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## A tale of two cities: is air pollution improving in Paris and London?☆

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## ABSTRACT

Paris and London are Europe's two megacities and both experience poor air quality with systemic breaches of the NO<sub>2</sub> limit value. Policy initiatives have been taken to address this: some European-wide (e.g. Euro emission standards); others local (e.g. Low Emission Zone, LEZ). Trends in NO<sub>x</sub>, NO<sub>2</sub> and particulate matter (PM<sub>10</sub>, PM<sub>2.5</sub>) for 2005–2016 in background and roadside locations; and trends in traffic increments were calculated in both cities to address their impact. Trends in traffic counts and the distribution in Euro standards for diesel vehicles were also evaluated. Linear-mixed effect models were built to determine the main determinants of traffic concentrations. There was an overall increase in roadside NO<sub>2</sub> in 2005–2009 in both cities followed by a decrease of ~5% year<sup>-1</sup> from 2010. Downward trends were associated with the introduction of Euro V heavy vehicles. Despite NO<sub>2</sub> decreasing, at current rates, roads will need 20 (Paris) and 193 years (London) to achieve the European Limit Value (40 µg m<sup>-3</sup> annual mean). Euro 5 light diesel vehicles were associated with the decrease in roadside PM<sub>10</sub>. An increase in motorcycles in London since 2010 contributed to the lack of significant trend in PM<sub>2.5</sub> roadside increment in 2010–16.

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## 1. Introduction

Paris and London are Europe's two megacities with more than 10 million people living in each. Both cities experience poor air quality. They currently exceed the European Limit Value for nitrogen dioxide (NO<sub>2</sub>) and particulate matter (PM) episodes are frequent during winter and early spring (Bessagnet et al., 2005; Favez et al., 2012; Rouil et al., 2015; Macintyre et al., 2016; Petit et al., 2017). Long-term exposure to poor air quality has been associated with premature deaths estimated to be ~55,000 annually in France and ~50,000 in the United Kingdom (Guerreiro et al., 2016).

Road transport accounts for a large proportion of primary emissions of nitrogen oxides (NO<sub>x</sub>) and PM<sub>10</sub> in both cities as estimated by inventories: 73% (NO<sub>x</sub>) and 42% (PM<sub>10</sub>) in Paris; 61% (NO<sub>x</sub>) and 28% (PM<sub>10</sub>) in Île-de-France (Airparif, 2016); and 50% for

both pollutants in London (GLA, 2017). Diesel engines emit NO<sub>x</sub> in the form of nitrogen monoxide (NO) and nitrogen dioxide (NO<sub>2</sub>). NO is rapidly oxidised to NO<sub>2</sub> in the atmosphere through its reaction with ozone leading to large concentrations near to roads and ultimately in urban areas. Incomplete combustion in diesel engines emit fine particles (particles with <2.5 µm). Particle emissions are also emitted by non-exhaust sources such as resuspension, tyre-wear and brake-wear, which represent an important fraction of coarse PM (particles with >2.5 µm) on roads (Amato et al., 2016; Font and Fuller, 2016).

Targeted policies to reduce vehicle emissions have been implemented at multiple scales. Some of these are European-wide such as the introduction of the Euro emission standards on new vehicles. Euro norms were introduced in 1991 and new stages were defined over time with more stringent exhaust emission standards for new vehicles. However, real-world emissions in new cars did not always align with expected type-approval tests. While test-cycle NO<sub>x</sub> emissions decreased by 80% since 1992, the real-driving emissions from diesel cars have increased ~20% (Carslaw et al., 2016; Weiss et al., 2012). The after-treatment devices to reduce PM emissions in diesel vehicles were proved to increase

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primary NO<sub>2</sub> emissions since the introduction of Euro 3/III diesel vehicles, representing around 50% of the on-road emissions of NO<sub>x</sub> (Carslaw et al., 2016; Grice et al., 2009; Weiss et al., 2012); and NO<sub>2</sub> emissions in Euro 6a were still higher than those in Euro 2 (Carslaw et al., 2016). Despite the effective reduction of on-road NO<sub>x</sub> emissions from Euro 6 diesel cars (Weiss et al., 2012), early Euro 6 diesel cars showed large variability in urban performance and exceeded the NO<sub>x</sub> standard limit (Degraeuwe and Weiss, 2017; O'Driscoll et al., 2016; Weiss et al., 2012). The new Euro 6d-temp standard for cars includes a real-world driving limit for NO<sub>x</sub> for the first time. In contrast to problems with diesels, on-road tests for gasoline Euro 3–5 light good vehicles (LGVs) were within emissions limits (Weiss et al., 2011).

Other policies to reduce air pollutant concentrations include the introduction of Low Emission Zones (LEZs). The LEZ in London came into operation in February 2008 (for heavy good vehicles of more than 12 tonnes in phase one) and July 2008 (for all other heavy good vehicles in phase two) setting a minimum Euro III PM emission standard for these vehicles. Third and fourth phases were introduced in January 2012 requiring larger vans, minibuses and other specialist vehicles to meet Euro III standards for PM; and heavy good vehicles standards were tightened to Euro IV. Euro III HGVs fitted with an approved filter or converted to an approved gas engine were allowed to continue to enter the LEZ. Buses operated by Transport for London (TfL) had to meet Euro IV standard from December 2015. A LEZ based on Euro norm vehicle classification was introduced in Paris in September 2015 banning the circulation of Euro II heavy good vehicles, coaches and buses in inner Paris. A second phase was introduced in July 2016 with the interdiction of pre-Euro 2 cars and motorcycles. A third phase introduced in July 2017 banning Euro 2 diesel cars and Euro III heavy good vehicles. All these restrictions are limited to weekdays from 8 a.m. to 8 p.m.

Other local measures include the introduction of emergency measures during pollution episodes in Paris (consisting in speed restrictions; banning heavy duty vehicles in inner Paris; free or reduced cost public transportation; free residential parking; alternate number plate traffic circulation until 2016 and differentiated circulation based on vehicle classification since 2017; and also restrictions on wood burning and industrial emissions); and the conversion to a pedestrian street of "Voie Georges-Pompidou" in September 2016, that previously carried 43,000 vehicles every day. Measures in London include the TfL retrofitting program of Euro III buses with Selective Catalytic Reduction (SCR) in specific routes in London; and the introduction of the Low Emission Bus Zone in 2017.

With all these policies being implemented simultaneously it is very difficult to isolate their individual response and evaluate their effectiveness. To overcome this, we evaluated trends in air pollution concentration in Paris and London seeking areas where the policy mix was effective and areas where it was not. Linear trends using the Theil-Sen estimator were calculated from 2005 to 2016 split in two: 2005–09; and 2010–16 for consistency with Font and Fuller (2016). These dates approximate the introduction of tighter Euro standards for new cars and LGVs - Euro 4 (January 2006) and Euro 5 (January 2011); 2010 is the mid-point. Trends in concentrations in urban background and traffic sites were compared to establish whether changes were driven by the regional component or by traffic sources. Trends in roadside increments were also calculated to better evaluate the effectiveness of traffic-related measures in reducing air pollutants concentrations. Linear-mixed effect models was applied to annual mean roadside increments to determine the main traffic variables driving ambient trends and to rank them as a function of their importance as modulators of the air pollution concentrations.

## 2. Methods

### 2.1. Air quality data

Hourly measurements of nitrogen oxides (NO<sub>x</sub>), nitrogen dioxide (NO<sub>2</sub>), particulate matter with aerodynamic diameter of less than 10 µm and 2.5 µm (PM<sub>10</sub> and PM<sub>2.5</sub>) were obtained from Airparif (Paris); and the London Air Quality Network (LAQN) and Defra's Automatic Urban and Rural Network (AURN) (London) from 2005 to 2016. This data set comprised a total of 44 monitoring sites across the Île-de-France region: 30 background locations and 14 roadside sites; and 130 monitoring sites in Greater London with 51 background and 79 roadside sites. Note that not all locations measured all pollutants (Supplementary Table 1, Supplementary Table 2). The distance to Paris or London's city centres was calculated for each monitoring site setting the centre of Paris at the "Point zéro des routes de France" located in the parvis of Notre-Dame (48°51'12.25"N, 2°20'55.63"E); and at Charing Cross in London (51°30'28.8"N, 0°7'30"W).

