Below are compiled the authors' responses to the anonymous Referees (RC) and the Editor.

RESPONSE TO RC#1

AC: We greatly appreciate RC#1's time and effort in reviewing our manuscript. We agree with most of the points raised and have thus made corresponding improvements to the texts. Below is the list of edits that follow the numbering made by RC#1.

To make the comments and responses easier to follow, we applied the following formatting to the text: the Anonymous Referee's comments are in black font; the Authors' Response is in blue italic font. Revisions made to the manuscript in response are summarized in a table with corresponding line numbers.

RC#1

1. Line 89: The results of the 2011 campaign were published in (Ježek et al., 2015a)… Change with "The results of the 2011 campaign were published in Ježek et al. (2015a)

AC: We apologize for this nuisance. Brackets were removed from around the reference and put around the publication year throughout the text.

RC#1

2. Line 92: the words "at the Ljubljana measurement station" are reported twice in the same sentence. Correct the sentence

AC: Deleted duplication.

RC#1

3. Line 98: improve the writing of (Ježek et al., 2015b) citation as reported at point 1. This comment is valid all along the manuscript. Please take care of citations.

AC: Citations were corrected accordingly throughout the text.

RC#1

4. Line 99: put in brackets the year of reference cited

AC: Brackets added accordingly.

RC#1

5. Line 102: please discuss in a quantitative way the maximum error that can be done "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)"

AC: We initially did not discuss the methodology in detail in this manuscript since the same method was used as in Ježek at al. (2015b), and Ježek at al. (2015a), where the method was tested, described in more detail, based on previous studies, and results were compared to other studies.

The assumption of complete combustion of all released pollutants was evaluated in:

- Dallman et al. (2012): "Although other carbon-containing species were measured in this study, these species are excluded from the carbon balance in eq 2 because they are present at low concentrations relative to CO_2 in diesel exhaust. For example, CO contributed less than 2% of the total carbon measured in the exhaust of the highest (top 10%) CO-emitting trucks. Concentrations of BC, HCHO, and C_2H_4 measured in truck exhaust accounted for <1% of total carbon "
- Hansen and Rosen (1990):" Excluding other carbonaceous species will not introduce an error larger than 10%."
- Dallmann et al. (2011):"..omitting CO and HC from the calculation yields only a 5% positive bias in calculated EFs."

Ježek et al. (2015b) performed tests on the plume-chasing method on cars, where they evaluated the effect of setting the background on the calculated BC EF. The variability of the background was described with two standard deviations of the measured background, which covered 90% of the variation. The two standard deviations were added and subtracted from the background to determine the influence of background variation on the calculated EFs. About -24/+26 % variation on the calculated EF was found.

The dilution did not affect the calculated BC EF (Ježek et al., 2015b). The dilution ratio was highest when the exhaust mass flow rate was low. This was consistent with the findings of Chang et al. (2009), who reported that the dilution ratio depends not only on speed but also on exhaust flow rate and other parameters (like tailpipe position and vehicle shape) that are more important in the near-wake region.

Wang et al. (2020) have obtained similar results. When comparing plume-chasing measurements against PEMS on an HGV, Wang et al. (2020), found relative errors of vehicle-specific emission factors of NOx were within ±20%.

Changes in text added in	Ježek et al. (2015b) found about -24/+26 % variation on the calculated EF due to the determination of the pollutant background concentration, and the dilution did not affect the calculated BC EF.

lines 123-126	Excluding other carbonaceous species from the denominator was estimated to produce a bias smaller than 10% (Hansen and Rosen, 1990). More recent studies found <1 and 5% positive bias when excluding other carbonaceous species (Dallman et al., 2011, 2012).
Added references	Dallmann, T. R., Harley, R. A., and Kirchstetter, T. W.: Effects of diesel particle filter retrofits and accelerated fleet turnover on drayage truck emissions at the port of Oakland, Environ Sci Technol, 45, 10773–10779, 2011. Dallmann, T. R., Demartini, S. J., Kirchstetter, T. W., Herndon, S. C., Onasch, T. B., Wood, E. C., and Harley, R. A.: On-road measurement of gas and particle phase pollutant emission factors for individual heavy-duty diesel trucks, Environ Sci Technol, 46, 8511–8518, 2012.

RC#1

6. Line 111: "NOx was treated as NO2 equivalent with a molar mass of 46 g mol-1 (Wang et al., 2012)" **This is wrong**. Compare for example Carslaw (2013; https://doi.org/10.1016/j.atmosenv.2013.09.026) where NO2/NOx ratio is well below 50% for Diesel passenger cars and other fleet vehicles. I suggest to review the literature (and the aforementioned paper) to find a proper average NO2/NOx ratio for each group of vehicles and apply a proper new molar mass on its base. **Please redo all the computations connected with NOx EF estimation.**

AC: We did not use the NO_2/NO_x ratio to calculate NO_x molar mass because during the 2011 and 2017 campaigns, we lacked instrumentation to measure both NO and NO_2 separately. To ensure comparability across all three campaigns, we maintained consistency in the data analysis and applied the same methodology throughout.

We used the NO_2 equivalent approach, which aligns with the reporting methods used in Wang et al. (2012) and Dallman et al. (2011), as required by the U.S. EPA (2010) under the Code of Federal Regulations Title 40: Protection of Environment, Part 86.

