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On-road vehicle emission measurements show a significant reduction of black carbon and nitrogen oxides emissions in Euro6c and 6d diesel-powered cars.

Irena Ježek Brecelj[1], Asta Gregorič[1,2], Lucijan Zgonik[3], Tjaša Rutar[3], Matic Ivančič[1], Balint Alfoldy[1], Griša Močnik[2,3], Martin Rigler[1]

[1]Aerosol d.o.o, Ljubljana, 1000, Slovenia
[2]Center for Atmospheric Research, University of Nova Gorica, Ajdovščina, 5270, Slovenia
[3]School for Environmental Sciences, University of Nova Gorica, 5271, Vipava, Slovenia

Correspondence to: Irena Ježek Brecelj (ibrecelj@aerosolmageesci.com)

Abstract. This study compares the results of three on-road chasing campaigns conducted in 2011, 2017, and 2023, making it the first to report how real-world emission factors (EF) for diesel vehicles have changed over a decade. By directly measuring emissions during real-world driving, this research provides critical insights into the effectiveness of emission control technologies and regulatory interventions. The findings highlight the transformative impact of Diesel Particulate Filters (DPFs), which have consistently reduced black carbon (BC) EF from diesel vehicles to levels comparable to gasoline-powered cars. This underscores the success of DPFs in controlling particulate emissions, a major contributor to air quality issues. Real Driving Emissions (RDE) regulations have also proven effective in significantly lowering nitrogen oxide (NO_x) emissions. By incentivizing the use of previously underutilized technologies, these regulations ensure better compliance with standards during typical driving conditions. Real-world EF measurements are crucial for bridging the gap between ambient pollution data and traffic emission models, enabling more accurate assessments of vehicle fleet contributions to air quality. This study employs an independent chasing method, demonstrating its value as a practical tool for monitoring fleet emissions, identifying super-emitters, and detecting vehicles with defeat devices. Overall, this decade-long analysis highlights the significant advancements in reducing diesel emissions, while emphasizing the importance of sustained efforts in monitoring and regulation to further improve air quality and refine emission modeling frameworks.

1. Introduction

Traffic contributes significantly to air pollution, compelling regulatory bodies to enact measures to reduce vehicle emissions. In the European Union (EU), regulations, such as 459/2012/EC (The European Commission, 2012) and 2016/646/EU (The European Commission, 2016) for passenger cars and light commercial vehicles, mandate compliance standards for all new vehicles. Over the years, the progressive tightening of European emission standards corresponds with the mandatory implementation of exhaust after-treatment systems in vehicles. The Euro standards, which started with Euro 1, in 1992, have first mandated the use of catalytic converters to meet initial carbon monoxide limits. Further reduction of nitrogen oxides

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(NO_x) emissions with Euro 3 (in 2000) necessitated the wider adoption of technologies like exhaust gas recirculation. More advanced systems like diesel particle filters (DPF) and selective catalytic reduction (SCR) became increasingly essential with the introduction of Euro 5 (in 2009) and Euro 6 (in 2014) standards, respectively (like goods vehicles standards denoted by Roman numerals Euro V and Euro VI). A more detailed description of progressive technological advances with respective Euro standards and associated costs can be found in Posada Sanchez et al., 2012.

In 2015, the "dieselgate" scandal shed light on critical issues within European legislation. The scandal underscored the discrepancy between emissions observed in laboratory tests, using the New European Driving Cycle (NEDC) (Economic Commission for Europe of the United Nations, 2011) – the cycle used for all new type approved vehicles, and emissions encountered during everyday driving conditions. The cycle-beating controversy was particularly concerning, wherein vehicle manufacturers manipulated engine performance to regulate emissions solely for regulatory tests. Employing so-called defeat devices, these manufacturers optimized engine control systems to recognize testing conditions and automatically switched to a mode optimized for emissions compliance. The manufacturers defended the use of defeat devices as a measure to prevent clogging and aging. However, the Court of Justice of the European Union ruling in case C-693/18 ruled that "only immediate risks of damage which create a specific hazard when the vehicle is driven… justify the use of a defeat device" (Court of Justice of the European Union, 2020).

In the wake of the dieselgate scandal, the EU and its member states have recalled 8.5 million cars in the Union. This initiative was aimed at repairing the affected vehicles. The rate of repair reported by the Volkswagen group reached 79.7% by July 2018 and they offered technical service for the affected vehicles free of charge by the end of 2020. However, Volkswagen would not offer a full and clear guarantee against potential problems post-repair (The European Commission, Dieselgate, 2021).

The ensuing scrutiny prompted regulatory reforms, notably the introduction of the Euro 6c standard (2018), which incorporated a more realistic driving cycle – the Worldwide-harmonized Light-duty Test Cycle (WLTC) (The European Commission, 2017) and mandated the use of a portable emissions measurement system (PEMS) in real driving emissions (RDE) tests alongside laboratory tests in conformity assessments.

Nevertheless, the response from authorities seems insufficient. Many of the inadequate vehicles, now between 9 to 14 years old, are still in use. An analysis by the International Council on Clean Transport (ICCT) involved 1,400 tests on Euro 5 and Euro 6 diesel cars made before RDE testing was implemented (Meyer et al., 2023). This study was carried out under controlled conditions by government bodies and revealed worrisome findings: "suspicious" NO_x emission levels were detected in 77% to 100% of tests and vehicles tested, while "extreme" NO_x emissions occurred in 40% to 75% of cases. These results respectively suggest the likely and almost certain use of prohibited defeat devices in Euro 5 and pre-RDE Euro 6 diesel cars.

The study by Giechaskiel et al. (2022) confirmed that expert tampering is still possible for vehicles that comply with current Euro standards, leading to extreme increases in emissions. They showed that tampered passenger car's NO_x and particle number emissions were ten times higher than those during a regeneration event.