NO<sub>x</sub> (NO + NO<sub>2</sub>) was measured in both cities by chemiluminescence and regular calibrations enabled the traceability of measurements to national metrological standards. In Paris, PM was measured by TEOM (Tapered Element Oscillating Microbalance) corrected by TEOM-FDMS (Tapered Element Oscillating Microbalance - Filter Dynamics Measurement System) from 2005 to 2011. From 2012 onwards PM was measured by TEOM-FDMS which is considered equivalent to the EU reference method based on 24-h sampling and gravimetric analysis. PM<sub>10</sub> in London was measured by either TEOM-FDMS, by TEOM and or by MetOne BAM (Beta Attenuation Monitor). PM<sub>10</sub> measurements by TEOM-FDMS were reference equivalent. PM<sub>10</sub> concentrations by TEOM were converted to reference equivalent using the Volatile Correction Model (VCM) (Green et al., 2009). PM<sub>10</sub> measurements by BAM were corrected to EU reference equivalent using a factor of 1/1.2 (DEFRA, 2010). PM<sub>2.5</sub> measurements were measured by TEOM-FDMS in both cities. In Paris, the Airparif laboratory and instrumentation were certificated by the French accreditation committee (COFRAC) for calibration and testing activities (European norm: ISO/CEI 17025). In London, all instruments were subject to twice yearly audit tests by the National Physical Laboratory or Ricardo AEA (London) which hold UKAS and ISO/CEI 17025 certifications.

### 2.2. Traffic data

Traffic data from Paris was provided by the Paris City Hall (Paris, 2017) and comprised average vehicle flow from 7 a.m. to 9 p.m. Monday to Friday and the ratio of vehicles between weekends and weekdays working hours. Annual Average Daily Flows (AADF, i.e. average number of vehicles per day) for roads in inner Paris and on the ring-road were calculated from the data provided. The proportion of each vehicle category in the fleet was available for given years (2005, 2010, 2012 and 2014) from Airparif based on the national inventory (CITEPA, 2017) corrected by local studies.

AADFs were available from the Department for Transport (DfT) for roads in London for different vehicle categories: cars & taxis, motorcycles, buses & coaches, light good vehicles (LGVs) and heavy goods vehicles (HGVs). Traffic data was extracted for the nearest traffic counter (<1 km) from each monitoring site located on the same road. A total of 79 traffic counts locations were used. The proportion of each vehicle category for London was calculated from the 79 roads for 2005, 2010, 2012 and 2014 (the same years as data was available for Paris).

### 2.3. Trend calculations

Linear trends in air pollutants concentrations, roadside increments and traffic counts were calculated for two periods of time: 1 January 2005 to 31 December 2009; and 1 January 2010 to 31 December 2016.

The methodology to calculate trends was identical to Font and Fuller (2016) and a summary can be found in the Supplementary Information. Trends were expressed in absolute values ( $\mu\text{g m}^{-3} \text{ year}^{-1}$ ) and as a percentage ( $\% \text{ year}^{-1}$ ). Trends in traffic increments ( $\text{incNO}_x$ ,  $\text{incNO}_2$ ,  $\text{incPM}_{10}$  and  $\text{incPM}_{2.5}$ ) were calculated by subtracting a background concentration from each hourly roadside measurement (“Lenschow approach”) (Lenschow, 2001). Note that the approach may not fully apportion the impact of traffic emissions to background concentrations. However, the formation of secondary air pollution generally takes place at time scales longer than hourly and outside the city (Abdalmogith et al., 2006). At the urban spatial scale this is not thought to lead to a large bias.

Trends were evaluated for each individual monitoring site in Paris and London and then overall trends were calculated using meta-analysis statistical tools. This is an effective methodology to evaluate the variability of ambient responses to polices across a large urban area, accounting for individual and population-wide variability among roads and other monitored locations as shown previously by Font and Fuller (2016).

Trends in traffic were calculated as the slope resulting from the least-square linear model of AADF per year and were expressed in units of  $\Delta \text{vehicles day}^{-1} \text{ year}^{-1}$ . In London, overall trends for AADF were also calculated by means of the Random Effects (RE) model using the traffic counts near to the monitoring sites.

### 2.4. Linear-mixed effect models

Linear-mixed-effect models were built to identify the main drivers of annual roadside increments concentrations in Paris and London. The annual roadside concentration for each pollutant ( $\text{incNO}_2$ ,  $\text{incNO}_x$ ,  $\text{incPM}_{10}$  and  $\text{incPM}_{2.5}$ ) at each monitoring site in Paris and London between 2005 and 2016 (Dormann et al., 2013) was taken as outcome and a random effect was specified on the monitoring site. All roadside sites in London were considered in the model. For Paris, data availability meant that only “intra-muros” and for “Boulevard Périphérique” mean roadside increments were used. All sites in Paris and London were combined in a single model.

Several model formulations were built using different fixed effects: total traffic flow (AADF); flow for diesel cars and LGVs (named light diesels); HGVs and buses & coaches (heavy vehicles) and motorcycles. Also, the traffic flow for Euro 4 and Euro 5 light diesels; Euro III, Euro IV and Euro V heavy vehicles; and Euro 2 and Euro 3 motorcycles were also included as fixed effects in some of the model formulations. The presence of a Low Emission Zone (LEZ) and city (Paris, London) were also included as categorical fixed effects. Supplementary Table 8 summarizes the model formulations.

The best model for each pollutant was selected based on the Conditional Akaike Information (cAIC); choosing the model with the lowest cAIC value. To test the significance of the selected model, a null model without the fixed effects was built and its cAIC value computed. Ideally, cAIC of the selected model should be lower than that from the null. Also, the likelihood ratio test was done comparing the selected and the null models using the anova test. The model was statistically different from the null model if  $p < 0.05$ . Linear-mixed effect models were computed using the ‘lmer’ function of the lme4/R package (version 1.1–17) (Bates et al., 2014). The 95% confidence intervals for the fixed effects were calculated using the ‘confint’ function from the MASS/R package (version 7.3–50).

Possible collinearity between the explanatory variables was assessed by means of pairwise Pearson correlation values, the conditional number (CN) and the maximum variance inflation factor (VIF). Thresholds of  $r > 0.70$ ,  $\text{CN} > 30$  and  $\text{VIF} > 10$  have been suggested to indicate possible collinearity (see references in Dormann et al., 2013). The majority of explanatory variables showed  $r < 0.70$  with the exception of AADF Motorcycles - AADF Heavy Vehicles ( $r = 0.82$ ); AADF Euro 4 light diesels - AADF Heavy vehicles ( $r = 0.8$ ); and AADF Euro 4 light diesels - AADF Motorcycles ( $r = 0.77$ ) in the  $\text{incPM}_{2.5}$  model; and AADF Euro 5 light diesels - AADF Euro V heavy vehicles ( $r = 0.81$ ); and AADF Euro 4 light diesels - AADF Euro IV heavy vehicles ( $r = 0.76$ ) in the  $\text{PM}_{10}$  model. CN and VIF were all below the 30 and 10 thresholds, indicating no collinearity. The models for  $\text{incPM}_{2.5}$  and  $\text{incPM}_{10}$  were re-run without one of the correlated variables and regression coefficients compared to the base model. Regression coefficients did not show a change of sign. Only the effect of AADF heavy vehicles in the  $\text{incPM}_{2.5}$  model increased when AADF motorcycles were excluded. Details can be found in the Supplementary Information S.4.