The rationale for using NO_2 equivalents lies in the atmospheric behavior of NO_x. While NO itself is not considered a major pollutant, it rapidly reacts with oxygen to form NO_2, which has well-documented adverse health and environmental effects, including respiratory issues, acid rain, and ozone formation. By assuming complete conversion of NO_x to NO_2 in the atmosphere, emissions are standardized as NO_2 equivalents for regulatory reporting and compliance. This approach enables consistent comparisons of emissions across different sources, such as power plants, vehicles, and industrial operations.

The NO_2/NO_x ratio can vary significantly—Carslaw and Rhys-Tyler (2013) reported values ranging from 12% to 55% for Euro 4 and Euro 5 vehicles. While we could apply a general or vehicle-specific NO_2/NO_x ratio to estimate molar mass, this would introduce additional uncertainty into the results.

This study builds upon the methodology of Ježek et al. (2015a), which also compared its results to other studies, including Carslaw and Rhys-Tyler (2013), as shown in the Table 7 (Ježek et al., 2015a). In Ježek et al. (2015a), emission ratios from Carslaw and Rhys-Tyler (2013) were converted to emission factors using the same molecular weights and carbon fractions as in Ježek et al. (2015a), ensuring methodological consistency and avoiding additional assumptions.

Added to line 122	USEPA, 2010
Added reference	USEPA: Code federal regulations - Title 40: Protection of environment Part 86, 2010.

RC#1

7. Line 112: “eBC”, referring to Savadkoohi et al. (2024; https://doi.org/10.1016/j.envint.2024.108553) ad a proper nomenclature

AC: *As per Savadkoohi et al. (2024), the recommendation for reporting equivalent BC or “eBC” was defined by Petzold et al. 2013. doi:10.5194/acp-13-8365-2013. We therefore added Petzold et al., 2013, reference. We did not use the Savadkoohi et al. (2024) harmonization factor.*

Added to line 128	*(Petzold et al, 2013)*
Added reference	Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S. M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., and Zhang, X. Y.: Recommendations for reporting black carbon measurements, Atmos Chem Phys, 13, 8365–8379, https://doi.org/10.5194/acp-13-8365-2013, 2013.

RC#1

8. Line 115: “The inlet was positioned on the right-hand side window of the mobile platform”. Please describe the inlet and its capability to work as isokinetic or not before the PM2.5 cyclone. And in the last case discuss the possible influence on the car speed on the capability of AE33 inlet to effectively collect the BC in a proper way.

AC: *The methodology used in this study was tested and reported in previous papers. Isokinetic sampling was addressed in Wang et al., 2011:" Isokinetic sampling was not attempted because particles studied were in the submicron size range, and the Stokes number was in the range of 10^{-7} to 10^{-3}, which was far less than unity. This indicated that the effects of particle inertia and isokinetic sampling were not significant ..."*

We did not find papers that would have directly measured how vehicle speed affects the AE33 inlet's BC collection efficiency. Ježek et al. (2015b) show that BC emissions are changing with driving but are independent of speed.

Added test in lines 131-132	, isokinetic sampling was not attempted as it is not deemed necessary at the vehicle speeds (Wang et al., 2011)

RC#1

9. Improve Figure 3 by also adding on the x-axis the label of each Euro standard referred to each numerical limit.

AC: *The x-axis scale on Figure 3 was customized to the Euro standard values, and each standard's label was added for clarification.*

The list of references used in this reply:

Carslaw, D. C., Rhys-Tyler, G., New insights from comprehensive on-road measurements of NOx, NO2 and NH3 from vehicle emission remote sensing in London, UK, Atmospheric Environment, Volume 81, 2013, Pages 339-347, https://doi.org/10.1016/j.atmosenv.2013.09.026.

Chang, V. W., Hildemann, L. M., and Chang, C. H.: Dilution rates for tailpipe emissions: effects of vehicle shape, tailpipe position, and exhaust velocity, J. Air Waste Manage, 59, 715–724, 2009.

Dallmann, T. R., Demartini, S. J., Kirchstetter, T. W., Herndon, S. C., Onasch, T. B., Wood, E. C., and Harley, R. A.: On-road measurement of gas and particle phase pollutant emission factors for individual heavy-duty diesel trucks, Environ Sci Technol, 46, 8511–8518, 2012.

Dallmann, T. R., Harley, R. A., and Kirchstetter, T. W.: Effects of diesel particle filter retrofits and accelerated fleet turnover on drayage truck emissions at the port of Oakland, Environ Sci Technol, 45, 10773–10779, 2011.

Hansen, A. D. A. and Rosen, H.: Individual Measurements of the Emission Factor of Aerosol Black Carbon in Automobile plumes, Journal of Air Waste Management Association, 40, 1654–1657, 1990.

Ježek, I., Drinovec, L., Ferrero, L., Carriero, M., and Močnik, G.: Determination of car on-road black carbon and particle number emission factors and comparison between mobile and stationary measurements, Atmos Meas Tech, 8, https://doi.org/10.5194/amt-8-43-2015, 2015b.

Ježek, I., Katrašnik, T., Westerdahl, D., and Mocnik, G.: Black carbon, particle number concentration and nitrogen oxide emission factors of random in-use vehicles measured with the on-road chasing method, Atmos Chem Phys, 15, https://doi.org/10.5194/acp-15-11011-2015, 2015a.