Adopting new exhaust after-treatment technologies brings additional production and maintenance expenses. In 2012, the cost for a complete exhaust after-treatment system compliant with Euro 6 standards for light-duty diesel vehicles was estimated between €1,300 and €1,750 (Posada Sanchez et al., 2012). One basic maintenance requirement for systems using SCR is refilling the diesel emission fluid, commonly known as AdBlue. The cost of refilling the fluid varies depending on the vehicle, its use, driving style, etc., for a heavy goods vehicle it can add up to €2,700 annually. The system also includes several sensors, which can trigger dashboard warnings and initiate a countdown to engine shutdown if issues arise. To circumvent the maintenance costs some have turned to using AdBlue emulators. These devices bypass the SCR system by sending false signals to the emission controls, suggesting that the system is functioning correctly and preventing the vehicle from entering the limp mode, where the vehicle's speed is reduced and less important parts like air-conditioning switched off (Ellermann et al., 2018). In a report to the Danish Road Traffic Authority (Janssen and Hagberg, 2018) the chasing method was tested against PEMS and was suggested as a possible screening tool to find emission noncompliant heavy-duty vehicles.

In this context, on-road vehicle emission measurements serve as a critical tool not only for assessing compliance but also for driving policy discussions and fostering transparency within the automotive industry.

This paper seeks to explore the implications of on-road emissions measurements on diesel-powered cars, with a focus on the significant reductions achieved in BC and NO_x emissions. Through a comprehensive analysis of three on-road vehicle-chasing campaigns, we aim to elucidate the effectiveness of regulatory measures, the role of independent monitoring, and the prospects for advancing toward cleaner and more sustainable transportation systems. Three on-road chasing measurement campaigns were conducted in the decade when exhaust aftertreatment system devices such as DPF and SCR were made mandatory for diesel-powered vehicles, capturing the dieselgate scandal and the effects of more stringent testing with PEMS and RDE for type approvals. The results of the three campaigns show a decreasing trend in vehicle BC and NO_x emission factors (EF). Categorizing vehicles by their respective vehicle emission standards we show the effectiveness of the standards and the consistency of the chasing method results. We also calculate the contribution of super emitters and present them with Lorentz curves and Gini indexes to compare the three campaigns.

2. Methods

Three on-road chasing campaigns were conducted in the 12 years in Slovenia (Europe). The first one was conducted in winter in December 2011, while the second and third campaigns were conducted in spring: March 2017 and May 2023. All three were carried out on regional roads and highways. The results of the 2011 campaign were published in (Ježek et al., 2015a), while the results of the 2017 and 2023 campaigns were not published before.

The average daily weather conditions of each campaign are summarized in Table 1. Temperature, relative humidity, and wind speed at the Ljubljana measurement station as reported by the Slovenian Environmental Agency at the Ljubljana measurement station. The entire list of measurement dates and the weather conditions are collected in Supplement table 1.

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Table 1 Summary of average daily conditions as reported by Slovenian Environmental Agency for Ljubljana Bežigrad station. https://meteo.arso.gov.si/met/sl/app/webmet/.

Campaign	Measurement period	Average daily T range (°C)	Daily avg. relative humidity range	Avg. wind speed (m/s)
2023	3rd May – 19th May	11.9 – 18.1	58 – 85%	0.7 – 2.3
2017	13th March – 23rd March	6.3 – 14.0	59 – 89%	0.6 – 3.1
2011*	6th December – 21st December	-1.3 – 9.3	76 – 94%	0.6 – 1.7

*Published in (Ježek et al., 2015a).

We employed the same on-road chasing approach as was tested in (Ježek et al., 2015b), and adopted in the 2011 campaign by Ježek et al., 2015a, where more details on the method are described and a comparison to other remote sensing campaigns was made. In short, with the on-road chasing measurements, the increase of pollutants over their background concentrations is determined by chasing a single vehicle on the road and then deducting the background concentrations obtained before and/or after the vehicle measurement. Assuming equal dilution of all released pollutants and complete combustion of fuel, in which practically all the carbon in the fuel is converted to CO_2 (Hansen and Rosen, 1990), the EF is then derived as the ratio of pollutant (P) to CO_2, where CO_2 is then used to estimate the amount of fuel burned:

$$EF_P = \frac{\int_{tj}^{ti}(P_{tj}-P_{ti})dt}{a \cdot \int_{tj}^{ti}(CO_{2\,tj}-CO_{2\,ti})dt} \cdot w_c \quad , \tag{1}$$

Where the coefficient “a” in the denominator represents the mass ratio between C and CO_2: a = 12:44 = 0.2727; thus, converting the mass concentration of CO_2 in Eq. (1) to units of mass concentration of C (mg C per m^{-3}). The carbon fraction in fuel w_c for both gasoline and diesel were set to 0.86 (Huss et al., 2013). The subscripts t_i and t_j denote the time of the beginning and end of the integration step, respectively. NO_x was treated as NO_2 equivalent with a molar mass of 46 g mol^{-1} (Wang et al., 2012). Our mobile platform was a car in which we installed an Aethalometer (model AE33, Aerosol Magee Scientific) to measure equivalent black carbon concentration (eBC; from here on expressed as BC), a NO_x monitor (CLD 86 in 2017 and CLDAL2 in 2023, EcoPhysics), and a CO_2 monitor (GMP 343, Vaisala). The list of the used instruments, their time resolution, and measurement uncertainties are provided in Table 2. The instruments were powered by three 100 Ah batteries. The inlet was positioned on the right-hand side window of the mobile platform. The AE33 was equipped with a PM2.5 cyclone as a size-selective inlet.

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Table 2 The list of instruments used in the 2017 and 2023 campaigns, their time resolutions, sampling flows, and measurement uncertainties.

Instrumentation	Species measured	Time resolution	Instrument flow	Measurement sensitivity
NDIR sensor Carbocap GMP343 (Vaisala)	CO_2	2 s	6 l/min (2017) 1 l/min (2023)	3 ppm
Aethalometer AE33 (Aerosol Magee Scientific)	BC	1 s	5 l/min	700 ng/m^3
CLD 86 (Eco Physics) (2017)	NO_x	1 s	0.1 l/min	1% of measurement value
AL2CLD (Eco Physics) (2023)	NO_x	1 s	1 l/min	

During each chase, we recorded the vehicle license plate number of each chased vehicle and thus obtained more detailed vehicle information such as vehicle age, fuel type used, and vehicle type (according to Directive 2001/116/EC, The Commission of the European Communities, 2001) from the registry of the Slovenian Ministry of Infrastructure and Spatial Planning. We disregarded the measurement of vehicles for which we could not get the registry information.

