	−8.3%
		LEZ 3			
	UB	−5.5%	(-6.3%, −4.7%)	<0.001	−3.4%
	TS	−16.8%	(-17.6%, −15.9%)	<0.001	−10.5%
Munich		LEZ 1 + HDV transit ban			
	UB	−4.4%	(-6.8%, −2.0%)	<0.001	−3.2%
	TS	−6.0%	(-8.0%, −4.0%)	<0.001	−4.4%
		LEZ 2 + HDV transit ban			
	UB	−7.5%	(-9.8%, −5.2%)	<0.001	NA
	TS	−11.3%	(-13.2%, −9.4%)	<0.001	NA
		LEZ 3 + HDV transit ban			
	UB	−14.7%	(-16.8%, −12.6%)	<0.001	−9.2%
	TS	−23.7%	(-25.3%, −22.1%)	<0.001	−14.8%

[a] UB: urban background sites; TS: traffic sites.
[b] Adjusted for renewal of vehicle fleet using the percentages in Table 2. Effects for LEZ 2 cannot be adjusted.

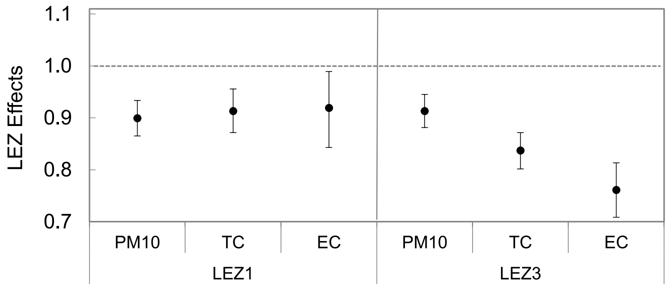

Fig. 4. Changes of biweekly PM_{10}, TC and EC at traffic sites in Berlin in LEZ stage 1 and 3 compared with the period before the introduction of LEZ. Error bars indicate the 95% confidence interval. The results for traffic sites were obtained from model 2, and urban background site is used as reference.

The effects by seasons are shown in part S4 of the Supplementary Material.

3.1.3. *Effects of LEZ - biweekly PM_{10}, TC and EC in berlin (model 2)*

In addition to PM_{10}, the effects of the LEZ on TC and EC were also evaluated in Berlin. Fig. 4 shows the effects of LEZ on biweekly PM_{10}, TC and EC concentrations in Berlin using model 2. Note that in model 2 the reference site is an UB site within the Berlin LEZ. All effects shown in the figure are statistically significant. All three air pollutants showed reductions of around 10% (PM_{10}: 11.1%, TC: 9.7%, EC: 9.1%) in LEZ stage 1. Reductions of TC and EC concentrations were much larger in LEZ stage 3 compared to PM_{10} (PM_{10}: 9.7%, TC: 17.3%, EC: 24.9%).

When adjusting the model 2 results for the net renewal of the vehicle fleet due to LEZ, the net effects of LEZ stage 1 become 8.0% (PM_{10}), 7.0% (TC) and 6.6% (EC), and the net effects of LEZ stage 3 become 6.1% (PM_{10}), 10.8% (TC) and 15.6% (EC).

3.2. NO_2

It should be noted that LEZs were implemented to regulate PM_{10} and other PM components, but not to regulate NO_2. Nevertheless, it might be of interest to observe the indirect effects on NO_2.

3.2.1. *Average NO_2 levels before and after LEZ implementation*

Fig. 5 shows the time series of the NO_2 yearly mean concentrations in two cities from 2004 to 2014. There was a clear gradient in NO_2 levels between TS, UB and RB sites (much more pronounced than PM_{10}). In Berlin, the NO_2 concentrations were generally stable from 2004 to 2012; however, in Munich, NO_2 decreased from 2005 to 2012, and stabilized from 2012 to 2014.

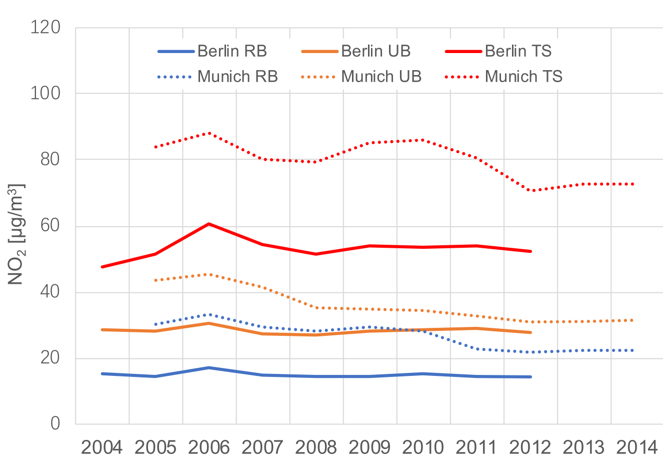

Fig. 5. Yearly mean NO_2 concentrations in Berlin and Munich. RB: regional background sites; UB: urban background sites; TS: traffic sites.

Table 5
Mean and median hourly NO_2 concentrations (µg/m^3) before LEZ (reference period, LEZ 0) and during different LEZ stages in Berlin and Munich.