Marjan Savadkoohi, Marco Pandolfi, Olivier Favez, Jean-Philippe Putaud, Konstantinos Eleftheriadis, Markus Fiebig, Philip K. Hopke, Paolo Laj, Alfred Wiedensohler, Lucas Alados-Arboledas, Susanne Bastian, Benjamin Chazeau, Álvaro Clemente María, Cristina Colombi, Francesca Costabile, David C. Green, Christoph Hueglin, Eleni Liakakou, Krista Luoma, Stefano Listrani, Nikos Mihalopoulos, Nicolas Marchand, Griša Močnik, Jarkko V. Niemi, Jakub Ondráček, Jean-Eudes Petit, Oliver V. Rattigan, Cristina Reche, Hilkka Timonen, Gloria Titos, Anja H. Tremper, Stergios Vratolis, Petr Vodička, Eduardo Yubero Funes, Naděžda Zíková, Roy M. Harrison, Tuukka Petäjä, Andrés Alastuey, Xavier Querol, Recommendations for reporting equivalent black carbon (eBC) mass concentrations based on long-term pan-European in-situ observations, Environment International, Volume 185, https://doi.org/10.1016/j.envint.2024.108553.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S. M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., and Zhang, X. Y.: Recommendations for reporting black carbon measurements, Atmos Chem Phys, 13, 8365–8379, https://doi.org/10.5194/acp-13-8365-2013, 2013.

USEPA: Code federal regulations - Title 40: Protection of environment Part 86, 2010.

Wang H, Wu Y, Zhang KM, Zhang S, Baldauf RW, Snow R, Deshmukh P, Zheng X, He L, Hao J. Evaluating mobile monitoring of on-road emission factors by comparing concurrent PEMS measurements. Sci Total Environ. 2020 Sep 20;736:139507. doi: 10.1016/j.scitotenv.2020.139507. Epub 2020 May 17. PMID: 32485371; PMCID: PMC7778828.

Wang, X., Westerdahl, D., Wu, Y., Pan, X., and Zhang, K. M.: On-road emission factor distributions of individual diesel vehicles in and around Beijing , China, Atmos Environ, 45, https://doi.org/10.1016/j.atmosenv.2010.09.014, 2011.

RESPONSE TO RC#2

AC: We appreciate RC2's time and effort in reviewing our manuscript. We agree with several of the points raised, particularly those highlighting areas where clarification or additional context can strengthen the manuscript, and we have made corresponding improvements. However, we respectfully disagree with some of the more general remarks regarding the relevance of our work, as we believe the study provides important and novel insights, especially in the context of long-term, real-world BC and NO_x emission measurements. Additionally, a few specific comments seem to reflect some misinterpretation of our methods and findings.

In the following, we respond to RC#2's comments point by point.

To make the comments and responses easier to follow, we applied the following formatting to the text: the Anonymous Referee's comments are in black font; the Authors' Response is in blue italic font. Revisions made to the manuscript in response are summarized in a table with corresponding line numbers.

RC#2: This paper summarises the findings of field campaigns that aim to measure/calculate the emission factors for NOx and BC over the period 2011 to 2023. The paper mostly focuses on how emissions have changed over this period, which is of potential interest to the scientific community. The results broadly reflect those already established in numerous emission measurements across Europe, with little new insight.

AC: We agree that the trends in vehicle emissions over the past decade are of broad interest, particularly in terms of how declared reductions compare to real-world performance. This study demonstrates how real-world NO_x and BC emissions have evolved, using the plume-chasing method applied consistently to a representative sample of in-use vehicles. While the general trends in NO_x reduction are in line with existing findings, our study provides the first real-world BC emission factors (EF) for Euro 5c, 6b, 6c, and 6d diesel vehicles, which is a significant contribution given the lack of decade-spanning BC EF data—particularly relevant for Europe's large diesel car fleet.

Although BC is not regulated as an exhaust pollutant, it represents a major portion of emitted particle mass, is a strong climate forcer, and has well-documented health impacts (Janssen et al., 2012; Bond et al., 2013). Its inclusion in the new EU Clean Air Directive 2024/2881 further highlights the need for robust real-world data. Our findings help fill this gap.

The plume chasing method has proven to be a practical, cost-effective tool for emission monitoring. While it has been extensively applied in China for HDVs (e.g., Yang, 2025, *2024; Wang et al., 2023), our study is the first to apply it across a decade to a random, mixed fleet in Europe, covering both diesel and gasoline vehicles and including cars, which are not represented in works in China or the US.*

Despite the inherent variability of the method, the results are consistent and align well with broader policy changes, including the introduction of diesel particulate filters (DPFs), selective catalytic reduction (SCR), and post-"Dieselgate" recalls. This consistency underscores the method's utility for ongoing monitoring and screening for high emitters. Super-emitters were shown to be an important contributor to ambient concentrations. Ježek et al. (2018) demonstrated that excluding super-emitters is a more effective measure to reduce traffic emissions than excluding older vehicles, showing that these high-emitting vehicles are not only older vehicles but may also be newer vehicles.