On average the duration of each chase was around a minute and a half in both the 2017 and 2023 campaigns, while in 2011 it was two minutes and a half (Ježek et al., 2015a). We discarded any chase that was shorter than half a minute. The longest chase in 2017 was 13.4 min and 4.5 min in 2023. The traveling speed was changing within each chasing episode. In 2011 most trucks' speed was between 80 and 90 km h^{-1} and for cars, it was between 100 and 130 km h^{-1} (Ježek et al., 2015a). In 2017 and 2023 more measurements were made on regional roads or traveling through small towns so the speed was lower, mostly between 50 and 90 km h^{-1}.

3. Results

Our total fleet size was 406 and 256 vehicles in 2017 and 2023, respectively. We grouped vehicles into three major categories: Gasoline cars, Diesel cars, and Goods vehicles as in (Ježek et al., 2015a). The sample size for each group and vehicle type as determined by European directive 2001/116/EC (The Commission of the European Communities, 2001) are included in Table 3 along with the sample size of the 2011 campaign by Ježek et al., 2015a, where more details on which types of vehicles were included in each group can be found. The total sample size in the 2011 campaign as reported by Ježek et al, 2015a, was 139, however, in this analysis, we could only use the results of the vehicles with the registry information available, which was 118 total.

In the 2011 campaign, Ježek et al., 2015a, reported that their focus was to measure diesel cars, for which BC EF were not published yet. The 2017 and 2023 campaigns were intended to monitor the vehicle fleet and the effectiveness of the emission standards after the "dieselgate" scandal. We therefore analyzed the changes in the total EU and Slovenian fleet compositions regarding the trends in fuel used, engine size, and vehicle age over time, and compared it with our vehicle fleet to check that the captured fleet is a representative sample of the measured fleet. We found that the Slovenian fleet is a good representation

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of the European average fleet as the size of both is increasing over time at the same rate, the share of diesel and gasoline-powered cars is approximately the same, and the engine size and car age groups are similar. The samples were representative for diesel and gasoline cars but not as representative for goods vehicles where our sample size was smaller. The fleet analysis can be found in Supplement Information 1. In section 3.1 we present the general trends and improvements in EF distributions, comparing the results of gasoline cars, diesel cars, and goods vehicles from the three campaigns 2011, 2017, and 2023. We then break the three main vehicle categories according to their vehicle emission standards in section 3.2 and explore the effectiveness of the implemented technology and legislation. In section 3.3 we demonstrate how a small fraction of vehicles disproportionally contributes to total fleet emissions and how the Lorentz curves became more skewed over the three measurement campaigns.

Table 3 Sample size in the 2023 and 2017 campaigns compared to the 2011 campaign by Ježek et al. 2015. Vehicle categories and their corresponding 2001/116/EC categories.

Category	Vehicle type	2001/116/EC	2023	Category total 2023	2017	Category total 2017	2011 (Ježek et al., 2015a)
Gasoline cars	Gasoline cars	M1	101	103	118	121	24
	Gasoline/LPG	M1	1		3		
	Light goods vehicles 1	N1	1		0		
Diesel cars	Diesel cars	M1	90	122	129	170	66
	Light goods vehicles 1	N1	32		41		
Goods vehicles	Light goods vehicles 2	N2	2	31	53	113	28
	Buses	M3	6		15		
	Minibus	M2	1		0		
	Heavy goods vehicles	N3	22		45		
TOTAL			**256**		**404**		**118**

3.1. Trends and Improvements in Emission Factor Distributions - Comparative Analysis of Gasoline Cars, Diesel Cars, and Goods Vehicles (2011-2023)

The trends of BC and NO_x EF over the three measurement campaigns for the three main vehicle groups—gasoline cars, diesel cars, and goods vehicles are presented — in Figure 1. This figure shows the EF distributions for each vehicle group across the three campaigns using box-and-whisker plots, which also include the minimum and maximum values.

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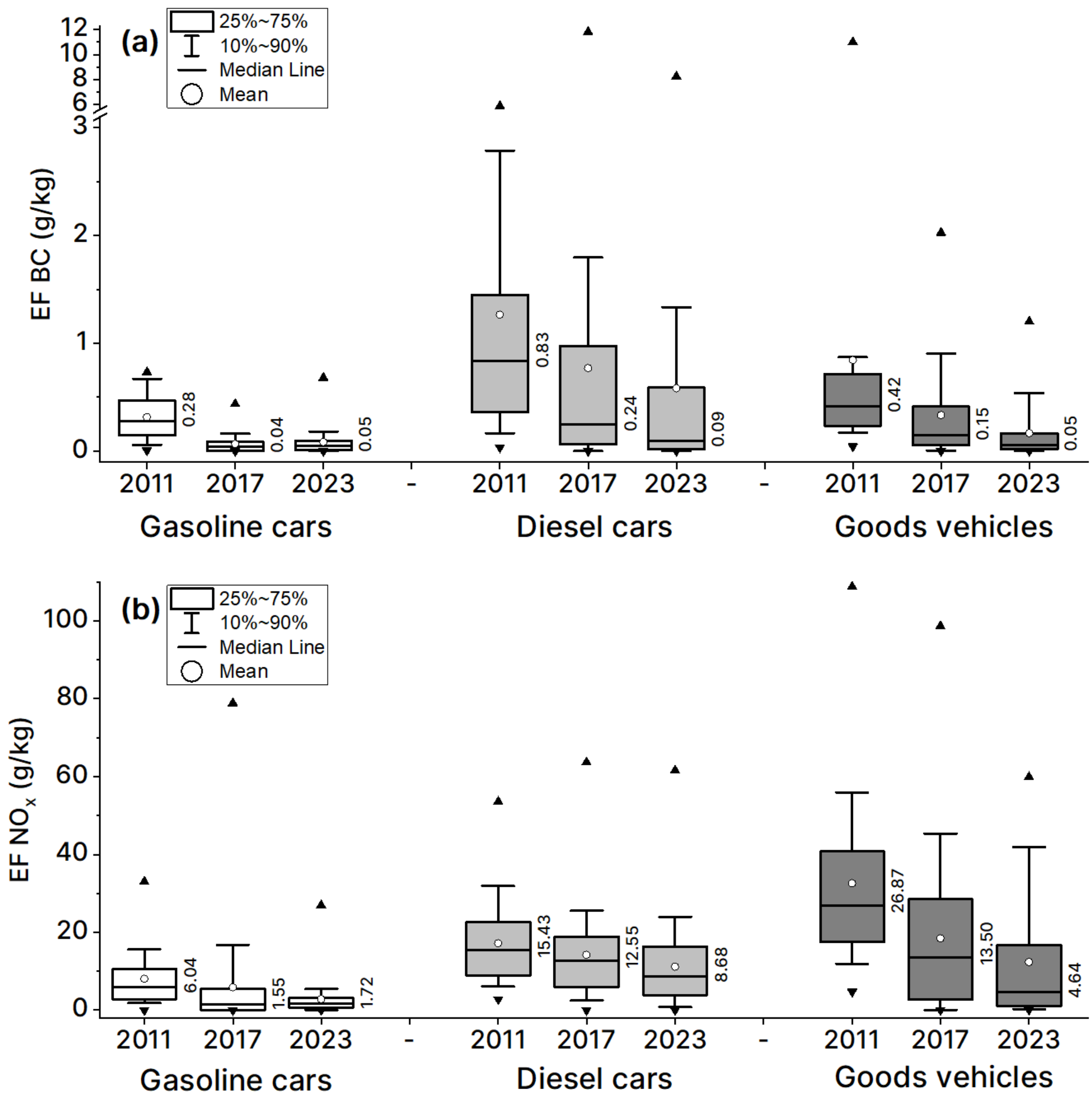