## 3. Results

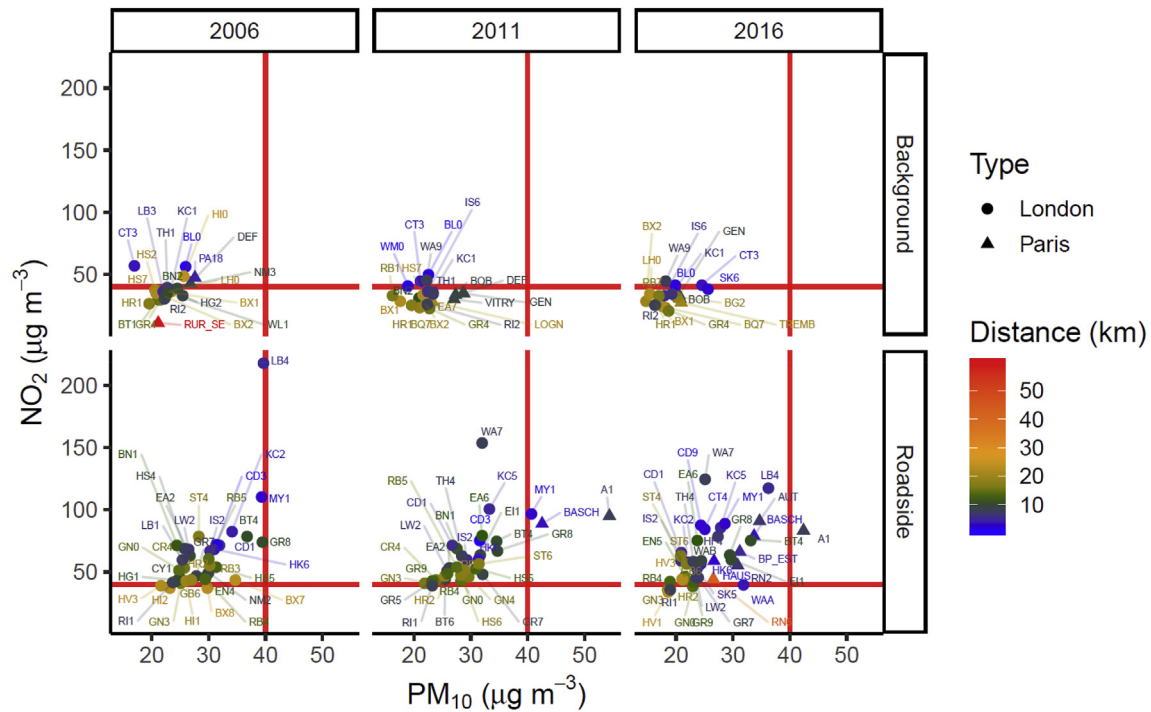
### 3.1. Annual mean concentrations

Throughout our study period, Paris and London measured exceedances of the European Limit Value (ELV) for  $\text{NO}_2$  ( $40 \mu\text{g m}^{-3}$  annual mean) across their networks. This limit value should have been met in 2010. Breaches took place in both background and roadside locations (Fig. 1). However, all background locations in Paris attained the ELV in 2016. That was not the case for London; in 2016 a few central background locations were still above the ELV. Annual  $\text{NO}_2$  concentrations in roadside locations were slightly higher in Paris (median annual  $\text{NO}_2$  across the network ranged from 53 to  $93 \mu\text{g m}^{-3}$  depending on the year) than in London (median across the network:  $55\text{--}59 \mu\text{g m}^{-3}$ ). However, they were quite similar in 2016 (median  $\text{NO}_2$  concentration across the roadside sites in the network was 59 and  $58 \mu\text{g m}^{-3}$  in Paris and London, respectively). Notably some roads in London observed annual mean concentrations more than five times the ELV (i.e. WA7 and LB4) whereas the worst case in Paris was 2.7 times the ELV (i.e. AUT). However, almost all roadside locations in Paris and London exceeded the ELV every year for  $\text{NO}_2$  between 2005 and 2016 (Fig. 1).

$\text{PM}_{10}$  concentrations were below the annual limit value ( $40 \mu\text{g m}^{-3}$ ) at all background locations in both Paris and London. But some roadside locations in Paris still had problems in meeting this limit; those located on the ring-road or next to major roads except for the BASCH site in inner Paris.  $\text{PM}_{10}$  in 2016 was exceeded only at A1 site. Roads in London did not exceed the annual ELV for  $\text{PM}_{10}$  during the study period except for LB4 in 2005 (Fig. 1).

### 3.2. Traffic data: counts and Euro norm distribution

The traffic count on the monitored roads in London was slightly greater ( $\sim 34,000$  to  $31,000 \text{ vehicles day}^{-1}$  from 2005 to 2014) compared with Paris ( $\sim 30,000$  to  $24,000 \text{ vehicles day}^{-1}$ ). It should be noted that the traffic flow for the Parisian ring-road was greater ( $\sim 100,500 \text{ vehicles day}^{-1}$ ) than other roads in Paris (Supplementary Tables 3 and 4). The vehicle fleet in both cities was dominated by cars and taxis representing  $\sim 70\text{--}75\%$  of the fleet; followed by LGVs ( $\sim 15\text{--}16\%$  in Paris and  $12\text{--}15\%$  in London); and HGVs ( $\sim 6\%$  and  $\sim 5\%$ , respectively). The rest of the fleet ( $\sim 7\%$ ) was dominated by motorcycles in Paris whereas it was split almost equally between motorcycles and buses & coaches in London. Notably, the proportion of



**Fig. 1.** Annual mean  $\text{NO}_2$  vs annual  $\text{PM}_{10}$  concentrations at background and roadside locations in 2006, 2011 and 2016 in Paris and London. Annual Limit Values are marked with red lines. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

buses in the Parisian fleet (~0.5–0.6% of the total) was less than one seventh of the share in London.

Diesel cars dominated the Parisian fleet in the whole study period and by 2015 they represented 74% of the cars driven in the city. The share of diesel in London was lower but increased from ~20% in 2005 to 43% in 2015. The number of diesel LGVs was slightly larger in London (95–98%) than Paris (84–91%) (Supplementary Fig. 20).

The distribution in Euro standard for each vehicle category can be found in Fig. 2. In 2005–09 diesel cars in Paris comprised Euro 3 (41–30%), Euro 2 (25–16%) and Euro 1 (18–7%). Euro 4 cars were introduced in 2006 and this was the main Euro class by 2009 representing ~50%. By contrast, for the same period, Euro 3 dominated the diesel car fleet in London (~70% in 2005) and reflecting the growth in the proportion of diesel cars (Supplementary section S3.6) these were slowly replaced by Euro 4 after its implementation in 2006. By 2009 Euro 4 represented 51% of the diesel cars in London. In 2010–16 the distribution of Euro classes in the diesel car fleet was quite similar in both cities, with a dominance of Euro 4 (40–50%); and the replacement of pre-Euro 4 cars by Euro 5. By 2016, Euro 4 and Euro 5 represented 80% of the car fleet in both cities. The distribution of LGVs by Euro class was quite similar to the car distribution in each city.

At the start of the 2005–09 period both Paris and London had similar HGV Euro distribution with Euro II and III dominating the fleet. But at the end of the period the distribution was very different in the two cities. Euro II HGVs were quickly replaced in London's fleet by Euro IV and had almost disappeared by 2008, when the LEZ was introduced. Euro IV was introduced into the fleet of the two cities but reached a higher share in London (49% by 2009) than Paris (30%). By 2009 the presence of Euro II HGVs was still notable in Paris at 20%. By 2010 HGVs in both Paris and London were mainly Euro III (~40%) and IV (40% and 52% in Paris and London, respectively). The introduction of phase 4 of the LEZ in London induced a faster decrease in Euro III compared to Paris at the end of the time

series. Most remaining Euro III HGVs after 2012 in London were adapted to meet Euro IV standards for PM emissions (63–93%) and therefore permitted in the LEZ. Euro V was introduced in 2010 and by 2015 it had become a large part of the fleet, 40% in Paris and 51% in London.

The Euro class distribution of buses & coaches in Paris was similar to that of HGVs for the whole time period. In London, Euro III buses dominated the fleet up to 2012 when Euro V was the main standard. Euro IV was introduced in 2006 and by 2008 reached 40%. Euro II buses represented ~40% in 2005 but were less than 10% from 2008 onwards. Euro VI buses & coaches were introduced in 2013 in Paris, a year before than London; and by 2015 its share was 20% compared to 12% in London.

### 3.3. Trends in air pollutants

Nitrogen oxides and particulate matter in both Paris and London have generally improved with downward trends in both periods of time (Table 1). Both cities had downward trends in background  $\text{NO}_2$  concentrations in 2005–09 ( $-2.1\% \text{ year}^{-1}$  and  $-1.4\% \text{ year}^{-1}$  in Paris and London, respectively) while roadside trends did not show any significant change. In 2010–16  $\text{NO}_2$  concentrations at background sites in London decreased faster ( $-2.1\% \text{ year}^{-1}$ ) than Paris ( $-1.7\% \text{ year}^{-1}$ ). However, roads in Paris and London showed a similar trend within uncertainties ( $-2.9$  [ $-3.4, -2.4$ ] and  $-2.3$  [ $-2.9, -1.8$ ]  $\% \text{ year}^{-1}$ , respectively).

Paris had no significant trend in  $\text{PM}_{10}$  during the first period of time but had a fast downward trend of  $-4.4\% \text{ year}^{-1}$  in both background and roadside locations during the second period. London showed significant downward trends in  $\text{PM}_{10}$  concentrations at background and roadside locations in both periods. However, in the first period background locations observed a faster decrease ( $-3.7\% \text{ year}^{-1}$ ) than roadside sites ( $-3.0\% \text{ year}^{-1}$ ). Downward trends in 2010–16 were similar at both background and roadside ( $-2.9\% \text{ year}^{-1}$  and  $-3.0\% \text{ year}^{-1}$ , respectively).