Moreover, this study emphasizes the value of such real-world measurements not only for regulatory and modelling purposes, but also for informing emission control strategies globally, especially in regions lacking similar data. By presenting evidence of policy effectiveness through real-world results, we aim to support both the scientific understanding and public trust in emission regulation.

Added text in lines 79-87	Although BC is not regulated as an exhaust pollutant, it represents 48% of the emitted particle mass (Landis et al., 2007), is a strong climate forcer, and has well-documented health impacts (Bond et al., 2013; Janssen et al., 2012). Traffic emissions are the second-largest contributor to BC emissions (González Ortiz et al., 2020). The inclusion of BC in the new EU Clean Air Directive 2024/2881 (European Parliament, 2024) further highlights the need for robust real-world data. Our findings help fill this gap. The on-road chasing method has thus far been extensively used in China to monitor the effectiveness of the heavy goods vehicles' emissions (Wang et al., 2023, 2015, 2011, 2012; Yang et al., 2025, 2024; Zeng et al., 2024). Ježek et al. (2015a) reported the first BC emission factors (EF) for diesel cars in Europe. In this study, we updated the BC EF for diesel-powered cars with previously unpublished Euro 5b-6d standards
Added text in lines 297-301	The results from the 2017 campaign, Euro V, Euro IV, and older (median of all ~ 20 g kg^{-1}, average ~ 24 g kg^{-1}) match the results from Yang et al. (2025) China V vehicles (19.8 – 23.2 g kg^{-1}). Still, our Euro VI NO_x EF (median in 2023 and 2017 campaigns were 2.4 and 4.05 g kg^{-1}, respectively, and averages 7.76 and 10.73 g kg^{-1}, respectively) are lower than China VI reported by Yang et al (2025), which were 14.1 and 17.4 g kg^{-1} for medium and heavy-duty trucks, respectively.
Added references	Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M.,

Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, Journal of Geophysical Research: Atmospheres, 118, 5380–5552, https://doi.org/10.1002/jgrd.50171, 2013.

Janssen, N. A. H., Gerlof-Nijland, M. E., Lanki, T., O, S. R., Cassee, F., Gerard, H., Fischer, P., Brunekreef, B., and Krzyzanowski, M.: Health effects of Black Carbon, WHO Regional Office for Europe, Bonn, 2012.

González Ortiz, A., Guerreiro, C., Soares, J., Antognazza, F., Gsella, A., Houssiau, M., Liberti, L., Lükewille, A., Öztürk, E., Horálek, J., Banyuls, L., TargaJaume, Solberg, S., Schneider, P., Walker, S.-E., Rouïl, L., Raux, B., Peuch, V.-H., and Barré, J.: Air quality in Europe - 2020 report, https://doi.org/doi:10.2800/786656, 2020.

The European Parliament: Directive EU 2024/2881, 2024.
Wang, H., Zhang, S., Wu, X., Wen, Y., Li, Z., and Wu, Y.: Emission Measurements on a Large Sample of Heavy-Duty Diesel Trucks in China by Using Mobile Plume Chasing, Environ Sci Technol, 57, 15153–15161, https://doi.org/10.1021/ACS.EST.3C03028/SUPPL_FILE/ES3C03028_SI_001.PDF, 2023.

Wang, J. M., Jeong, C.-H., Zimmerman, N., Healy, R. M., Wang, D. K., Ke, F., and Evans, G. J.: Plume-based analysis of vehicle fleet air pollutant emissions and the contribution from high emitters, Atmospheric Measurement Techniques Discussions, 8, 2881–2912, https://doi.org/10.5194/amtd-8-2881-2015, 2015.

Wang, X., Westerdahl, D., Wu, Y., Pan, X., and Zhang, K. M.: On-road emission factor distributions of individual diesel vehicles in and around Beijing , China, Atmos Environ, 45, https://doi.org/10.1016/j.atmosenv.2010.09.014, 2011.

Wang, X., Westerdahl, D., Hu, J., Wu, Y., Yin, H., Pan, X., and Zhang, K. M.: On-road diesel vehicle emission factors for nitrogen oxides and black carbon in two Chinese cities, Atmos Environ, 46, 45–55, https://doi.org/10.1016/j.atmosenv.2011.10.033, 2012.

Yang, J., Che, X., Tan, J., Qin, X., Duan, J., Liu, D., Duan, Y., Xiang, S., Shen, N., Zhai, X., Zhang, Y., Ning, Z., and Li, L.: Real-world emission characteristics and driving factors of diesel trucks: Insights from plume chasing experiments, Atmos Environ X, 25, 100311, https://doi.org/10.1016/J.AEAOA.2025.100311, 2025.

	Yang, P., Wang, H., Wu, X., Xiao, S., Zheng, X., You, Y., Zhang, S., and Wu, Y.: Long-term plume-chasing measurements: Emission characteristics and spatial patterns of heavy-duty trucks in a megacity, Environmental Pollution, 361, https://doi.org/10.1016/J.ENVPOL.2024.124819, 2024.

RC#2: The paper is lacking in the literature cited. There is a considerable literature available on this topic, which already captures the main findings of this paper. I feel that if the authors had considered more literature they would have focused their paper differently i.e. not summarising what we already know. As it stands, this paper provides an overall summary of emissions but does not present compelling new evidence of wider interest to the community. I have suggested major revision for this (and other) reasons.