Figure 1 Box-and-whisker plot for (a) BC EF (b) NO_x EF of Gasoline cars, Diesel cars, and Goods vehicles in the three on-road measurement campaigns conducted in 2011 (Ježek et al. 2015), 2017, and 2023. The black triangles represent the group's minimum and maximum EF measured in the respective campaign. The numbers right of the box plot are the median values. Note the scale break for BC EF.

Gasoline-powered cars exhibit the lowest BC and NO_x EF distributions among the three vehicle categories (median BC EF values are 0.28, 0.04, and 0.05 g kg^{-1} for 2011, 2017, and 2023 campaigns respectively). In the 2011 campaign, gasoline-powered cars' BC and NO_x EF were higher compared to those in the subsequent campaigns in 2017 and 2023. Gasoline-powered cars' maximum BC EF from the three campaigns were all in the range of the median diesel-powered cars in 2011

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(0.83 g kg^{-1}) or average 2023 (0.58 g kg^{-1}), while the maximum NO_x EFs (33.01 g kg^{-1}, and 26.88 g kg^{-1} in 2011 and 2023 respectively) were in the range of the 90th percentile of diesel-powered cars (or higher like in 2017 when it was 78.83 g kg^{-1}). Diesel-powered cars show a consistent reduction in the average, median, and interquartile range of BC EF across the three campaigns: the median values decreased from 0.83 g kg^{-1} in the 2011 campaign to 0.24 g kg^{-1} and 0.09 g kg^{-1} in 2017 and 2023 campaigns, respectively. This trend indicates significant progress in reducing particulate emissions from diesel-powered cars. However, the NO_x EF for diesel-powered cars only shows a slight reduction from the 2011 to 2017 campaign (median 15.43 g kg^{-1} and 12.55 g kg^{-1}, respectively) and some improvement in 2023 (median 8.86 g kg^{-1}). This suggests that while particulate emissions have been effectively addressed, NO_x emissions from diesel-powered cars remain a challenge, albeit with some improvements. The high maximum BC EF values skew the distribution of diesel-powered cars so the median and the average values don't show the same level of improvement in the group, whereas the NO_x EF median and average are more aligned and show the same trend.

The BC EF distributions for goods vehicles generally feature slightly lower values than those of diesel cars within the same campaigns, and they exhibit a decreasing trend over time (median values in 2011, 2017, and 2023 campaigns were 0.42, 0.15, and 0.05 g kg^{-1} respectively). This indicates progress in reducing particulate emissions from goods vehicles with DPFs. The goods vehicles' NO_x EF distribution values were higher than diesel-powered cars' NO_x EF distribution in the 2011 campaign (median 26.9 g kg^{-1}), similar in the 2017 campaign (median 13.5 g kg^{-1}), and lower than diesel cars in the 2023 campaign (median 4.6 g kg^{-1}). Therefore, goods vehicles showed a more significant improvement in NO_x emissions over the three campaigns.

Overall, the EF for BC and NO_x have generally decreased across all vehicle categories over the study period, reflecting advancements in vehicle technology and emissions control strategies. However, the varying reduction rates between BC and NO_x EFs among the vehicle types highlight the ongoing challenges and areas for further improvement in emission reductions.

3.2. Analysis by Vehicle Emission Standards.

To investigate the influence of the vehicle emission standards we broke down each main vehicle category into smaller sub-groups according to vehicle emission standards for all three campaigns. Figure 2 illustrates the comparison of median values for each sub-group across the three campaigns.

Figure 2 shows that the results for each subgroup are consistent over the three campaigns. This consistency highlights the reliability of our measurements, and the trends observed in the total groups' EF distributions. The exceptions, as previously noted, are the gasoline cars in 2011 and the NO_x EF of goods vehicles. The discrepancies observed for gasoline cars in the 2011 campaign are likely due to different weather conditions that may have influenced the emissions of the smaller sample size. In the case of goods vehicles, the diverse range of vehicle types and the smaller sample size contribute to the variability in NO_x emissions, making inconsistencies in these results less unexpected. The sample size of each subgroup is reported in Supplement table 2, and the median and interquartile ranges for BC and NO_x EF are in Supplement table 3 and 4, respectively.

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Overall, the consistency in the results over the three campaigns underscores the effectiveness of emission standards and the improvements in vehicle technologies to reduce emissions. This detailed breakdown by vehicle emission standards provides a clearer understanding of how specific groups have progressed over time.