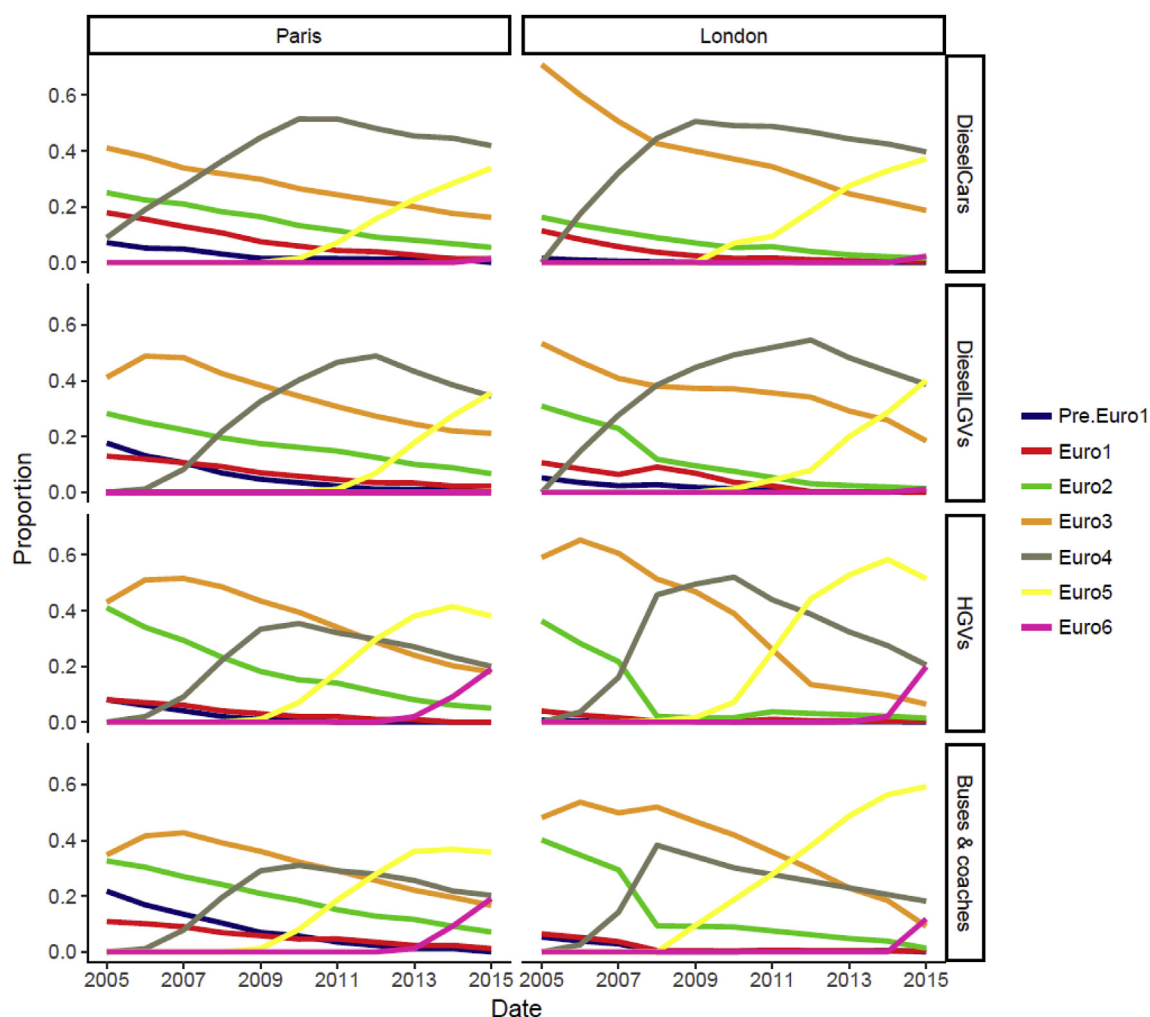


Fig. 2. Distribution of Euro classes for diesel vehicles (cars, LGVs, HGVs and buses & coaches) for Paris and London.

Table 1

Overall percentage trends calculated by means of the random-effects linear model for  $\text{NO}_x$ ,  $\text{NO}_2$ ,  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ , for the periods 2005–2009 and 2010–2016 in London and Paris by station type. Trends in roadside increments are also reported. Brackets denote 95% confidence intervals. \*\*\* significant at the 0.001 level; \*\* significant at the 0.01 level; \* significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant.

City	Pollutant	2005–09			2010–16		
		Background trend (% year <sup>-1</sup> )	Roadside trend (% year <sup>-1</sup> )	Roadside increment (% year <sup>-1</sup> )	Background trend (% year <sup>-1</sup> )	Roadside trend (% year <sup>-1</sup> )	Roadside increment (% year <sup>-1</sup> )
Paris	$\text{NO}_x$	-1.7 [-2.5, -0.9]***	-3.1 [-5.7, -0.5]*	-3.5 [-7.6, 0.6] <sup>+</sup>	-1.5 [-1.9, -1.1]***	-1.9 [-2.5, -1.4]***	-2.7 [-3.5, -1.8]***
	$\text{NO}_2$	-2.1 [-2.82, -1.3]***	-0.5 [-2.4, 1.5]	0.7 [-2.9, 4.2]	-1.7 [-2.0, -1.4]***	-2.9 [-3.4, -2.4]***	-5.5 [-7.0, -4.0]***
	$\text{PM}_{10}$	0.2 [-1.0, 1.5]	-0.03 [-1.8, 1.8]	-0.3 [-2.3, 1.7]	-4.3 [-5.0, -3.6]***	-4.3 [-4.7, -3.9]***	-5.4 [-6.4, -4.3]***
	$\text{PM}_{2.5}$	-4.2 [-7.5, -1.0]*	--	--	-5.2 [-6.2, -4.1]***	-6.6 [-7.3, -5.9]***	-10.5 [-12.6, -8.4]***
London	$\text{NO}_x$	-2.1 [-2.6, -1.5]***	-1.3 [-2.0, -0.7]***	1.1 [0.1, 2.2]*	-1.6 [-2.2, -1.0]***	-1.5 [-2.1, -0.8]***	-1.7 [-2.7, -0.8]***
	$\text{NO}_2$	-1.4 [-2.1, -0.7]***	-0.2 [-0.9, 0.5]	10.4 [7.9, 12.8]***	-2.1 [-2.7, -1.6]***	-2.3 [-2.9, -1.8]***	-5.0 [-3.1, -6.9]***
	$\text{PM}_{10}$	-3.7 [-4.7, -2.7]***	-3.0 [-3.6, -2.4]***	-3.0 [-5.9, -0.1]*	-2.9 [-3.9, -2.0]***	-3.0 [-3.6, -2.4]***	-8.4 [-4.4, 12.5]***
	$\text{PM}_{2.5}$	--	-5.1 [-8.6, -1.6]**	--	-4.4 [-5.3, -3.5]***	-3.7 [-5.2, -2.1]***	-2.6 [-12.5, 7.3]

$\text{PM}_{2.5}$  concentrations at background locations in Paris decreased at  $-4.2\% \text{ year}^{-1}$  in 2005–09 (note that only two sites were available; Supplementary Fig. 6) and faster in 2010–16 at  $-5.2\% \text{ year}^{-1}$ .  $\text{PM}_{2.5}$  along roads was only available in the second period and showed a decrease of  $-6.6\% \text{ year}^{-1}$ , faster than their background counterparts. London had downward trends in both periods. However, downward trends in roadside locations were slower in 2010–16 ( $-3.7\% \text{ year}^{-1}$ ) than in 2005–09 ( $-5.1\% \text{ year}^{-1}$ ). Trends along roads in 2010–16 were slower ( $-3.7\% \text{ year}^{-1}$ ) than those

observed in background ( $-4.4\% \text{ year}^{-1}$ ).

Overall trends in roadside  $\text{NO}_2$  increments changed sign from the 2005–09 period, which had an upward trend in both cities, to a significant downward trend in 2010–16 ( $-6\% \text{ year}^{-1}$  and  $-5\% \text{ year}^{-1}$  in Paris and in London, respectively) (Table 1). Trends in roadside  $\text{PM}_{10}$  increment were flat in 2005–09 in both cities and then showed a downward trend from 2010 ( $-5\% \text{ year}^{-1}$  in Paris and  $-8\% \text{ year}^{-1}$  in London). Trends in roadside  $\text{PM}_{2.5}$  increment were only available in the second period. Paris observed a

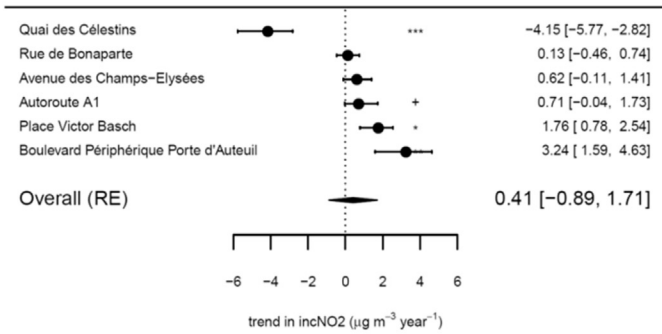
significant downward trend ( $-11\% \text{ year}^{-1}$ ) and London showed a downward but non-significant trend ( $-2.6\% \text{ year}^{-1}$ ) (Table 1). It should be highlighted that trends in roadside  $\text{PM}_{2.5}$  increment in Paris was estimated from two monitoring sites.