AC: We have expanded the literature cited according to specific points raised by RC1 and RC2. While there have been studies on NO_x emissions using different methods, this is the first time the chasing method has been used as a monitoring tool over a decade on diesel-powered cars, gasoline-cars, and goods vehicles. With this study, we show the utility of the chasing method for measuring car emissions, measure the changes over more than a decade, and for the first time report the real-world BC EF of the car fleet, which includes Euro5-Euro6 standard vehicles. According to other points raised, we have adjusted the text to make these points clearer, namely, we have added the importance of BC emission measurements in the introduction (see previous point for specifics) and adjusted the abstract (see next point for specifics).

The following literature was added:

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, Journal of Geophysical Research: Atmospheres, 118, 5380–5552, https://doi.org/10.1002/jgrd.50171, 2013.

Chen, Y., Sun, R., and Borken-Kleefeld, J.: On-Road NO xand Smoke Emissions of Diesel Light Commercial Vehicles-Combining Remote Sensing Measurements from across Europe, Environ Sci Technol, 54, 11744–11752, https://doi.org/10.1021/ACS.EST.9B07856/SUPPL_FILE/ES9B07856_SI_001.PDF, 2020.

Carslaw, D. C., Farren, N. J., Vaughan, A. R., Drysdale, W. S., Young, S., and Lee, J. D.: The diminishing importance of nitrogen dioxide emissions from road vehicle exhaust, Atmos Environ X, 1, 100002, https://doi.org/10.1016/J.AEAOA.2018.100002, 2019.

Dallmann, T. R., Demartini, S. J., Kirchstetter, T. W., Herndon, S. C., Onasch, T. B., Wood, E. C., and Harley, R. A.: On-road measurement of gas and particle phase pollutant emission factors for individual heavy-duty diesel trucks, Environ Sci Technol, 46, 8511–8518, 2012

Dallmann, T. R., Harley, R. A., and Kirchstetter, T. W.: Effects of diesel particle filter retrofits and accelerated fleet turnover on drayage truck emissions at the port of Oakland, Environ Sci Technol, 45, 10773–10779, 2011.

González Ortiz, A., Guerreiro, C., Soares, J., Antognazza, F., Gsella, A., Houssiau, M., Liberti, L., Lükewille, A., Öztürk, E., Horálek, J., Banyuls, L., TargaJaume, Solberg, S., Schneider, P., Walker, S.-E., Rouïl, L., Raux, B., Peuch, V.-H., and Barré, J.: Air quality in Europe - 2020 report, https://doi.org/doi:10.2800/786656, 2020.

Grange, S. K., Farren, N. J., Vaughan, A. R., Rose, R. A., & Carslaw, D. C. (2019). Strong Temperature Dependence for Light-Duty Diesel Vehicle NOx Emissions. Environmental Science and Technology, 53(11), 6587–6596. https://doi.org/10.1021/acs.est.9b01024

Janssen, N. A. H., Gerlof-Nijland, M. E., Lanki, T., O, S. R., Cassee, F., Gerard, H., Fischer, P., Brunekreef, B., and Krzyzanowski, M.: Health effects of Black Carbon, WHO Regional Office for Europe, Bonn, 2012.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S. M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., and Zhang, X. Y.: Recommendations for reporting black carbon measurements, Atmos Chem Phys, 13, 8365–8379, https://doi.org/10.5194/acp-13-8365-2013, 2013.

The European Parliament: Directive EU 2024/2881, 2024.

USEPA: Code federal regulations - Title 40: Protection of environment Part 86, 2010.

Wang, H., Zhang, S., Wu, X., Wen, Y., Li, Z., and Wu, Y.: Emission Measurements on a Large Sample of Heavy-Duty Diesel Trucks in China by Using Mobile Plume Chasing, Environ Sci Technol, 57, 15153–15161, https://doi.org/10.1021/ACS.EST.3C03028/SUPPL_FILE/ES3C03028_SI_001.PDF, 2023.

Wang, J. M., Jeong, C.-H., Zimmerman, N., Healy, R. M., Wang, D. K., Ke, F., and Evans, G. J.: Plume-based analysis of vehicle fleet air pollutant emissions and the contribution from high emitters, Atmospheric Measurement Techniques Discussions, 8, 2881–2912, https://doi.org/10.5194/amtd-8-2881-2015, 2015.

Wang, X., Westerdahl, D., Hu, J., Wu, Y., Yin, H., Pan, X., and Zhang, K. M.: On-road diesel vehicle emission factors for nitrogen oxides and black carbon in two Chinese cities, Atmos Environ, 46, 45–55, https://doi.org/10.1016/j.atmosenv.2011.10.033, 2012.

Wang, X., Westerdahl, D., Wu, Y., Pan, X., and Zhang, K. M.: On-road emission factor distributions of individual diesel vehicles in and around Beijing , China, Atmos Environ, 45, https://doi.org/10.1016/j.atmosenv.2010.09.014, 2011.

Yang, J., Che, X., Tan, J., Qin, X., Duan, J., Liu, D., Duan, Y., Xiang, S., Shen, N., Zhai, X., Zhang, Y., Ning, Z., and Li, L.: Real-world emission characteristics and driving factors of diesel trucks: Insights from plume chasing experiments, Atmos Environ X, 25, 100311, https://doi.org/10.1016/J.AEAOA.2025.100311, 2025.