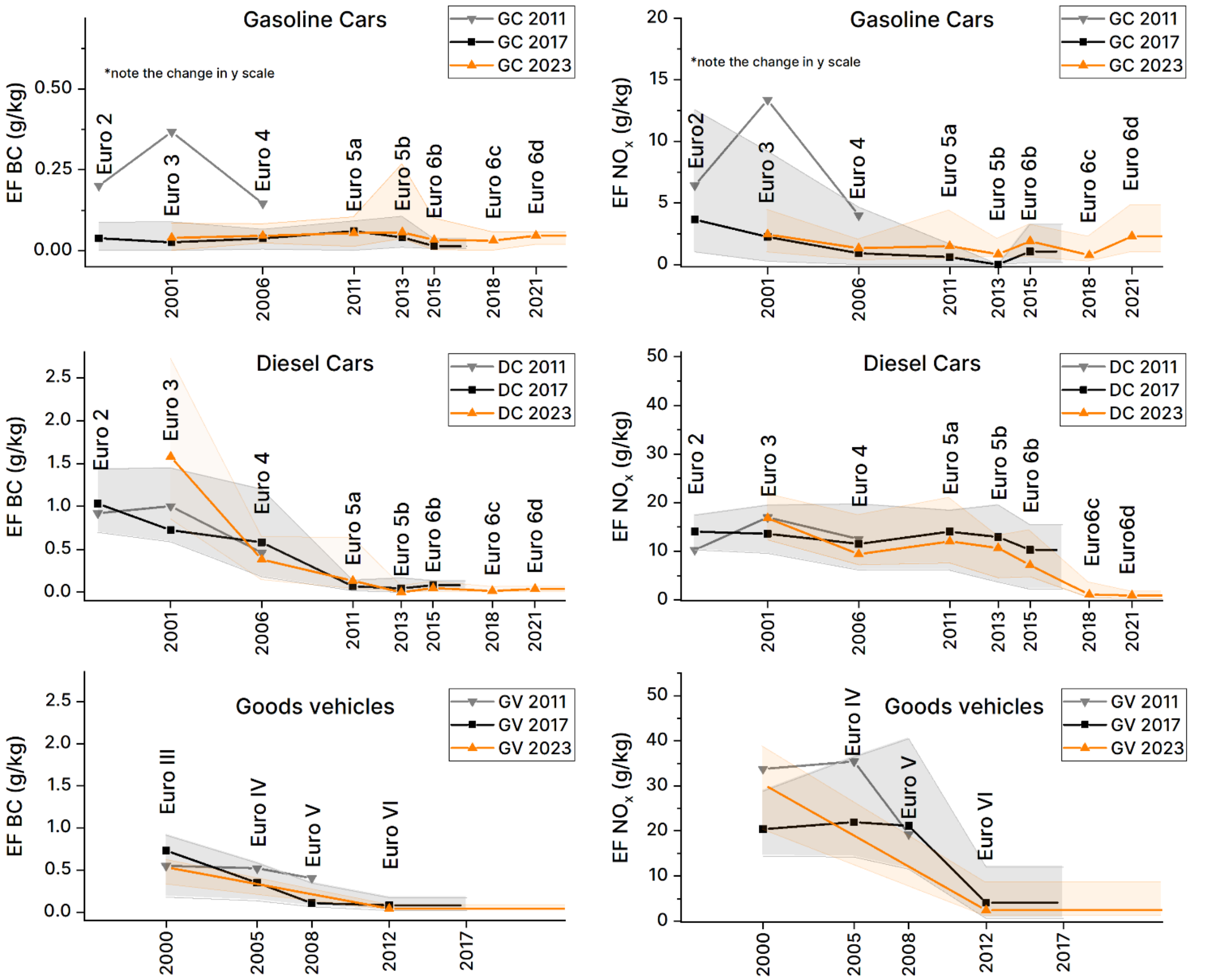

Figure 2 Black carbon (left column) and NO_x (right column) emission factors (EF) for the Gasoline cars (top row), Diesel cars (middle row), and Goods vehicles (bottom row). The points represented with grey inverted triangles, black squares, and orange triangles are the results for the 2011, 2017, and 2023 campaigns, respectively. They represent the median values for all vehicles that were registered after the point and 'belong to specific emission standard', the interpolated lines indicate the trend between two standards, and the shaded area is an interpolated interquartile range of the period.

A notable drop in BC EF was observed for diesel-powered cars and goods vehicles with the introduction of Euro 5 and Euro V standards, respectively. This reduction in both the median and interquartile range can be attributed to the mandatory implementation of DPF to meet the desired PM emission reductions set by these standards.

For diesel-powered cars with Euro 5a, the median BC EF dropped below the 25th percentile of Euro 4 in both the 2017 and 2023 campaigns. In the 2017 campaign, the Euro 5a median was 0.07 g kg^{-1} and was much lower compared to the Euro 4 25th percentile of 0.18 g kg^{-1}. In the 2023 campaign, both values were 0.13 g kg^{-1}. With Euro 5b and later standards, the 75th percentile of BC EF dropped below the 25th percentile of Euro 4 in both the 2017 and 2023 campaigns.

These reductions brought diesel-powered car BC EF in line with gasoline-powered car BC EF, reflecting the legislation that introduced PM emission limits for gasoline-powered cars with Euro 5a standard and set the same PM emission limit for both gasoline- and diesel-powered cars (0.005 g km^{-1} for Euro 5a, 0.0045 g km^{-1} for subsequent standards, see also Supplement figure 5).

Additionally, while the Euro VI standard managed to reduce the NO_x EF of goods vehicles through the availability of new technologies such as SCR, a notable drop in NO_x EF of diesel-powered cars was only observed after the enforcement of more stringent type of approval tests with RDE under the Euro 6c and Euro 6d standards. Our Euro 6 diesel-powered cars results match the results from the TRUE project (Bernard et al., 2021), which reports pre-RDE Euro 6 fuel-specific NO_x emissions of around 7.5 g kg^{-1} (we found Euro 6b EF 7.12 g kg^{-1}) and Euro 6d around 2.0 g kg^{-1} (in our work Euro 6d was 0.9 g kg^{-1}) by using the remote sensing method.