When looking at the behaviour of individual roads, we can see considerable heterogeneity. Trends in roadside  $\text{incNO}_2$  in Île-de-France showed an increase in 2005–09 at almost all roads except for Quai des Célestins (Fig. 3A) that showed a significant downward trend. This response was due to the change of the layout of the road with a construction of a bicycle lane between the monitoring site and the road. Boulevard Périphérique Porte d’Auteuil observed the

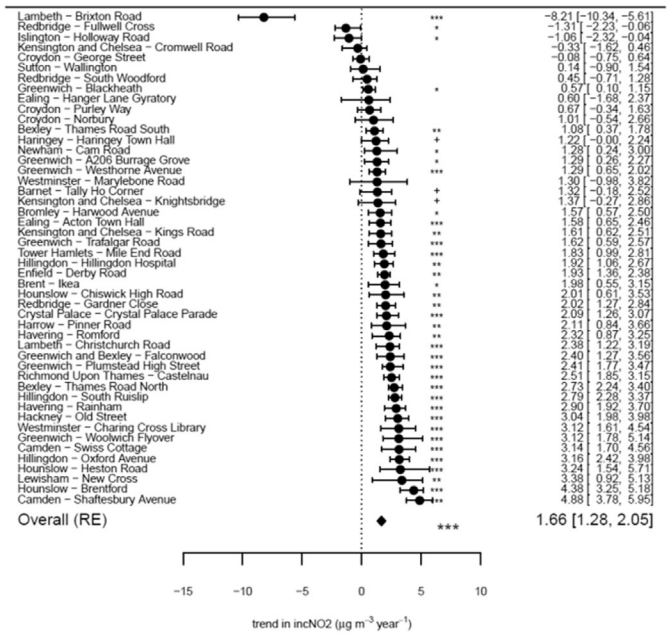
fastest increase ( $3.2 \mu\text{g m}^{-3} \text{ year}^{-1}$ ). In 2010–16 all roads showed a significant downward trend for all pollutants (Fig. 4) although the rate of response varied. The trend in  $\text{incNO}_2$  and  $\text{incPM}_{10}$  in 2010–16 in Boulevard Périphérique Porte d’Auteuil was twice as fast ( $-4.3$  and  $-1.7 \mu\text{g m}^{-3} \text{ year}^{-1}$ , respectively) as the overall trend ( $-1.9$  and  $-0.9 \mu\text{g m}^{-3} \text{ year}^{-1}$ , respectively) (Fig. 4A, C).

Compared with Paris, London roads had a greater variability of responses for all pollutants in the two periods of time but especially in 2010–16. Despite the overall downward trend in  $\text{incNO}_2$  and  $\text{incPM}_{10}$  in 2010–16, there were some roads with significant upward trends (Fig. 4B, D). Trends in  $\text{incPM}_{2.5}$  also had a variability of

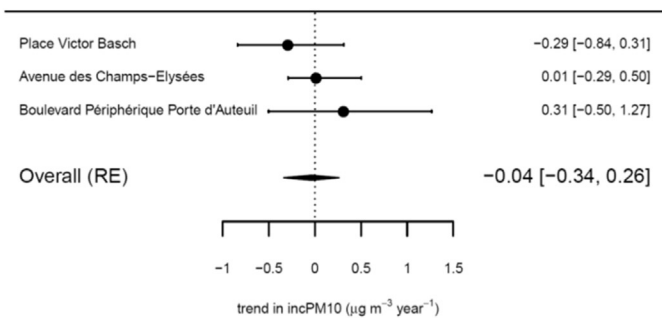
**A. Paris trends in  $\text{incNO}_2$  2005-09**



**B. London trends in  $\text{incNO}_2$  2005-09**



**C. Paris trends in  $\text{incPM}_{10}$  2005-09**



**D. London trends in  $\text{incPM}_{10}$  2005-09**

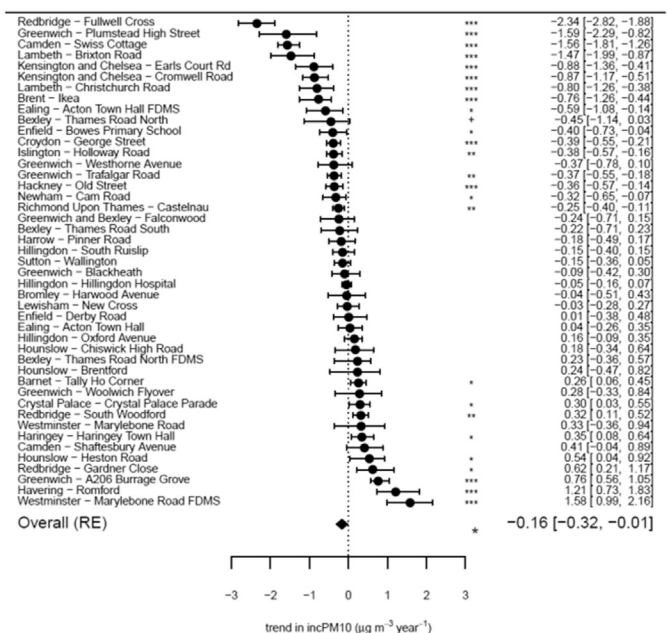
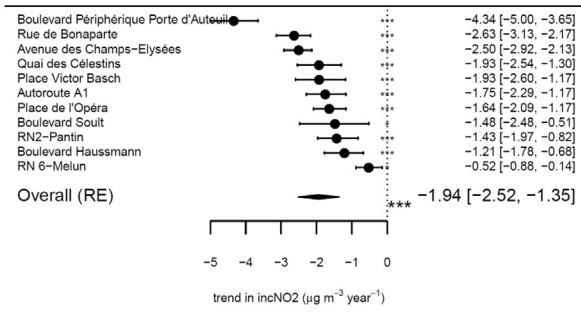
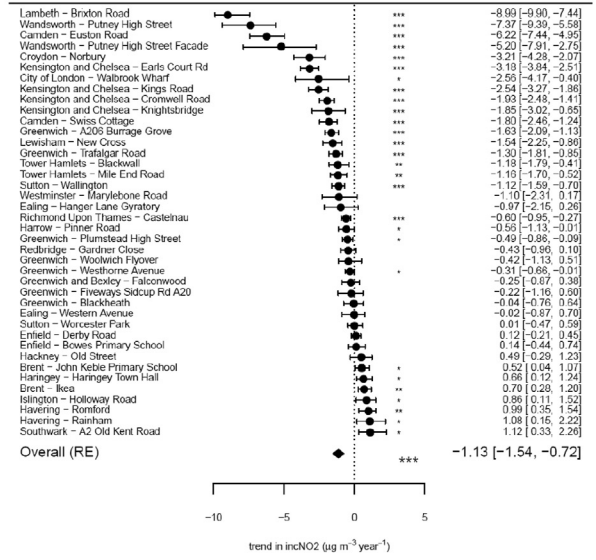


Fig. 3. Forest plots for the trends in roadside increments in  $\text{NO}_2$  ( $\text{incNO}_2$ ) and  $\text{PM}_{10}$  ( $\text{incPM}_{10}$ ) for Paris and London for 2005–09.

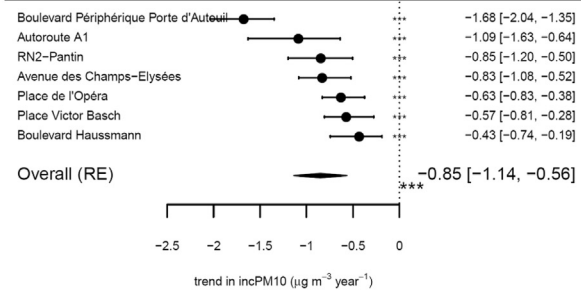
**A. Paris trends in incNO<sub>2</sub> 2010-16**



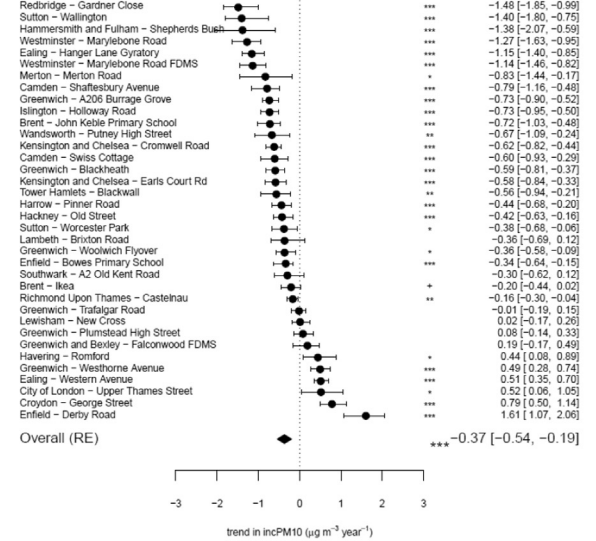
**B. London trends in incNO<sub>2</sub> 2010-16**



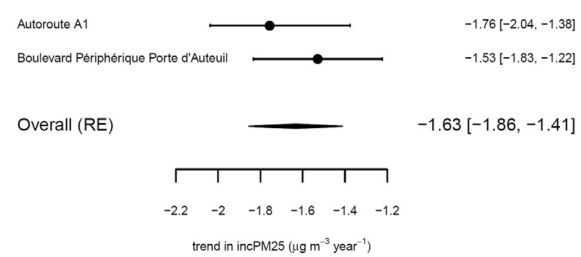
**C. Paris trends in incPM<sub>10</sub> 2010-16**