Yang, P., Wang, H., Wu, X., Xiao, S., Zheng, X., You, Y., Zhang, S., and Wu, Y.: Long-term plume-chasing measurements: Emission characteristics and spatial patterns of heavy-duty trucks in a megacity, Environmental Pollution, 361, https://doi.org/10.1016/J.ENVPOL.2024.124819, 2024

RC#2: The Abstract is rather general and could be made stronger by being more quantitative e.g. how much have NOx emissions reduced through RDE testing, or the extend to which BC has reduced due to DPFs.

AC: We have changed the abstract to include more specific results of this study: namely the % of reduction in BC EF achieved after DPF were introduced with Euro5 and after RDE were implemented with Euro 6c and d, the % of reduction achieved with RDE tests, and the results of higher contribution of super emitters contribution to total vehicle fleet over the decade.

Abstract changed. Lines 10 - 24	This study compares the results of three on-road chasing campaigns conducted in 2011, 2017, and 2023 to assess the direct impact of control technologies and the indirect effect of the regulations on the real-world on-road emissions of diesel- and gasoline-powered vehicles. The findings from these campaigns are consistent, highlighting the effectiveness of installed Diesel Particulate Filters (DPF) in reducing black carbon (BC) emission factors (EF) of diesel-powered cars by 88% compared to pre-DPF Euro 4 diesel-powered cars. These reduce the diesel car EF to those at the level of gasoline-powered cars, as intended with the legislation. The results demonstrate the success of Real Driving Emissions (RDE) regulations in lowering nitrogen oxide (NO_x) emissions by 86% compared to pre-RDE Euro 6b, using technologies that were previously existing but underutilized. By presenting evidence of policy effectiveness through real-world results, we aim to support both scientific understanding and public trust in emission regulation. Using the Lorenz curves to evaluate super emitters' contribution to

	total emissions, we found that over the decade, the curves became more skewed for BC emissions, with 10% of diesel cars with the highest BC EF contributing 36%, 51%, and 65% of total diesel cars' emissions in 2011, 2017, and 2023 respectively. For NO_x emissions, the top 10% most polluting diesel-powered cars contributed 22%, 28%, and 29% of total NO_x emissions in the same years. Despite the inherent variability of the method, the results are consistent over the three campaigns and align well with broader policy changes, thus proving that the independent chasing method is a valuable screening tool for identifying super-emitters and serves as an effective monitoring tool for the entire vehicle fleet.

RC#2: There aspects of the data and methods that require further attention.

The field campaigns were conducted in very different conditions - especially the 2011 data; summarised in Table 1. the 2011 data was collected in very low ambient temperatures (-1.3 to -9.3) compared with 2023 (11.9 to 18.1). There are therefore very likely strong differences in emissions between these two years simply because of the conditions. Emissions of NOx (and BC) were likely much higher in 2011 for two reasons: older fleet compared with 2017 and 2023 and much lower temperatures. Such low temperatures are known to affect vehicle NOx emissions e.g. see

Grange, S. K., Farren, N. J., Vaughan, A. R., Rose, R. A., & Carslaw, D. C. (2019). Strong Temperature Dependence for Light-Duty Diesel Vehicle NOx Emissions. *Environmental Science and Technology*, *53*(11), 6587–6596. https://doi.org/10.1021/acs.est.9b01024

Suarez-Bertoa, R., & Astorga, C. (2018). Impact of cold temperature on Euro 6 passenger car emissions. *Environmental Pollution*, *234*, 318–329. https://doi.org/10.1016/j.envpol.2017.10.096

Wærsted, E. G., Sundvor, I., Denby, B. R., & Mu, Q. (2022). Quantification of temperature dependence of NOx emissions from road traffic in Norway using air quality modelling and monitoring data. *Atmospheric Environment: X*, *13*. https://doi.org/10.1016/j.aeaoa.2022.100160

The emissions for NOx and BC are therefore very likely higher in 2011 than they would have been if the temperatures were similar to the later campaigns - but the effect is unknown. However, there is an opportunity to investigate this important aspect of emissions.

AC: While temperature is an important influence on vehicle emissions, we disagree with these comments. The temperature range in 2011 was not -1.3 to -9.3°C as RC2 stated, *but -1.3 to + 9.3°C, and only gasoline cars showed higher emissions in the 2011 campaign; diesel vehicles were in the same range for all three measurement campaigns. This is because the temperatures were on average ~14° in 2023, ~11° in 2017, and ~4° in 2011. The temperature difference was not as large as the one reported in Wærsted et al. (2022), where they were comparing results with temperature differences exceeding 14°C to -7 °C and -13°C. Grange et al. (2019) did not show that gasoline vehicles' NO_x emissions would be affected by the temperature, and in the average temperature range in which we conducted the measurements, there was the least variability shown (12.5 ~ 15 g kg^{-1}) for all diesel cars. We have amended the text to clarify this.*

Changed text in lines 224-226	The discrepancies observed for gasoline cars in the 2011 campaign are likely due to the smaller sample size. Different weather conditions may have influenced the emissions, although not significantly, since Grange et al. (2019) did not show temperature dependency on gasoline cars NO_x emissions.
Added reference	Grange, S. K., Farren, N. J., Vaughan, A. R., Rose, R. A., & Carslaw, D. C. (2019). Strong Temperature Dependence for Light-Duty Diesel Vehicle NOx Emissions. *Environmental Science and Technology*, *53*(11), 6587–6596. https://doi.org/10.1021/acs.est.9b01024

RC2: Figure 2 needs an x-axis label. I also think it could be plotted differently with the x-axis as Euro standard (not year of manufacture). This would make it much easier to contrast the emissions by Euro standard. It would also be worth considering plotting the mean and the 95% confidence interval in the mean to help demonstrate whether there are significant differences between the Euro classes. I don't think considering the interquartile range adds much and it makes it difficult to see the main changes.