City	Site type[a]	Variable	LEZ 0	LEZ 1	LEZ 2	LEZ 3
Berlin	RB	mean ± std[b]	15.7 ± 10.0	14.5 ± 9.0		14.6 ± 9.8
		median	13.6	12.5		12.3
	UB	mean ± std	29.3 ± 15.6	27.5 ± 15.1		28.4 ± 16.1
		median	26.1	24.3		24.8
		ratio[c]	1.86	1.89		1.90
		diff[d]	13.6	13.0		13.8
	TS	mean ± std	56.1 ± 24.0	52.7 ± 22.6		53.3 ± 24.2
		median	53.3	50.5		50.7
		ratio	3.56	3.63		3.6
		diff	40.3	38.2		38.7
Munich	RB	mean ± std	31.7 ± 19.7	28.9 ± 18.5	23.3 ± 16.8	22.6 ± 15.1
		median	26.5	24.5	18.5	19.0
	UB	mean ± std	43.8 ± 23.6	34.5 ± 21.1	32.6 ± 20.2	31.5 ± 18.7
		median	38.0	30.0	28.0	27.5
		ratio	1.38	1.2	1.4	1.39
		diff	12.1	5.7	9.2	8.9
	TS	mean ± std	84.7 ± 32.8	86 ± 33.6	77.4 ± 30.3	71.7 ± 29.3
		median	82.5	82.8	74.8	69.3
		ratio	2.67	2.98	3.31	3.17
		diff	53.0	57.1	54.0	49.1

[a] RB: regional background site; UB: urban background site; TS: traffic sites.
[b] Std: standard deviation.
[c] The ratios of the mean NO_2 between UB or TS sites and the RB sites.
[d] The differences of the mean NO_2 between UB or TS sites and the RB sites.

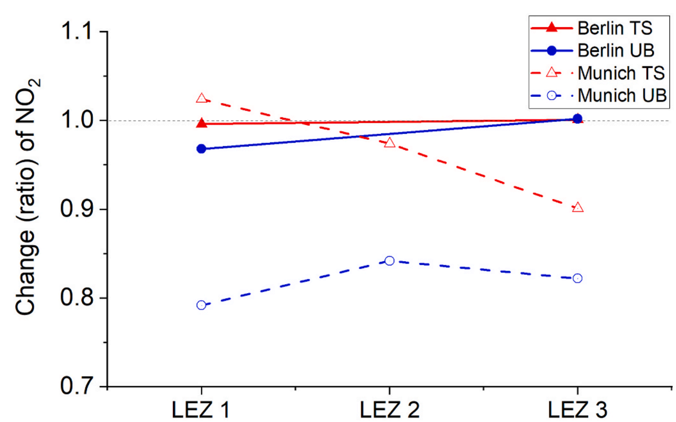

Fig. 6. Changes of NO_2 concentrations in Berlin and Munich in LEZ stage 1–3 compared with the period before the introduction of LEZ. The results for TS and UB were obtained from model 1, and RB is used as reference.

Table 5 shows the NO_2 concentrations before and during the operation of LEZs (LEZ 0, LEZ 1, LEZ 2 and LEZ 3) at all three types of measurement sites in Berlin and Munich. The ratios and differences were calculated against the regional background concentrations, and were used as indicators of 他the absolute and relative deviation between traffic or urban background site and regional background site. The ratios and differences of NO_2 (TS vs. RB, UB vs. RB) in Berlin did not change with the stages of LEZ (LEZ 1, LEZ 3) compared to LEZ 0. In Munich, an increasing trend in NO_2 ratios between TS and RB was observed.

3.2.2. *Effects of LEZ - hourly NO_2 (model 1)*

Fig. 6 shows the effects of the LEZs (LEZ with HDV transit ban in Munich) on reducing NO_2 concentrations in Berlin and Munich. Detailed results are presented in Table 6. In Berlin, the effects for NO_2 were small

Table 6
Changes of NO_2 concentrations in Berlin and Munich in LEZ stage 1–3 compared with period before the introduction of LEZ. The results for TS and UB were obtained from model 1, and RB is used as reference.

City	Site type[a]	Effect	95% CI	p-value	Net LEZ effect[b]
Berlin		LEZ 1			
	UB	−3.2%	(-4.3%, −2.1%)	<0.001	−2.3%
	TS	−0.4%	(-1.7%, 0.9%)	0.517	−0.3%
		LEZ 3			
	UB	0.2%	(-0.8%, 1.2%)	0.667	0.1%
	TS	−0.1%	(-1.0%, 1.3%)	0.856	−0.1%
Munich		LEZ 1 + HDV transit ban			
	UB	−20.8%	(-22.3%, −19.4%)	<0.001	−14.9%
	TS	2.4%	(0.9%, 3.9%)	0.002	1.7%
		LEZ 2 + HDV transit ban			
	UB	−15.8%	(-17.4%, −14.3%)	<0.001	NA
	TS	−2.6%	(-4.0%, −1.1%)	0.001	NA
		LEZ 3 + HDV transit ban			
	UB	−17.8%	(-19.3%, −16.4%)	<0.001	−9.0%
	TS	−9.9%	(-11.2%, −8.6%)	<0.001	−5.0%