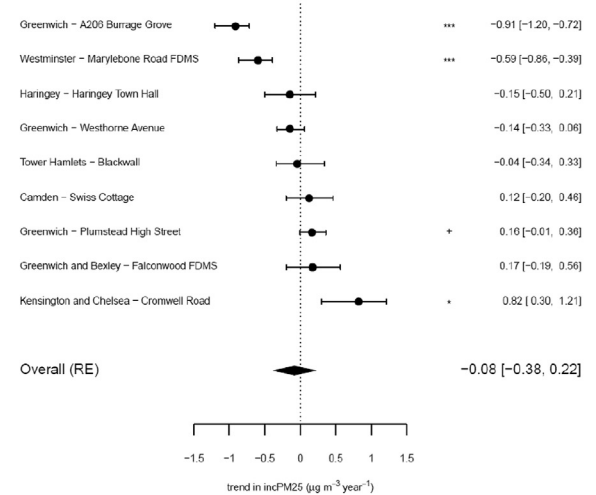
**D. London trends in incPM<sub>10</sub> 2010-16**



**E. Paris trends in incPM<sub>2.5</sub> 2010-16**



**F. London trends in incPM<sub>2.5</sub> 2010-16**



**Fig. 4.** Forest plots for the trends in roadside increments in NO<sub>2</sub> (incNO<sub>2</sub>), PM<sub>10</sub> (incPM<sub>10</sub>), and PM<sub>2.5</sub> (incPM<sub>2.5</sub>) for Paris and London for 2010–16.

response; some roads had fast downward trends (Greenwich – A206 Burrage Grove and Westminster-Marylebone Road) and others had significant upward trends (e.g. Kensington and Chelsea – Cromwell Road) (Fig. 4F).

### 3.4. Traffic: counts and trends

The total number of vehicles decreased during the study period in the two cities. Paris observed similar rates in both periods of time (around -2% year<sup>-1</sup>) while London observed a faster decrease in 2005–09 (-1.4% year<sup>-1</sup>) than in 2010–16 (-0.2% year<sup>-1</sup>) (Supplementary Table 5). Trends on the ring-road in Paris showed a different behaviour, with a non-significant upward trend in the 2005–09 and a modest downward trend in 2010–16 (-0.3% year<sup>-1</sup>, significant at *p* < 0.1) (Supplementary Table 6).

Only London had data to calculate trends in each vehicle category. Here, the downward trend in the total number of vehicles was largely explained by a downward trend in the number of cars & taxis (-1.5 and -0.3% year<sup>-1</sup> in the first and second period, respectively) and a change in the number of HGVs (-0.8 and -0.6% year<sup>-1</sup>). The number of LGVs increased in both periods but not at a significant rate. Buses & coaches increased in 2005–09 (1.7% year<sup>-1</sup>) but decreased after 2010 (-0.6% year<sup>-1</sup>). Motorcycles increased in both time periods but remarkably in the second period at a rate of 11% year<sup>-1</sup> (Supplementary Table 7).

### 3.5. Influence of traffic variables to roadside increment concentrations

Table 2 indicates the selected linear-mixed effect model for each pollutant and the statistical parameters. The cAIC values and the R<sup>2</sup> for each model including the null model can be found in the Supplementary Table 9; and the formulation for the selected model for each pollutant is shown in Supplementary Table 10.

The traffic parameters with greatest impact on roadside NO<sub>2</sub> increments were AADF Euro 4 heavy vehicles (2.9 μg m<sup>-3</sup> every 1000 vehicles) > AADF motorcycles (1.8 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>) > AADF Euro 3 heavy vehicles (1.7 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>) > AADF light diesel vehicles (0.6 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>). AADF Euro 5 heavy good vehicles lead to a decrease on incNO<sub>2</sub> levels at a rate of -4.1 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>.

The linear-mixed effect model for incNO<sub>x</sub> did not separate the different Euro norms for heavy vehicles but did it for light diesels. Also, the AADF of motorcycles and heavy vehicles were associated with an increase in incNO<sub>x</sub> (4.8 and 3.5 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>) and AADF Euro 5 light diesels was associated with a decrease (-2.4 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>).

Roadside increments in PM<sub>10</sub> were influenced by AADF Euro 3

and 5 heavy vehicles; and Euro 2 motorcycles (0.7, 1.2 and 3.5 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>, respectively) whereas AADF Euro 5 light diesels was associated with a decrease in incPM<sub>10</sub> (-0.4 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>). Roadside PM<sub>2.5</sub> increments only showed two traffic parameters with significant coefficients: AADF motorcycles (1.2 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>) and AADF Euro 5 light diesels (-0.3 μg m<sup>-3</sup> 1000 vehicles<sup>-1</sup>).

## 4. Discussion

### 4.1. Nitrogen dioxide (NO<sub>2</sub>)

#### 4.1.1. Trends in roadside increments in 2005–09

Roadside locations in Paris and in London did not benefit from the downward trends observed in background locations in 2005–09. Trends in incNO<sub>2</sub> showed an upward trend in both cities (the lack of statistically significant trend in Paris was due to the fast-downward trend due to the road layout changes at Quai des Célestins site. When this is removed from the analysis, the overall trend in incNO<sub>2</sub> was an increase of 2.5% year<sup>-1</sup>\*\*).

Counterintuitively, the increase in incNO<sub>2</sub> took place when the number of vehicles in the urban centres decreased fastest, around -2.1% year<sup>-1</sup> in Paris and -1.5% year<sup>-1</sup> in London. However, the proportion of diesel vehicles increased: from 56% (2005) to 74% (2009) of cars in Paris; and 20%–33% of in London (Supplementary Fig. 20). The linear-mixed effect model indicated that AADF of light diesel vehicles (cars + LGVs) was associated with an increase in incNO<sub>2</sub>. The mean number of light diesel vehicles along roads in Paris and London increased from ~12,200 in 2005 to ~15,200 in 2009, representing an increase of 1.8 μg m<sup>-3</sup> in incNO<sub>2</sub>. The other traffic parameters associated with higher incNO<sub>2</sub> with significant change between 2005 and 2009 were Euro IV heavy vehicles, that were introduced in the fleet with a mean of 1200 vehicles by 2009, adding ~3.5 μg m<sup>-3</sup>.

In 2005–09, Euro 4 cars and LGVs were introduced into the fleet of the two cities and was the main standard by 2009 (Fig. 2). Despite the stringent emission standards for NO<sub>x</sub> (the pollutant regulated in vehicle performance tests) in Euro 4 diesel cars and LGVs, evidence has shown that under real driving conditions emissions did not align with improved performance in approval tests (Carslaw and Rhys-Tyler, 2013; Carslaw et al., 2016; Sjödin et al., 2017). NO emitted in urban areas would have been transformed to NO<sub>2</sub> by O<sub>3</sub> reaction. Furthermore, there is evidence that primary NO<sub>2</sub> emissions from light diesel vehicles measured in London increased from 16% (Euro 3) to 28% (Euro 4) of total NO<sub>x</sub> (Carslaw et al., 2016). Slightly higher figures were reported in Göteborg for Euro 4 (~35%) (Sjödin et al., 2017). The high NO<sub>2</sub> emissions from Euro 4 were due to the vehicle after-treatment

**Table 2** Statistical parameters (intercept and regression coefficients) of the selected linear-mixed effect models for each pollutant. Values denote the 95% confidence interval. Units for fixed effects are expressed per 1000 vehicles. Bold numbers indicate significant at 95% level.

	incNO <sub>x</sub>	incNO <sub>2</sub>	incPM <sub>10</sub>	incPM <sub>2.5</sub>
Best model formulation	#5	#6	#8	#5
Intercept	93.8 [71.5, 116.5]	17.4 [8.9, 25.8]	4.2 [2.8, 5.7]	1.2 [-0.5, 2.8]
AADF light diesels		<b>0.6 [0.3, 1.0]</b>		
AADF heavy vehicles	<b>3.5 [1.1, 5.8]</b>			0.4 [-0.1, 1.0]
AADF motorcycles	<b>4.8 [1.5, 8.3]</b>	<b>1.8 [0.2, 3.3]</b>		<b>1.2 [0.9, 1.6]</b>
AADF Euro 4 light diesels	0.3 [-0.2, 0.8]		0.1 [-0.03, 0.3]	0.0 [-0.2, 0.1]
AADF Euro 5 light diesels	<b>-2.4 [-3.0, -1.9]</b>		<b>-0.4 [-0.5, -0.2]</b>	<b>-0.3 [-0.5, -0.2]</b>
AADF Euro III heavy vehicles		<b>1.7 [0.1, 3.3]</b>	<b>0.7 [0.1, 1.2]</b>	
AADF Euro IV heavy vehicles		<b>2.9 [1.6, 4.3]</b>	-0.5 [-1.3, 0.3]	
AADF Euro V heavy vehicles		<b>-4.1 [-5.6, -2.6]</b>	<b>1.2 [0.6, 1.8]</b>	
AADF Euro 2 motorcycles			<b>3.5 [1.9, 5.1]</b>	
AADF Euro 3 motorcycles			0.2 [-0.6, 0.9]	



technologies to reduce carbon monoxide and hydrocarbons; and to aid control of PM diesel emissions (Carslaw et al., 2016). Conversely, on-road emissions from Euro IV heavy vehicles showed a reduction in the primary NO<sub>2</sub> fraction (Carslaw et al., 2016) despite an increase in NO<sub>x</sub> emissions (Carslaw and Rhys-Tyler, 2013; Sjödin et al., 2017).