AC: We have added the x-axis label with Euro classes; however, we have kept the year of first registration to retain the timeline of introduction of emission standards, since they were not rolled out periodically, and there was a gap between cars and goods vehicles.

The reason for using the median and the interquartile ranges is to exclude the high influence the super-emitters have on the mean, and show how the distributions of the Euro classes are behaving. Since the EF distributions are not normal Gaussian and are not symmetrical, the median is a more robust parameter to describe the central tendency of a distribution than the mean. The large variability in the vehicles' EF is represented by the first and third quartiles. The skewness of the data may be seen in Figure 1, where in some cases the mean overlaps with the 75^{th} percentile - i.e., BC EF for diesel cars and goods vehicles in 2023. Since we are not making any assumptions about the underlying statistics of the distribution, we are using the median, the first and third quartiles to describe the locality, spread, and skewness of each group's EF distribution. While most of the remote sensing studies use the average, we have added a table summarizing the average values for each vehicle subgroup in the supplementary material. Comparing the values, we can see that high emitters influence the mean the most when the BC EF decreased after Euro 5 introduction. The median is more than 41% lower than the mean. In NO_x EF, we can see that the two values were similar in 2017 (5 - 27% difference), while in 2023, we saw larger discrepancies between the two values after Euro 6b (30% or higher).

Added text in lines 214-221	To describe the locality, spread, and skewness of each group's EF distribution, we used the median, the first and third quartiles. Since the EF distributions are not normal Gaussian and are not symmetrical, the median is a more robust parameter to describe the central tendency of a distribution than the mean. The large variability in the vehicles' EF is represented by the first and third quartiles. The skewness of the data may be seen in Figure 1, where in some cases the mean overlaps with the 75th percentile - i.e., BC EF for diesel cars and goods vehicles in 2023. Since we are not making any assumptions about the underlying statistics of the distribution, we are using the median, the first and third quartiles to describe the locality, spread, and skewness of each group's EF distribution. Figure 2 illustrates the comparison for each subgroup across the three campaigns.
Added text in lines 228-230	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 tables 3a and 4a, respectively, while the averages for each subgroup are in Supplement tables 3b and 4 b, respectively.
Added text in lines 258 - 263	Carslaw et al. (2019), who also used the remote sensing method, reported slightly higher diesel-powered cars' NO_x EF, 9.1 g kg^{-1} for Euro 6 and 17.2 g kg^{-1} for Euro 5 in the United Kingdom. Compared to the averages of our fleet, we find that their results were similar or slightly higher. While measuring with different methods in a similar temperature range, we attribute the difference to the difference between the Slovenian and the United Kingdom fleets, which would be consistent with Chen et al. (2020), who showed that the United Kingdom fleet generally had higher NO_x EF than the fleets in Spain, Switzerland, and Sweden.
Added references	Carslaw, D. C., Farren, N. J., Vaughan, A. R., Drysdale, W. S., Young, S., and Lee, J. D.: The diminishing importance of nitrogen dioxide emissions from road vehicle exhaust, Atmos Environ X, 1, 100002, https://doi.org/10.1016/J.AEAOA.2018.100002, 2019. Chen, Y., Sun, R., and Borken-Kleefeld, J.: On-Road NOx and Smoke Emissions of Diesel Light Commercial Vehicles-Combining Remote Sensing Measurements from across Europe, Environ Sci Technol, 54, 11744–11752,

	https://doi.org/10.1021/ACS.EST.9B07856/SUPPL_FILE/ES9B07856_SI_001.PDF, 2020.

RC2: Figure 3 looks somewhat questionable if I understand it correctly. Why consider median and not mean emissions? It would help if a different symbol was used for each Euro class. It might also be useful to split Euro 6 into pre RDE (b/c) and RDE (d-temp/d) given that there were large changes in emissions through Euro 6 from lab-based factors to on-road measurements and limits.

AC: Figure 3 is related to using the median and the interquartile ranges to describe locality, spread, and skewness of each subgroup's EF distribution (as explained in the answer just above), thus exploring the effect of the legislation on fleet EF distribution. We wanted to demonstrate that even though the NOx EF do not seem to have improved much, we could consistently detect improvement (reduction) of the 25^{th} percentile, and some improvement in the median values of Euro 6b after the recall (if we compare the trends from the 2017 and the 2023 campaign) . The improvement in the EF distribution due to these vehicles' lower emissions is diminished by high-polluting vehicles, namely the 75^{th} percentile, which does not show a significant change.

We specifically state that the figure does not include Euro 6 c and d, exactly because of the large changes in the testing procedure.