[a] UB: urban background sites; TS: traffic sites.
[b] Adjusted for renewal of vehicle fleet using the percentages in Table 2. Effects for LEZ 2 cannot be adjusted.

and not statistically significant, except for LEZ 1 at UB. At the traffic sites in Munich, there were very small effects for LEZ 1 (increment of 2.4%) and LEZ 2 (reduction of 2.6%). A statistically significant reduction of about 10% was observed in LEZ 3. Overall, the effect of the LEZ on reducing NO_2 concentration was stronger from LEZ 1 to LEZ 3 at Munich traffic sites. In contrast, a large decrease of 20.8% in NO_2 concentration was seen in LEZ 1 at the Munich urban background site (Lothstraβe), while during the subsequent stages of the LEZ the reductions became smaller (15.8% in LEZ 2, 17.8% in LEZ 3). There was no clear trend of NO_2 reduction with LEZ stages.

Table 6 also shows the net effects on NO_2 adjusted for the net renewal of the vehicle fleet by LEZ using the percentages from Table 2, which were 72% and 51% of the GAM model results for LEZ stage 1 and 3, respectively.

The effects by seasons are shown in part S4 of the Supplementary Material.

4. Discussion

4.1. Trend of PM_{10} and NO_2 concentrations

As Figs. 2 and 5 show, the time trend in Germany is quite different for PM_{10} and NO_2. The decreasing trend of PM_{10} concentration reflects the use of particle traps in vehicles, the introduction of LEZs and the reduction of PM emissions from industry (Luft, 2002) and fuel combustion and residential heating (European Environment Agency, 2018). In contrast, the situation of NO_2 is based on four influencing factors (Bruckmann et al., 2019) which together result in the absence of a decreasing trend for NO_2. Firstly, the percentage of passenger cars with Diesel engines increased from about 14% in 2000 to 33% in 2018 (Kraftfahrt-Bundesamt, 2019). Secondly, there was a shift in the conversion of NO to NO_2 in the exhaust gas in Diesel cars by the use of oxidation catalysts (Kurtenbach et al., 2008). Thirdly, the real driving emissions of NO_x from Euro 4 and 5 diesel cars haven't been improved much compared with Euro 3 (Lutz, 2018); therefore, the vehicle fleet update may have a limited effect in reducing NO_x. And lastly, the manipulation of the software of some automobile companies with frequent deactivation of exhaust cleaning in real driving mode further underestimated the real NO_2 emissions (Borgest, 2017).

Furthermore, one should keep in mind that the observation of health effects from $PM_{2.5}$ on mortality, respiratory and cardiovascular morbidity is considered as causal or likely causal (US EPA, 2010) whereas for NO_2 this causal relationship is used only for the risk of asthma and other respiratory endpoints (US EPA, 2016). WHO comes to similar conclusions that with respect to mortality, a risk coefficient for 1 $\mu g/m^3$ PM_{10} is clearly higher than the risk coefficient for 1 $\mu g/m^3$ NO_2 (WHO, 2013a, b). As a result, the estimated years of life lost in Germany in 2014 were 5 times higher from $PM_{2.5}$ than for NO_2 (Wichmann, 2018).

4.2. Effects of LEZ on PM_{10} and PM components

Significant decreases of PM_{10} concentrations after the enforcement of LEZs (in Munich in combination with HDV transit ban) were observed in Berlin and Munich for both UB and TS. The more rigorous restrictions from LEZ (i.e., stage 3 LEZs) led to a larger reduction of PM_{10}. Stronger effects on PM_{10} were found at traffic sites in comparison to UB sites, which is in line with the fact that the share of PM_{10} attributable to vehicular exhaust is larger at a TS site than at an UB site. Our results are in line with the results obtained by Fensterer et al. (2014), who in a first analysis of the Munich data from 2006 to 2010, used a very similar statistical model to estimate the changes in PM_{10} concentrations after the implementation of the LEZ. The reduction of PM_{10} concentrations after the implementation of LEZ stage 1 was larger at a single traffic site (13.0%) and smaller in the UB site (4.5%). A very similar reduction of 4.4% at the background site was observed, whereas the reduction at two traffic sites was somewhat smaller (6.0%) compared to Fensterer et al. (2014). However, in Fensterer et al. (2014) data from the traffic site at Prinzregentenstraβe were used, while in present study data from two other sites: Stachus and Landshuter Allee were used. The monitoring sites at Prinzregentenstraβe was closed and the data for the time periods of LEZ 2 and LEZ 3 were not available. Compared to Fensterer et al. (2014), the current study is extended to LEZ stages 2 and 3 in Munich (showing further reduction of PM_{10}), included the effects of LEZ on NO_2, and additionally includes Berlin LEZ for PM_{10}, NO_2, TC and EC.