The greater increase in incNO<sub>2</sub> in 2005–09 observed on the Paris ring-road (Boulevard Périphérique Porte d'Auteuil) compared to the inner roads in Paris, and based on the results of the linear mixed-effect model for incNO<sub>2</sub>, is explained by the larger increase in light diesel vehicles (14% compared with 5.5%, respectively) and an increase of ~2300 Euro IV heavy vehicles compared with ~600 in inner Paris.

#### 4.1.2. Trends in roadside increments 2010–16

In contrast to 2005–09, there were clear downward trends in incNO<sub>2</sub> in 2010–16 in both cities at 5–6% year<sup>-1</sup>. The linear-mixed effect model indicated that Euro V heavy vehicles were responsible for reductions in incNO<sub>2</sub>. This agrees with real-world emission tests from Euro V HGVs that show a reduction of 22–85% in primary NO<sub>2</sub> emissions compared with Euro II/III standards (Carslaw et al., 2016; Sjödin et al., 2017). By 2015, Euro V dominated the HGV and bus & coach fleets in both cities (Fig. 2). Therefore, it is likely that the downward trends in incNO<sub>2</sub> concentrations in 2010–16 in the two cities reflected the reduction of NO<sub>2</sub> emissions from Euro V heavy vehicles. Another factor that might have hastened the decrease in incNO<sub>2</sub> in 2010–16 is the increase of light diesel vehicles, most likely explained by the negative coefficient in incNO<sub>x</sub> (Table 2).

There was a faster downward trend in incNO<sub>2</sub> on the ring-road site compared with inner Paris roads. This was consistent with the greater increase of Euro V heavy vehicles (380%) on the ring-road compared with inner roads (332%).

London had a faster HGV fleet turnover than Paris due to the introduction of the LEZ in 2008, as seen in Fig. 2 and as shown separately by Ellison et al. (2013). Pre-Euro III HGVs were removed from the fleet and replaced by Euro IV (up to 2010) and then by Euro V. The tightening of the LEZ in 2012 led to the remaining Euro III HGVs being retro-fitted to meet Euro IV PM standards (e.g. fitting a diesel particle filter - DPF). However, the LEZ did not produce faster reductions in incNO<sub>2</sub> concentrations when compared with Paris. The LEZ in London was designed to reduce PM emissions but was also predicted to reduce NO<sub>x</sub> concentrations in London by 18% (Cloke et al., 2000) based on assumed real-world emissions declining in line with Euro standards. The linear-mixed effect model suggests that reductions in incNO<sub>2</sub> would be achieved tightening standards of heavy vehicles to Euro V.

The Parisian fleet still had Euro II and III HGVs in 2015 accounting for 30% of the HGVs. A LEZ was introduced in Paris in September 2015 banning pre-Euro III HGVs. Based on the London's experience replacing pre-Euro III HGVs is not expected to provide rapid NO<sub>2</sub> reductions if these are solely replaced by Euro IV as shown by the trends in incNO<sub>2</sub> in 2005–09 in London and confirmed by the linear-mixed effect model. The LEZ in Paris also bans diesel cars and LGVs (pre-Euro 3) and motorcycles (pre-Euro 2). However, on-road primary NO<sub>2</sub> from pre-Euro 3 cars and LGVs was less than Euro 4 and 5 (Carslaw et al., 2016). The effectiveness of the LEZ would probably be maximised by tightening the policy to also exclude Euro 4 and/or Euro 5 LGVs; and Euro pre-V HGVs.

#### 4.1.3. Future directions and considerations

Despite the recent downward trends, both Paris and London are still a long way away from attaining the NO<sub>2</sub> ELV which was set for 2010. To place current rate of progress in context, based on trends between 2010 and 2016, roads in Paris will still need between 4 and 20 years (average of 10) to attain the ELV. For London's roads this is

between 2 and 193 years (average of 21). The background sites in London that exceeded might still need 6 years (ranging from 1 to 23 years) to comply at current rates. However, it should be remembered that these calculations do not include new policies such as the introduction of Euro 6 (a-d)/VI in the fleet. Grice et al. (2009) estimated that the annual mean concentration of NO<sub>2</sub> in London is expected to remain above the limit value until 2020 or beyond even with new Euro VI standards to address the emissions from heavy duty vehicles. Despite tightening NO<sub>x</sub> emission limits on new Euro 6 vehicles and the introduction of the real-driving emissions tests in Euro 6d-temp, the allowed conformity factors will delay the widespread adoption of available emission control technologies (Degrauwe et al., 2017) and alternative measures such as the reduction of diesel cars and move to a gasoline urban fleet would be a more effective route to achieve compliance with the EU standards.

The roads that had faster downward trends in 2010–16 in NO<sub>2</sub> in London were those with the greatest annual mean NO<sub>2</sub> concentration in 2010 (CD9, LB4, WA7 and WA8) (Supplementary Fig. 21). In the case of WA7 and WA8 an intervention program fitting Euro III buses with Selective Catalyst Converters (SCRs) in 2013 successfully reduced roadside NO<sub>x</sub> and NO<sub>2</sub> concentrations by 20% (Barratt and Carslaw, 2014). Despite the success of this intervention, it cannot be repeated (although new intervention programmes can be deployed). The overall downward trend at these locations decreased when the step-change was viewed over the 2010–16 period compared with 2010–14 in Font and Fuller (2016). Further policies will clearly be required to reduce NO<sub>2</sub> ambient levels.

The emission performance of buses & coaches will be particularly relevant for London and especially on those roads where they represent an important fraction of heavy vehicles. TfL buses within the London's LEZ should comply to Euro IV standards from December 2017 (Holman et al., 2015). On-road tests of Euro IV (Liu et al., 2011) and Euro V buses (Zhang et al., 2014) found an improved performance in NO<sub>x</sub> emissions compared to previous emission standards but they still exceeded the regulated test limit. According to our model results, Euro IV heavy vehicles were not associated with a decrease in incNO<sub>2</sub> concentrations either. Zhang et al. (2014) suggested that hybrid and natural gas buses as the alternatives to control NO<sub>x</sub> emissions however others (Howarth, 2014) have suggested large leakages from the use of methane as a road fuel could be harmful for climate change.

It is also important to note that some roads in London that exceed NO<sub>2</sub> ELV concentrations had upward trends in incNO<sub>2</sub> (Fig. 4B). Clearly policies were not being successful along these roads. Except for Islington – Holloway Road (IS2) and Hackney – Old Street (HK6), the majority of these roads are located far from the city centre (>10 km) (Supplementary Fig. 21). In 2010–14, roads with increased incNO<sub>2</sub> were associated with an increase in the number of heavy vehicles (buses and HGVs) (Font and Fuller, 2016).

#### 4.2. Particulate matter (PM<sub>10</sub> and PM<sub>2.5</sub>)

Trends in PM<sub>10</sub> at the background and the roadside in Paris were similar in magnitude for each period; however, trends in roadside increments showed a faster decrease. The same pattern was observed in London. The traffic increment represented on average ~35% and 18% of the total PM<sub>10</sub> concentration measured at the roadside locations in Paris and London, respectively. Therefore, trends at roadside locations would have been largely determined by the downward trends in the urban background. To be effective, the reduction of urban PM<sub>10</sub> concentrations should also include wider urban and regional policies such as tackling urban wood burning and secondary PM precursors, including vehicle emissions from outside the city boundaries.

#### 4.2.1. Trends in roadside increments 2005–09

Policies reducing vehicular PM emissions were successful in London since 2005–09 and in Paris since 2010–16 (Table 2) with downward trends in the traffic increment.

Paris observed flat non-significant trends in 2005–09 while London observed significant downward trends. The linear-mixed effect model suggested that the downward trend in London was associated with the replacement of Euro III HGVs with Euro IV. Although the model for  $\text{incPM}_{10}$  did not result in a statistically significant coefficient for Euro IV HGVs, the central estimate showed a negative coefficient (Table 2). Moreover, less Euro III heavy vehicles was associated with less  $\text{incPM}_{10}$ . The benefit in replacing Euro III HGVs in Paris may have been offset by the large proportion of motorcycles.