The reduction in NO_x emissions through legislation was more gradual compared to PM emissions. The NO_x emission limits were progressively tightened, first being halved from Euro 3 to Euro 4, then decreased by a third from Euro 4 to Euro 5, and halved again from Euro 5 to Euro 6. For gasoline-powered cars, the NO_x emission limit was set at 0.15 g km^{-1} with the Euro 3 standard. Until the introduction of Euro 6b, gasoline car limits were approximately one-third of diesel car NO_x limits. After Euro 6b, they were set at 75% of diesel car limits (graphically presented in Supplement figure 5).

Our results, however, did not reflect this regulatory trend. Before Euro 6b, the median NO_x emissions for gasoline cars were 0-16% of their diesel counterparts. For Euro 6b, gasoline-powered cars' emissions were 9% (2017) and 27% (2023) of diesel car emissions. After Euro 6b, diesel cars had similar or higher median NO_x emissions compared to gasoline cars.

We compared our NO_x EF distributions against legislative trends (Figure 3), plotting the median, 25th, and 75th percentiles of each campaign's subgroups against NO_x standards from Euro 3 to Euro 6b. Euro 6c and d were excluded due to identical limits to Euro 6b but different testing methods (RDE). Linear regression and ANOVA analysis ($\alpha = 5\%$) revealed that for diesel cars, the 2023 campaign showed a significant downward trend in median and 25th percentile values, while the 2017 campaign showed a significant downward trend only in 25th percentile values. For gasoline cars, only the 75th percentile values in the 2017 campaign showed a significant trend.

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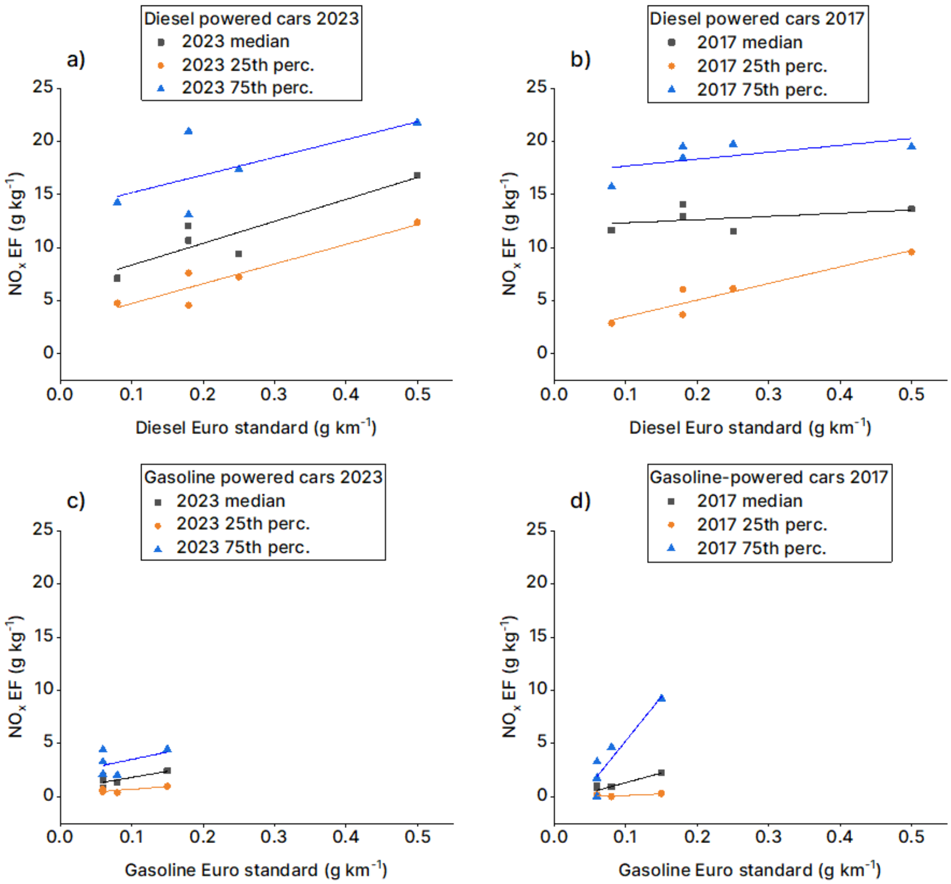

Diesel	2023 median	2023 25th perc.	2023 75th perc.	2017 median	2017 25th perc.	2017 75th perc.
R-Square (COD)	0.82	0.88	0.47	0.18	0.88	0.38
Intercept	6.30 ± 1.55	2.89 ± 1.12	13.49 ± 2.84	12.02 ± 1.04	1.95 ± 0.90	17.02 ± 1.32
Slope	20.60 ± 5.61	18.57 ± 4.04	16.67 ± 10.24	3.05 ± 3.74	6.49 ± 4.78	6.49 ± 4.78
Prob>F	0.035	0.019	0.202	0.475	0.017	0.267
Gasoline	2023 median	2023 25th perc.	2023 75th perc.	2017 median	2017 25th perc.	2017 75th perc.
R-Square (COD)	0.56	0.58	0.21	0.76	0.56	0.86
Intercept	0.66 ± 0.53	0.23 ± 0.21	2.10 ± 1.38	-0.47 ± 0.52	-0.10 ± 0.11	-3.05 ± 1.72
Slope	11.54 ± 5.94	4.69 ± 2.33	13.97 ± 15.51	18.02 ± 5.79	83.23 ± 19.27	83.23 ± 19.27
Prob>F	0.147	0.137	0.434	0.053	0.148	0.023

Figure 3 NO_x EF median (black squares), 25th (orange circles), and 75th percentiles (blue triangles) against the NO_x legislation standards. The top row shows the results for diesel-powered cars in the 2023 campaign (a) and 2017 campaign (b). The bottom row shows the results for gasoline-powered cars in the 2023 campaign (c) and 2017 campaign (d). The lines in coordinating colors are the linear regression lines with their parameters (R-square, intercept, slope, and Prob> F, where α = 5%) are listed in the table below.

These results suggest an improvement in a quarter of diesel cars' NO_x emissions, with vehicles achieving lower emissions in successive standards, as indicated by lowering 25th percentiles in both campaigns. However, there was no significant change

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in the 75th percentile, implying that a quarter of vehicles remained unaffected by stricter legislation. In 2017, median values for diesel cars didn't change significantly with legislation, while in 2023, median values for diesel cars correlated with legislative changes. This discrepancy between the two campaigns may be the effect of the recall of 8.5 million cars in Europe which reached 79.7% by July 2018 by the Volkswagen group.