TC and EC concentrations in Berlin were reduced more strongly than PM_{10} concentrations after the introduction of LEZ stage 3 (PM_{10}: 10%, TC: 17%, EC: 25%). The results confirm the argument of Cyrys et al. (2014) that EC or BC would be much better indicators when evaluating the effectiveness of LEZs. Both parameters are more specific metrics for combustion related particles than PM_{10}, and in this way also for traffic exhaust emissions. The combination of PM_{10} and BC monitoring in urban areas could potentially generate a useful approach in evaluating the impact of road traffic emissions on air quality. Unfortunately, BC or EC concentrations are not currently routinely measured in urban air quality networks. As BC is considered as a highly health-relevant particle fraction, the implementation of LEZs may lead to a higher health benefit than its literal reduction to the PM_{10} mass concentration.

4.3. Effects of LEZ on NO_2

The NO_2 concentrations responded differently to LEZs (in Munich in combination with HDV transit ban) than PM_{10}, and differently between Berlin and Munich. Only very small and mainly non-significant effects without a clear tendency were found in Berlin. In contrast, NO_2 at UB in Munich significantly decreased with the introduction of the LEZ but the NO_2 levels did not further decrease during LEZ 2 and LEZ 3. Reductions of NO_2 concentrations at TS in Munich were observed only from LEZ 2. The decrease of NO_2 concentrations due to the introduction of LEZs was much weaker than for particulate matter. In 2012 to 2017, at around 60% of the traffic sites in Germany, exceedances of the limit values for NO_2 are still being observed (UBA, 2017).

In Munich, HDV transit ban was implemented before LEZ 1, while no such measure was introduced in Berlin. The HDV transit ban may have contributed to the decrease of NO_2 in Munich. There are two possible explanations for the observed reductions of NO_2 at the Munich UB site.

First, the HDV transit ban came into force eight months before LEZ 1. This might have had an overall effect on the urban background concentration of NO_2 in Munich. As the effects do not become stronger from LEZ 1 to LEZ 3, it is likely that the LEZ does not contribute to the reduction of NO_2 at urban background site. However, the HDV transit ban seemes to have a weaker effect on NO_2 at traffic sites. The second explanation could be the local influences on NO_2 at Munich urban background site. In this study, only one urban background site is available in Munich (Lothstraβe). Some potentially unknown changes in local NO_2 sources near Lothstraβe before LEZ 1 implementation may have caused such results. This underlines the disadvantage of relying on one single monitoring site. In contrast, the current study includes two traffic sites in Munich, four urban background sites and five traffic sites in Berlin.

4.4. Strengths and limitations

The regression models developed in this study used the variable "concentrations of air pollutants at regional background" as reference. This allowed us to account for many confounders including meteorology, diurnal variation, long-range transport of air pollution, and secondary aerosol formation that may influence the absolute levels of air pollutants. In addition, the variable "the stages of LEZs" already implies the impact on traffic emissions, which may be caused by vehicle fleet update (modal shift) or change in the traffic volume. As a result, including the traffic variables (such as traffic volume and modal shift in the car fleet composition) in the model is not necessary. This model was also used to evaluate the LEZ on the key components of PM_{10} (EC and TC).

Some limitations are inherent to our methodology. When comparing the relative difference of air pollutants between TS and RB, and between UB and RB adjusting for confounding variables, it cannot be ruled out that other mitigation measures (e.g., speed limit control, HDV transit ban, traffic rerouting, and natural vehicle renewal) than the LEZ were the cause for the observed differences. Therefore, other mitigation measures or policies that influenced the car fleet or emission of vehicular exhaust could not be distinguished by the GAM models. During the economic crisis from 2008, the German government initiated a car scrappage program in the beginning of 2019 to stimulate the economy; however, it was launched one year later than the LEZs which already banned the old cars in 2008. In Munich, a HDV transit ban was introduced at the similar time with LEZ. In Berlin, no additional control measures on traffic were introduced. We tried to differentiate the effects of natural vehicle renewal from net LEZ effects in reducing the PM_{10} and NO_2 concentrations by estimation of the emission reductions in a real-world scenario with LEZ and in a normal fleet turnover without LEZ, respectively.

The second limitation is due to the availability of monitoring sites and data. In Berlin, there is a sufficient number of monitoring sites (4 RB, 4 UB and 5 TS), while in Munich, the number of monitoring stations is more limited (1 RB, 1 UB and 2 TS). In the case that only a single monitoring station is available, the result is more susceptible to, e.g., the change of any local sources in the long-term measurement. In contrast, when multiple sites of the same category are available, the mean urban air pollution level represents the estimations more robustly.

4.5. Final remarks on LEZ

There have been doubts before the implementation of LEZs about whether they could be a suitable and effective measure in improving urban air quality. Many studies have been carried out to evaluate the effects of LEZs. In the case of Germany, most studies have found an effect in reducing PM_{10} concentrations (Cyrys et al., 2014; Fensterer et al., 2014; Lutz and Rauterberg-Wulff, 2009; Malina and Scheffler, 2015). Our study showed a clear effect for PM_{10} in the range of 2%–24%, depending on the site category (background vs. traffic) and the LEZ stage. The effect for EC in Berlin is much stronger than for PM_{10}. The effect on NO_2 is not clear and not consistent, which might be caused by the fact that NO_2 emissions in real-life driving conditions have not been significantly reduced.