Trends in  $\text{incPM}_{2.5}$  were only available at the roadside locations in London. It showed a faster decrease ( $-5.1\% \text{ year}^{-1}$ ) than  $\text{incPM}_{10}$  ( $-3.0\% \text{ year}^{-1}$ ).

#### 4.2.2. Trends in roadside increments 2010–16

Paris and London observed downward trends in  $\text{incPM}_{10}$  in 2010–16. According to the linear-mixed effect model for  $\text{incPM}_{10}$  (Table 2), the introduction of Euro 5 light diesels significantly reduced roadside  $\text{PM}_{10}$  concentrations. This is due to the exhaust diesel particle filters (DPFs) (Fiebig et al., 2014) that were effectively compulsory in Euro 5 cars and LGVs (2011). DPFs in heavy vehicles were first introduced in Euro IV (2009). The slower decrease in  $\text{incPM}_{10}$  in Paris compared to London might be due to the greater proportion of motorcycles that offset the benefits of Euro 5 light diesel vehicles.

Surprisingly, the model gave non-significant factor for Euro IV heavy vehicles and positive factors for Euro V, despite the DPFs. A possible explanation is that the reduction of  $\text{PM}_{10}$  emissions from tail-pipe (in the fine fraction) might have been counteracted by increased non-exhaust particle emissions (resuspension, brake-wear, tyre-wear) (dominant in the coarse fraction). An increase in the  $\text{incPM}_{10}$  from non-exhaust emissions might be influenced by an increase in the traffic flow, especially that of heavy vehicles. An increase in kerbside coarse particle concentrations were observed in a central location in London following a higher number of buses with increased vehicle weight associated with larger resuspension rates (Carslaw et al., 2006). However, heavy vehicles, both HGVs and buses & coaches, showed a decreasing trend in 2010–16 in London (Supplementary Table 7). Despite the overall downward trend in  $\text{incPM}_{10}$  observed across London's roads in 2010–16 (Table 1), there were some suburban roads that had upwards trends (Fig. 4D). With one exception, the eight roads with upwards trends in  $\text{incPM}_{10}$  were suburban (distance from the city centre > 10 km) (Supplementary Fig. 21). The central London site (City of London – Upper Thames Street) was affected by nearby construction dust in 2016. All other central roads measured a decrease. This indicates that the control on vehicular  $\text{PM}_{10}$  emissions did not have the same response everywhere. Unfortunately, most roads with positive  $\text{incPM}_{10}$  trends did not have collocated  $\text{PM}_{2.5}$  measurements (Fig. 4D, F; Supplementary Fig. 23) to confirm if an increase in the coarse fraction was the cause. The plots relating trends in  $\text{incPM}_{10}$  vs trends in AADF for different vehicle categories in London (Supplementary Fig. 24) indicate that sites in outer London with positive trends had an increase in LGVs. That appears to contradict the results from the linear mixed effect model. However, the light diesel vehicle category in the model was not LGV specific and included both cars and LGVs; and also the Euro norm. It might be the case that these locations with upward  $\text{incPM}_{10}$  trends might have older LGVs and the reduced  $\text{PM}_{10}$  traffic concentrations from Euro 5 diesel cars might be offset with the light diesel vehicle count.

Decreasing trends in  $\text{incPM}_{2.5}$  concentrations on the two

Parisian roads with  $\text{PM}_{2.5}$  data were greater than those observed in background locations indicating the success of traffic-related policies (Fig. 4E); however, it was the opposite in London where trends in roadside  $\text{incPM}_{2.5}$  in 2010–16 showed a non-significant downward trend (Table 1). This behaviour contrasted with earlier analysis of 2010–14 when most roads in London observed significant downward trends (Font and Fuller, 2016). Trends in  $\text{incPM}_{2.5}$  were not monotonic in 2010–16 and the consistent downward trend observed in 2010–14 was broken by an increase in the roadside  $\text{PM}_{2.5}$  concentration in 2015–16 (Supplementary Fig. 19). This later increase was more prominent on some roads (e.g. CD1: Camden – Swiss Cottage) than others (e.g. MY7: Westminster – Marylebone Road) resulting in a non-significant downward trend when considering the period 2010–16. Some roads in London had slower trends in  $\text{PM}_{2.5}$  than that at the background (KC1) (Supplementary Fig. 22).

The linear-mixed effect model for  $\text{incPM}_{2.5}$  identified Euro 5 light diesels as the vehicle category associated with reducing roadside  $\text{PM}_{2.5}$ ; and motorcycles with increasing factors (Table 2). The number of motorcycles in London has increased rapidly at a rate close to  $11\% \text{ year}^{-1}$  (Supplementary Table 7). This increase might be associated with the proliferation of motorcycles based deliveries in recent years (e.g. home meal-delivery) or the permanent opening of London's bus lanes to motorcycles in 2011 (TfL, 2011). This is an important issue since Euro standards for motorcycles do not regulate PM emissions (except for quads). Despite small engine size, motorcycles emissions tests carried out by Pham et al. (2013) showed that they emitted more PM than petrol light duty gasoline vehicles reported in Robert et al. (2007) with PM emissions from motorcycles that were much greater even than non-catalyst petrol vehicles (model years 1965–2003) for the same distance. Motorcycles emit significant quantities of primary organic aerosols, aromatic volatile carbon components and also are responsible for the production of secondary organic aerosols (Platt et al., 2014). The poor combustion in motorcycles in terms of air fuel ratio is often responsible for high unburned hydrocarbons and particle emission levels, with submicron particles dominating the exhaust (Yang et al., 2005; Chien and Huang, 2010). The increase in the number of motorcycles in London might therefore have led to an increase in fine particle emissions, offsetting the expected benefits of DPFs on other vehicle classes.

The Parisian fleet has a larger share of motorcycles compared with London. The relationship between motorcycles and  $\text{PM}_{2.5}$  concentrations in London therefore has clear implications for Paris. The two Parisian sites that observed a  $\text{PM}_{2.5}$  decrease in 2010–16 are located on the ring-road and therefore had a lower share of motorcycles than inner-Paris roads, which would be consistent with the evidence here of their large  $\text{PM}_{2.5}$  emissions.

## 5. Conclusions

This paper evaluated recent trends (2005–2016) in traffic related pollutants ( $\text{NO}_2$ ,  $\text{NO}_x$ ,  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ ) in Europe's two megacities, Paris and London. Monitoring in two different cities allowed us to evaluate the success of policies (Euro norm, vehicle traffic flow and Low Emission Zone) in a wider perspective. Linear-mixed effect models provided information of which traffic variables drove the trend in roadside increment concentrations. The main results were:

- Light diesel vehicles, motorcycles and Euro III and IV heavy vehicles were associated with increased traffic  $\text{NO}_2$  concentrations in 2005–09 in both cities.
- The downward trend in traffic  $\text{NO}_2$  concentrations in 2010–16 in both Paris and London roads was associated with the

introduction of Euro V heavy vehicles. The introduction of Euro 5 Euro norm in cars and light good vehicles did not result in a clear decrease in traffic NO<sub>2</sub> in line with the evidence on the real-world emissions tests (dieselgate) (Degraeuwe and Weiss, 2017).

- London observed a greater heterogeneity of behaviour with some roads displaying an upward trend in incNO<sub>2</sub> in 2010–16. Roads with greatest concentrations were improving; those with increasing NO<sub>2</sub> had one to two times the annual limit value in 2010.
- An overall downward trend in traffic increment of PM<sub>10</sub> was observed in London since 2005; and since 2010 in Paris. Notably, this was not observed on all London roads.
- The fast replacement of Euro III HGVs in London, partly induced by the introduction of the LEZ, explained the earlier downward trend in incPM<sub>10</sub> when compared with Paris. The greater proportion of motorcycles in Paris might have offset the benefit of replacing Euro III HGVs.
- Euro 5 light diesel vehicles were responsible for downward trends in incPM<sub>10</sub> since 2010.
- Increment PM<sub>2.5</sub> concentrations had a statically significant downward trend in Paris in 2010–16 which was associated with Euro 5 light diesel vehicles. London did not observe such clear trend due to the increase of the motorcycle flow.

The success of current and future policies relies on the real-world emissions of new vehicles being less than the ones they replace. It is clear from comparing the traffic composition between the two cities that the London's LEZ successfully replaced the intended vehicle types. However, it did not provide the expected impact on ambient pollutant concentrations, especially in NO<sub>2</sub>. The responsibility for the transference of this success into ambient concentrations must therefore rest with the vehicle manufacturers that have previously engineered differences between type-approval tests and real-world emissions. Euro 6/VI vehicles will include on-road testing as part of their standard emission testing and an improvement in ambient air quality in urban areas is therefore expected. However, results from this study highlights that motorcycles may be causing hotspots of air pollution in European cities. A continued increase in the number of motorcycles and the diesel car growth, especially light goods vehicles, could confound policies to reduce NO<sub>2</sub> and PM concentrations.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2019.01.040>.

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