While our goods vehicles sample size was relatively small in the 2023 campaign and most of the captured vehicles were compliant with Euro VI, we can see from both the 2017 and 2023 campaign the decrease in NO_x and BC emissions, as both drop to the level of cars NO_x EFs.

3.3. Super-emitter Contribution

Lorenz curves are used to assess the impact of so-called super-emitters. The curves show the proportion of total emissions (y-axis) produced cumulatively by the bottom x% of the fleet, ranking vehicles from least to most polluting. A straight line between the origin (no emissions) and maximum emissions on a 1:1 scale (and hence the line at 45°) would indicate equal pollution contribution by all vehicles. To compare these lines between the three campaigns we calculated the Gini index, which represents the ratio between the area between the Lorenz curve and the perfect equality line at 45° and the entire area under the perfect equality line. We analyzed each campaign, vehicle group, and each pollutant separately. Figure 4 reveals that super-emitters' contribution is more skewed for BC emissions (a, b, c) than NO_x emissions (d, e, f), as demonstrated by generally higher Gini indexes closer to 1. The curves become typically more skewed over the three campaigns, as shown by generally increasing Gini indexes, a small fraction of vehicles contribute more pollution than most vehicles. A Gini index closer to 0 would indicate that all vehicles in the group contribute the same to total fleet emissions.

The top 10% of gasoline-powered car emitters with the highest BC EF contributed 21%, 44%, and 47% of total gasoline car fleet BC emissions in 2011, 2017, and 2023, respectively (Figure 4 a). The top 10% of diesel cars with the highest BC EF contributed 36%, 51%, and 65% of total diesel car fleet BC emissions in the same years (Figure 4b). For goods vehicles, the top 10% contributed 53%, 43%, and 48% of total BC emissions in 2011, 2017, and 2023, respectively (Figure 4c).

In terms of NO_x emissions, the top 10% of gasoline-powered cars contributed 31%, 59%, and 44% of total gasoline car fleet NO_x emissions in 2011, 2017, and 2023, respectively (Figure 4 d). For diesel-powered cars, the top 10% contributed 22%, 28%, and 29% of total NO_x emissions in the same years (Figure 4 e). The top 10% of goods vehicles contributed 22%, 30%, and 41% of total goods vehicles' fleet NO_x emissions in 2011, 2017, and 2023, respectively (Figure 4 f). The contribution is increasing over the campaigns, which means that by lowering EF in general, the super emitters are even more important if we want to further improve the air quality in the cities.

Excluding vehicles that disproportionately contribute to total fleet emissions from city access would be a more effective measure to reduce traffic emissions than excluding vehicles based on specific emission standards (Ježek et al., 2018). This approach would also notify and incentivize vehicle owners to maintain their vehicles better.

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To prepare an effective strategy to reduce overall pollution in a city, the focus should be on targeting the highest-emitting vehicles, regardless of whether they are diesel or gasoline-powered. The Gini index analysis showed that equal contribution to emissions does not necessarily mean the emissions are good or desirable – as is shown on Figure 4 diesel cars with a more uniform NO_x emissions profile had problematic overall NO_x levels.

However, to find vehicles that may be using defeat devices, a more granular analysis of different vehicle groups would be beneficial. The Lorenz curves could be used as a monitoring tool to identify outliers within specific vehicle categories, as those could indicate cars with malfunctioning emissions control systems or illegal tampering.

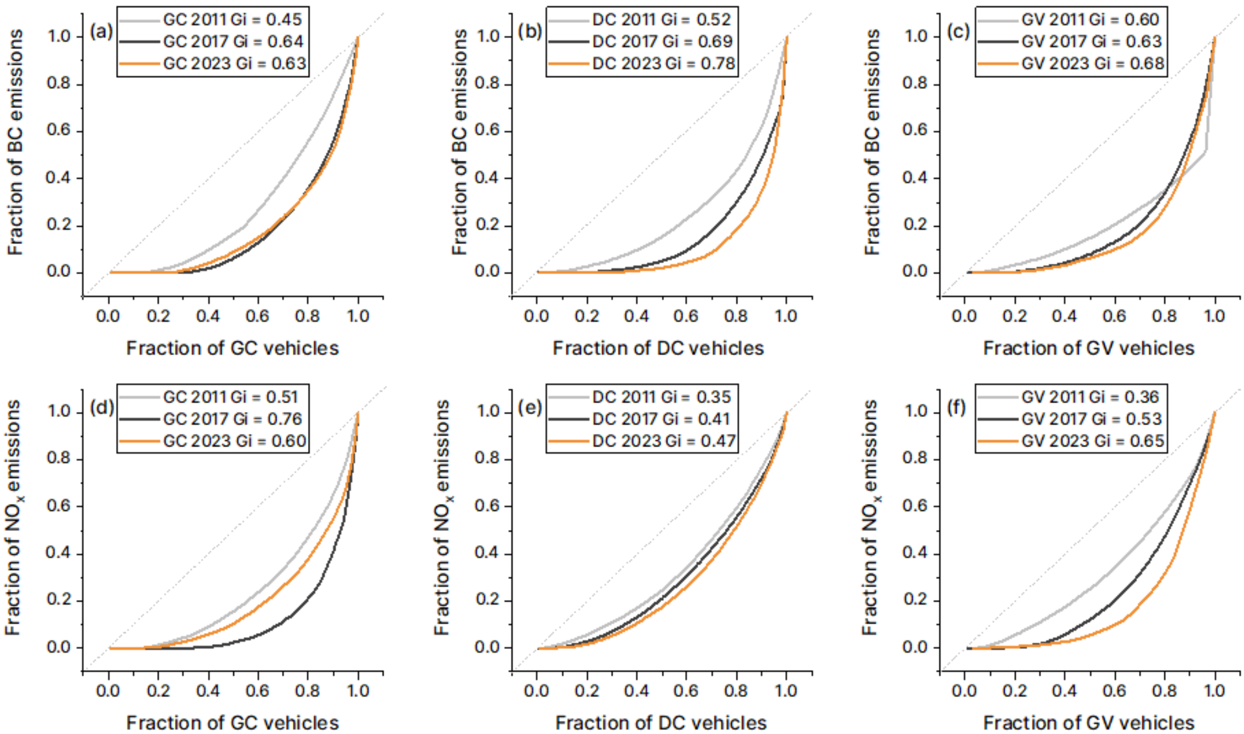

Figure 4 Supper emitter curves are presented as Lorenz curves, where we distribute vehicles within each group from least to most polluting and sum their emissions; and Gini indexes (Gi). We did this separately for Gasoline Cars (GC), Diesel Cars (GC), and Goods vehicles (GV), and compared their results in the 2023 (orange), 2017 (black), and 2011 (gray) campaigns.