After many years of implementation of LEZs, the vehicle fleet today meets the most stringent requirement of LEZ stage 3; therefore, the additive effect of LEZ now may be smaller than right after its implementation. Nevertheless, a mitigation measure, such as LEZs, which targets the major primary air pollution source in a highly populated city center could be a feasible way to improve the urban air quality.

5. Conclusions

Overall, our study showed that LEZs (in Munich in combination with the HDV transit ban) proved to be effective in reducing PM_{10}. The strongest effects were observed in LEZ stage 3, the strictest stage so far. LEZs are were effective in reducing PM_{10} concentrations near traffic than in urban background areas. In contrast, it is not clear whether the LEZ was responsible for the reduced NO_2 concentrations; more likely the HDV transit ban contributed to the observed NO_2 reduction in Munich.

The results clearly demonstrated that the LEZ had a much larger effect in reducing the EC concentration near traffic than for PM_{10} (25% vs. 10% in LEZ 3 in Berlin). This has a twofold implication: firstly, LEZ is more efficient in mitigating the more toxic fraction of PM_{10} (soot particles); secondly, the health benefit of the LEZ may be larger than the estimation based on the reduction of PM_{10}.

Author contribution

Jianwei Gu: Conceptualization, Investigation, Writing - Original Draft **Veronika Deffner**: Methodology, Investigation, Software, Writing - Review & Editing **Helmut Küchenhoff**: Investigation **Regina Pickford**: Writing - Review & Editing **Susanne Breitner**: Writing - Review & Editing **Alexandra Schneider**: Writing - Review & Editing **Michal Kowalski**: Investigation **Annette Peters**: Writing - Review & Editing **Martin Lutz**: Methodology, Formal analysis, Writing - Review & Editing **Andreas Kerschbaumer**: Investigation, Resources **Rémy Slama**: Writing - Review & Editing **Xavier Morelli**: Writing - Review & Editing **Heinz-Erich Wichmann**: Writing - Review & Editing, Supervision **Josef Cyrys**: Conceptualization, Supervision, Writing - Review & Editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2021.114048.

References

Anenberg, S.C., Miller, J., Minjares, R., Du, L., Henze, D.K., Lacey, F., Malley, C.S., Emberson, L., Franco, V., Klimont, Z., Heyes, C., 2017. Impacts and mitigation of excess diesel-related NOx emissions in 11 major vehicle markets. Nature 545, 467.

Anenberg, S.C., Henze, D.K., Tinney, V., Kinney, P.L., Raich, W., Fann, N., Malley, C.S., Roman, H., Lamsal, L., Duncan, B., Martin, R.V., van Donkelaar, A., Brauer, M., Doherty, R., Jonson, J.E., Davila, Y., Sudo, K., Kuylenstierna, J.C.I., 2018. Estimates of the global burden of ambient PM2.5, ozone, and NO2 on asthma incidence and emergency room visits. Environ. Health Perspect. 126, 14.

Beelen, R., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z.J., Weinmayr, G., Hoffmann, B., Wolf, K., Samoli, E., Fischer, P., Nieuwenhuijsen, M., Vineis, P., Xun, W.W., Katsouyanni, K., Dimakopoulou, K., Oudin, A., Forsberg, B., Modig, L., Havulinna, A.S., Lanki, T., Turunen, A., Oftedal, B., Nystad, W., Nafstad, P., De Faire, U., Pedersen, N.L., Östenson, C.-G., Fratiglioni, L., Penell, J., Korek, M., Pershagen, G., Eriksen, K.T., Overvad, K., Ellermann, T., Eeftens, M., Peeters, P.H., Meliefste, K., Wang, M., Bueno-de-Mesquita, B., Sugiri, D., Krämer, U., Heinrich, J., de Hoogh, K., Key, T., Peters, A., Hampel, R., Concin, H., Nagel, G., Ineichen, A., Schaffner, E., Probst-Hensch, N., Künzli, N., Schindler, C., Schikowski, T., Adam, M., Phuleria, H., Vilier, A., Clavel-Chapelon, F., Declercq, C., Grioni, S., Krogh, V., Tsai, M.-Y., Ricceri, F., Sacerdote, C., Galassi, C., Migliore, E., Ranzi, A., Cesaroni, G., Badaloni, C., Forastiere, F., Tamayo, I., Amiano, P., Dorronsoro, M., Katsoulis, M., Trichopoulou, A., Brunekreef, B., Hoek, G., 2014. Effects of long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts within the multicentre ESCAPE project. Lancet 383, 785–795.

Belis, C.A., Karagulian, F., Larsen, B.R., Hopke, P.K., 2013. Critical review and meta-analysis of ambient particulate matter source apportionment using receptor models in Europe. Atmos. Environ. 69, 94–108.

Boogaard, H., Janssen, N.A.H., Fischer, P.H., Kos, G.P.A., Weijers, E.P., Cassee, F.R., van der Zee, S.C., de Hartog, J.J., Meliefste, K., Wang, M., Brunekreef, B., Hoek, G., 2012. Impact of low emission zones and local traffic policies on ambient air pollution concentrations. Sci. Total Environ. 435, 132–140.

Borgest, K., 2017. Manipulation von Abgaswerten. Springer, Wiesbaden, Germany.

Brook, R.D., Rajagopalan, S., Pope 3rd, C.A., Brook, J.R., Bhatnagar, A., Diez-Roux, A.V., Holguin, F., Hong, Y., Luepker, R.V., Mittleman, M.A., Peters, A., Siscovick, D., Smith Jr., S.C., Whitsel, L., Kaufman, J.D., 2010. Particulate matter air pollution and cardiovascular disease: an update to the scientific statement from the American Heart Association. Circulation 121, 2331–2378.

Bruckmann, P., Krämer, U., Wichmann, H.E., 2019. Wissenschaft trifft Politik – die Basis der europäischen Grenzwerte für Stickstoffdioxid und Feinstaub. Umweltmed – Hygiene – Arbeitsmed 24, 83–100.

Brunekreef, B., Künzli, N., Pekkanen, J., Annesi-Maesano, I., Forsberg, B., Sigsgaard, T., Keuken, M., Forastiere, F., Barry, M., Querol, X., Harrison, R.M., 2015. Clean air in Europe: beyond the horizon? Eur. Respir. J. 45, 7–10.

Cesaroni, G., Boogaard, H., Jonkers, S., Porta, D., Badaloni, C., Cattani, G., Forastiere, F., Hoek, G., 2012. Health benefits of traffic-related air pollution reduction in different socioeconomic groups: the effect of low-emission zoning in Rome. Occup. Environ. Med. 69, 133–139.

Clemen, S., Kaupp, H., 2018. Two decades of soot measurements in the Berlin air monitoring network (in German). Gefahrst. Reinhalt. Luft 78, 109–116.

Council of the European Union, 2008. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on Ambient Air Quality and Cleaner Air for Europe. Official Journal of the European Union, pp. 1–44.

Cyrys, J., Peters, A., Soentgen, J., Wichmann, H.E., 2014. Low emission zones reduce PM10 mass concentrations and diesel soot in German cities. J. Air Waste Ma. 64, 481–487.

Degraeuwe, B., Thunis, P., Clappier, A., Weiss, M., Lefebvre, W., Janssen, S., Vranckx, S., 2017. Impact of passenger car NOX emissions on urban NO2 pollution – scenario analysis for 8 European cities. Atmos. Environ. 171, 330–337.

Ellison, R.B., Greaves, S.P., Hensher, D.A., 2013. Five years of London's low emission zone: effects on vehicle fleet composition and air quality. Transport Res D-Tr. E. 23, 25–33.

Emplan, 2010. Lufthygienische Untersuchung (Augsburg, Germany).

European Environment Agency, 2018. Air Quality in Europe — 2018 Report. EEA Report No 12/2018.

Fensterer, V., Kuchenhoff, H., Maier, V., Wichmann, H.E., Breitner, S., Peters, A., Gu, J. W., Cyrys, J., 2014. Evaluation of the impact of low emission zone and heavy traffic ban in Munich (Germany) on the reduction of PM10 in ambient air. Int. J. Environ. Res. Publ. Health 11, 5094–5112.

Franco, V., Sánchez, F.P., German, J., Mock, P., 2014. Real-World Exhaust Emissions from Modern Diesel Cars - A Meta-Analysis of PEMS Emissions Data from EU (EURO 6) and US (Tier 2 BIN 5/ULEV II) Diesel Passenger Cars. Part 1: Aggregated Results. International Council on Clean Transportation Europe, Berlin.

Hoek, G., Krishnan, R.M., Beelen, R., Peters, A., Ostro, B., Brunekreef, B., Kaufman, J.D., 2013. Long-term air pollution exposure and cardio- respiratory mortality: a review. Environ. Health 12, 43.

Holman, C., Harrison, R.M., Querol, X., 2015. Review of the efficacy of low emission zones to improve urban air quality in European cities. Atmos. Environ. 111, 161–169.

Janssen, N.A.H., Hoek, G., Simic-Lawson, M., Fischer, P., van Bree, L., Brink, H.t., Keuken, M., Atkinson, R.W., Anderson, H.R., Brunekreef, B., Cassee, F.R., 2011. Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM10 and PM2.5. Environ. Health Perspect. 119, 1691–1699.

Jiang, W., Boltze, M., Groer, S., Scheuvens, D., 2017. Impacts of low emission zones in Germany on air pollution levels. Transp. Res. Proc. 25, 3370–3382.

Kraftfahrt-Bundesamt, 2019. Fahrzeugzulassungen (FZ) Bestand an Kraftfahrzeugen nach Umwelt-Merkmalen. Kraftfahrt-Bundesamt, Flensburg, Germany.

Kurtenbach, R., Becker, K.H., Bruckmann, P., Kleffmann, J., Niedojadlo, A., Wiesen, P., 2008. Das innerstädtische stickstoffdioxid (NO2)-problem: welchen einfluss haben direkte verkehrsemissionen? Mitt Umweltchem Ökotox 14, 69–73.

LfU, 2010. Minderungswirkung von Umweltzonen auf die Luftschadstoffbelastung-Umweltzone München. Bayerisches Landesamt für Umwelt.