4. Discussion

Ježek et al., (2015a), found good consistency between the results of different calculation approaches to chasing data and between results of various study types, including remote sensing, chasing, mobile measurements, and the European Environmental Agency's emission inventory. This further demonstrates the consistency and reliability of the measurement methods used in these studies. In recent years, new methodologies and dedicated instrumentation have been developed (Farren

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et al., 2023; Olin et al., 2023), with increased interest in determining the most robust independent methodology for screening many vehicles for defeat devices (Ellermann et al., 2018; Janssen and Hagberg, 2018).

One significant challenge in measuring vehicle emissions in real-world conditions is the high variability in vehicle operation, which affects emissions. Considerable effort has been made to enhance the robustness and repeatability of on-road measurements using PEMS. The procedures for RDE testing with PEMS are summarized in the Joint Research Centre's Report (Valverde Morales and Bonnel, 2018). This report guides the preparation, execution, and data quality checks for emissions tests with PEMS on light-duty vehicles, following EU legislation. Additionally, Zardini and Bonnel, (2020) present a detailed analysis of 79 tests conducted on 11 passenger cars, summarizing the latest EU-RDE procedure (RDE-4, Regulation EU 2018/1832). They emphasize the responses provided by the RDE data analysis tool, EMROAD version 6.03, developed and maintained by the Joint Research Center.

The data collection includes RDE tests designed to cover a broad range of environmental conditions (e.g., temperature and altitude) and to challenge the trip dynamics requirements defined by legislation to represent typical vehicle use. The EMROAD tool evaluates trip validity based on factors such as trip duration, distance, distance shares in specific driving regimes (e.g., urban), vehicle speed and speed shares, trip dynamics, ambient conditions, elevation gain, trip severity relative to the WLTP driving cycle (based on CO_2), and emissions of pollutants, along with their correction for ambient boundary conditions and excess severity. For better understanding and comparison, it would be useful to make such detailed assessments also with the chasing method.

PEMS measurements typically last about 90 minutes per vehicle, whereas our chasing measurements averaged 90 seconds per vehicle in the 2017 and 2023 campaigns. Given the limited parameters controlled with the chasing method, further investigation is warranted. However, studies suggest it can be a useful screening tool (Farren et al., 2023; Vojtisek-Lom et al., 2020).

5. Conclusions

Our analysis of three on-road chasing campaigns conducted in 2011, 2017, and 2023 demonstrates the robustness of the chasing method as a consistent and reliable approach for determining BC and NO_x EF under real driving conditions. The study provides critical insights into the temporal evolution of vehicle fleet emissions and their implications for atmospheric science.

The significant reductions in BC EF achieved through the widespread adoption of DPFs and the drastic decreases in NO_x emissions resulting from the implementation of SCR systems and stricter regulations are notable advancements. These improvements have brought emissions from diesel-powered vehicles, including goods vehicles, closer to those of gasoline-powered vehicles. Such advancements are essential for refining the input parameters of atmospheric dispersion and chemical transport models, which rely on accurate real-world EF data to predict pollutant distributions and transformations.

Despite these technological achievements, challenges such as improper vehicle maintenance and tampering with exhaust systems remain significant obstacles to achieving further reductions in traffic-related emissions. Addressing these issues is critical not only for regulatory compliance but also for improving the accuracy of emissions inventories used in modeling.

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Real-world EF measurements are indispensable for bridging the gap between ambient pollution observations and traffic emission models. They provide a direct link between vehicle emissions and their environmental impact, enabling more robust validation and improvement of pollution models' results. This integration enhances our ability to model atmospheric processes, predict air quality, and develop targeted mitigation strategies informed by reliable data.

The Gini index analysis has shown that even when the emissions are more equally distributed across the vehicle fleet, the overall pollution levels can still be problematic. This suggests that targeting the highest-emitting vehicles, whether they are diesel or gasoline-powered, should be the focus of any effective pollution reduction strategy.

A robust method for measuring vehicle emissions in real driving conditions, as suggested by our work, would be a valuable tool for monitoring vehicle emissions independently of the vehicle driver or owner's knowledge, thereby bypassing any potential interference from manufacturers and vehicle owners who may be using defeat devices or failing to properly maintain their vehicles.

Additionally, a comprehensive measurement base would facilitate the development of more accurate models for forecasting traffic emissions. It would also enhance the efficiency of measures implemented by cities to reduce traffic pollution and provide a means to monitor and evaluate their effectiveness. This comprehensive data collection could guide policymakers in making informed decisions and implementing effective strategies to mitigate urban air pollution.

By combining the insights from the Gini index and Lorenz curve analyses, policymakers can identify the highest-emitting vehicles regardless of fuel type, while also using real-world emissions monitoring to detect vehicles with malfunctioning or tampered emissions control systems. This holistic approach, focused on the actual emissions performance rather than just the vehicle technology, is essential for developing comprehensive policies to significantly improve air quality in cities.

Author contribution: IJB, AG, MR, and GM designed the experiments. IJB, LZ, and TR carried them out, and MI and BA provided technical assistance. IJB, LZ, TR, and MI performed the data analysis. GM and AG acquired funds for the projects. MR, AG, IJB, and GM supervised the projects. IJB wrote the initial draft. All authors discussed the results and contributed to the final manuscript.

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Conflict of interests:

The authors IJB, AG, MI, BA, GM, and MR were employed by Aerosol d.o.o., the manufacturer of Aethalometers, at the time or part of the time while the study was conducted.

Data availability:

Due to the Personal Data Protection Act, the distribution of the dataset is limited, parts may be available upon request.

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