Löschau, G., Wiedensohler, A., Birmili, W., Rasch, F., Spindler, G., Müller, K., Wolf, U., Hausmann, A., Böttger, M., Bastian, S., Anhalt, M., Dietz, V., Herrmann, H., Uwe, B., 2015. Messtechnische Begleitung der Einführung der Umweltzone in der Stadt Leipzig, Teil 4: Immissionssituation von 2010 bis 2014 und Wirkung der Umweltzone. Landesamt für Umwelt, Landwirtschaft und Geologie (Dresden).

Löschau, G., Wiedensohler, A., Birmili, W., Rasch, F., Spindler, G., Müller, K., Wolf, U., Hausmann, A., Anhalt, M., Dietz, V., Herrmann, H., Uwe, B., Horst-Günter, K., 2016. Messtechnische Begleitung der Einführung der Umweltzone in der Stadt Leipzig, Teil 5: Immissionssituation von 2010 bis 2015 und Wirkung der Umweltzone. Landesamt für Umwelt, Landwirtschaft und Geologie (Dresden).

Luft, T., 2002. Erste Allgemeine Verwaltungsvorschrift zum Bundes-Immissionsschutzgesetz: Technische Anleitung zur Reinhaltung der Luft – TA Luft vom 24.7.2002. Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit, pp. 511–605.

Lutz, M., 2012. Modellierung der verkehrsnahen NO2-Belastung 2009: Symposium: Wie gesund ist unsere Stadt? Senatsverwaltung für Stadtentwicklung und Umwelt. Referat Immissionsschutz, Graz, Austria.

Lutz, M., 2013. Mitigation strategies, Berlin, Germany. In: Cruz, M., Minguillón, M Viana, Querol, X. (Eds.), Particulate Matter, Environmental Monitoring and Mitigation. Future Science Ltd, Ebook, ISBN 978-1-909453-13-5, pp. 74–95. https://doi.org/10.4155/ebo.13.489. Nov. 2013.

Lutz, M., 2018. Successes and challenges of twenty years air quality management in Berlin, Germany. In: Querol, X. (Ed.), Air Quality in Cities. A Global Challenge. Fundación gasNatural Fenosa, ISBN 978-84-09-01905-2. Barcelona, Spain. http://www.fundacionnaturgy.org/en/product/libro-la-calidad-del-aire-las-ciudades-reto-mundial/.

Lutz, M., Rauterberg-Wulff, A., 2009. Ein Jahr Umweltzone Berlin: Wirkungsuntersuchungen. Senatsverwaltung für Gesundheit, Umwelt und Verbraucherschutz, Berlin.

Malina, C., Scheffler, F., 2015. The impact of Low Emission Zones on particulate matter concentration and public health. Transport. Res.: Pol. 77, 372–385.

Morfeld, P., Groneberg, D.A., Spallek, M., 2014a. Effectiveness of low emission zones of stage 1: analysis of the changes in fine dust concentrations (PM10) in 19 German cities. Pneumologie 68, 173–186.

Morfeld, P., Groneberg, D.A., Spallek, M.F., 2014b. Effectiveness of low emission zones: large scale Analysis of changes in environmental NO2, NO and NOx concentrations in 17 German cities. PLoS One 9.

Panteliadis, P., Strak, M., Hoek, G., Weijers, E., van der Zee, S., Dijkema, M., 2014. Implementation of a low emission zone and evaluation of effects on air quality by long-term monitoring. Atmos. Environ. 86, 113–119.

Querol, X., Alastuey, A., Ruiz, C.R., Artiñano, B., Hansson, H.C., Harrison, R.M., Buringh, E., Brink, H.M.t., Lutz, M., Bruckmann, P., Straehl, P., Schneider, J., 2004. Speciation and origin of PM10 and PM2.5 in selected European cities. Atmos. Environ. 38, 6547–6555.

Raaschou-Nielsen, O., Andersen, Z.J., Beelen, R., Samoli, E., Stafoggia, M., Weinmayr, G., Hoffmann, B., Fischer, P., Nieuwenhuijsen, M.J., Brunekreef, B., Xun, W.W., Katsouyanni, K., Dimakopoulou, K., Sommar, J., Forsberg, B., Modig, L., Oudin, A., Oftedal, B., Schwarze, P.E., Nafstad, P., De Faire, U., Pedersen, N.L., Östenson, C.-G., Fratiglioni, L., Penell, J., Korek, M., Pershagen, G., Eriksen, K.T., Sørensen, M., Tjønneland, A., Ellermann, T., Eeftens, M., Peeters, P.H., Meliefste, K., Wang, M., Bueno-de-Mesquita, B., Key, T.J., de Hoogh, K., Concin, H., Nagel, G., Vilier, A., Grioni, S., Krogh, V., Tsai, M.-Y., Ricceri, F., Sacerdote, C., Galassi, C., Migliore, E., Ranzi, A., Cesaroni, G., Badaloni, C., Forastiere, F., Tamayo, I., Amiano, P., Dorronsoro, M., Trichopoulou, A., Bamia, C., Vineis, P., Hoek, G., 2013. Air pollution and lung cancer incidence in 17 European cohorts: prospective analyses from the European Study of Cohorts for Air Pollution Effects (ESCAPE). Lancet Oncol. 14, 813–822.

Regierung von Oberbayern, 2007. Luftreinhalte-/Aktionsplan für die Stadt München, 1. Fortschreibung.

Rückerl, R., Schneider, A., Breitner, S., Cyrys, J., Peters, A., 2011. Health effects of particulate air pollution: a review of epidemiological evidence. Inhal. Toxicol. 23, 555–592.

Sadler, 2011. Low Emission Zones in Europe, for ADEME, Final Report. Sadler Consultants, Emmendingen, Germany.

Santos, F.M., Gómez-Losada, Á., Pires, J.C.M., 2019. Impact of the implementation of Lisbon low emission zone on air quality. J. Hazard Mater. 365, 632–641.

SenGUV, 2011. Ein Jahr Umweltzone Stufe 2 in Berlin: Untersuchungen zur Wirkung auf den Schadstoffausstoß des Straßenverkehrs und die Luftqualität in Berlin (*One year of stage 2 of the low emission zone: Investigation of the impact on the pollutant emissions of road traffic and on the air quality in Berlin)*. Senatsverwaltung für Umwelt, Gesundheit und Verbraucherschutz. Berlin. https://www.berlin.de/sen/uvk/_assets/umwelt/luft/luftreinhaltung/umweltzone/umweltzone_1jahr_stufe2_bericht.pdf. (Accessed 27 August 2021).

Silva, F.N.d., Custódio, R.A.L., Martins, H., 2014. Low emission zone: lisbon's experience. J. Traffic Logist. Eng. 2, 133–139.

Stadt München, 2012. Münchner Statistik, 3. Quartalsheft, Jahrgang 2012: 2012/3 - Grün, Gelb oder Rot? - Ab Oktober 2012 sind nur noch grüne Umweltplaketten in der Münchner Umweltzone erlaubt. City of Munich, 2012/2 - Pkw-Neuzulassungen in

Münch. https://www.muenchen.de/rathaus/dam/jcr:fe5baecc-35d4-44a8-b210-552b8148031c/mb120303.pdf. (Accessed 27 August 2021).
Tartakovsky, D., Kordova – Biezuner, L., Berlin, E., Broday, D.M., 2020. Air quality impacts of the low emission zone policy in Haifa. Atmos. Environ. 232, 117472.
Thorpe, A., Harrison, R.M., 2008. Sources and properties of non-exhaust particulate matter from road traffic: a review. Sci. Total Environ. 400, 270–282.
UBA, 2017. Air Quality 2017 - Preliminary Evaluation. Umweltbundesamt, Dessau-Roßlau, Germany.
US EPA, 2010. Quantitative Health Risk Assessment for Particulate Matter, pp. 1–596. https://www3.epa.gov/ttn/naaqs/standards/pm/data/PM_RA_FINAL_June_2010.pdf.
US EPA, 2016. Integrated Science Assessment for Oxides of Nitrogen – Health Criteria. https://cfpub.epa.gov/ncea/isa/recordisplay.cfm?deid=310879.
VDI 2465, 2016. Part 2, Measurement of Soot (Ambient Air) Thermographic Determination of Elemental Carbon after Thermal Desorption of Organic Carbon. Verein Deutscher Ingenieure e.V., Düsseldorf.
Viana, M., Kuhlbusch, T.A.J., Querol, X., Alastuey, A., Harrison, R.M., Hopke, P.K., Winiwarter, W., Vallius, M., Szidat, S., Prévôt, A.S.H., Hueglin, C., Bloemen, H., Wåhlin, P., Vecchi, R., Miranda, A.I., Kasper-Giebl, A., Maenhaut, W., Hitzenberger, R., 2008. Source apportionment of particulate matter in Europe: a review of methods and results. J. Aerosol Sci. 39, 827–849.
Watkiss, P., Allen, J., Anderson, S., Beevers, S., Browne, M., Carslaw, D., Emerson, P., Fairclough, P., Francsics, J., Freeman, D., Haydock, H., Hidri, S., Hitchcock, G., Parker, T., Pye, S., Smith, A., Ye, R., Young, T., 2003. London Low Emission Zone Feasibility Study. Phase II. Final Report to the London Low Emission Zone Steering Group.
WHO, 2006. Air Quality Guidelines for Particulate Matter, Ozone, Nitrogen Dioxide and Sulfur Dioxide, Global Update 2005: Summary of Risk Assessment.
WHO, 2013a. Review of Evidence on Health Aspects of Air Pollution – REVIHAAP Project: Final Technical Report. Regional Office for Europe, Copenhagen, Denmark.
WHO, 2013b. Review of Evidence on Health Aspects of Air Pollution. Technical Report. WHO Regional Office for Europe. http://www.euro.who.int/_data/assets/pdf_file/0004/193108/REVIHAAP-Final-technical-report-final-version.pdf?ua=1.
Wichmann, H.E., 2018. Gesundheitliche Risiken von Stickstoffdioxid im Vergleich zu Feinstaub und anderen verkehrsabhängigen Luftschadstoffen. Umweltmedizin – Hygiene – Arbeitsmedizin 23, 57–71.