Changed text in lines 264-265	Our measurements showed that gasoline-powered cars' emissions were much lower compared to diesel-powered cars until the introduction of RDE tests.
Changed text in 268-275	While examining the impact of legislation on the measured fleet EF distribution, we observed that, although the median NOx EF values did not show substantial improvement, there was a consistent reduction in the 25th percentile. Additionally, some improvement was noted in the median values of Euro 6b vehicles following the recall, as evidenced when comparing trends from the 2017 and 2023 campaigns. However, the positive shift in EF distribution driven by lower-emission vehicles is offset by the persistence of high-polluting vehicles, particularly those at the 75th percentile, which exhibit no significant change. Despite the relative stability of median NOx EF values among pre-RDE diesel-powered cars, a reduction in emissions can still be observed through the lowering of the 25th percentile. To evaluate the reductions in diesel cars'

	NO_x EF fleet measured in real driving conditions, reflected the legislation at all, ...
Changed Figure 2. The scale was customized to the legislative values, and additional labels were added to clarify the Emission standard.	--

RC2: In the Conclusions it is stated: "...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." This may be true but it is a conclusion that is not based on the work that was done e.g. tampered vehicles were not identified, nor any impacts of improper maintenance. In general the Conclusions need to be strengthened e.g. by providing a more quantitative understanding of the changes and whether the result presented challenge wider understanding of these issues.

AC: We have strengthened the conclusions with a more specific description of the observed changes, and highlighting the importance of identifying and excluding high emitters from the vehicle fleet to improve air quality.

Added text in lines 371 - 393	Our results show that the measures to reduce vehicle emissions with increasingly stricter vehicle emissions standards were reflected to some extent in the lower values of the real-world driving EF, regardless of the high variability in vehicle maintenance, environmental conditions, driving accelerations, and speeds. The effectiveness of DPF was reflected in most of the fleet, since the 75th percentile of Euro 5b was reduced to below the 25th percentile of Euro 4, reducing the median by 88%. The NO_x EF showed a gradual decrease in the cleanest 25% of the vehicles, while the highest emitters' NO_x EF remained stable until RDE tests were introduced with Euro 6c, when the NO_x EF median was reduced by 86% compared to pre-RDE Euro 6b. Despite significant technological advancements, such as the installation of DPFs and the introduction of SCR systems that have drastically reduced the NO_x emissions of diesel-powered cars and goods vehicles to levels comparable to gasoline vehicles, a small fraction of high-polluting vehicles is now the primary obstacle to substantially reducing traffic-related emissions. As demonstrated in Ježek et al. (2018), excluding high-emitting vehicles is a more efficient strategy to reduce

	traffic emissions than excluding vehicles based on older emission standards, since some super emitters are newer vehicles. With the three major vehicle groups BC and NO_x EF now being nearly the same, targeting the highest-emitting vehicles, whether they are diesel or gasoline-powered, should be the focus of any effective pollution reduction strategy. We showed how Lorentz curves for BC emissions became more skewed, with 10% of diesel cars with the highest BC EF contributing 36%, 51%, and 65% of total diesel cars' emissions in 2011, 2017, and 2023, respectively. The contributions of these high emitters remained more similar for NO_x emissions, with the top 10% most polluting diesel-powered cars contributing 22%, 28%, and 29% of total NO_x emissions in the same years. Due to the success of RDE tests in lowering diesel vehicles NO_x EF, we expect that Lorenz curves for NO_x emissions will also become more skewed in the future as most of the older cars are eliminated from the fleet.

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Author's response to Editor's comments on 15 May 2025:

Editor:

»Thank you for addressing the reviewers' comments. This article is now publishable, however I would ask that the abstract conform to ACP's style guidelines (https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html). Currently, no explicit research gap is identified here; if the authors could modify the abstract accordingly, this will be more accessible to nonspecialists. I'm hoping that this will not take too much work.«

AC: We would like to thank the Editor for the constructive input. We have rewritten the abstract to include all ACP guidelines:

Abstracts should have fewer than **250 words** and provide a concise and accessible summary of the purpose, results, and implications of the research. ACP expects that abstracts will normally include the following components:

1. The **topic** of the article and why it is important;
2. The **status** of scientific understanding;
3. The **gap** in knowledge being addressed;
4. The **objectives**, questions, or hypotheses of the study;
5. The **approach** such as modelling, measurements, machine learning, etc.;
6. The main **results** with important quantitative information, if appropriate;
7. The **importance** and implications of the results.

The abstract now reads as:

Real-world vehicle emission measurement methods were developed to bridge the gap between laboratory tests and actual on-road emissions, including the variability in vehicles, drivers, and environmental conditions. The on-road chasing method is a cost-effective approach, capable of capturing emissions from numerous vehicles. While it has been extensively used to measure truck emissions, it has not been systematically applied to diesel- and gasoline-powered cars. This study addresses that gap by comparing data from three on-road chasing campaigns conducted in 2011, 2017, and 2023 to evaluate the impact of emissions control technologies and regulatory policies on diesel- and gasoline-powered vehicles. Results show that Diesel Particulate Filters (DPFs) reduce black carbon (BC) emission factors by 88% compared to pre-DPF Euro 4 diesel-powered cars, while Real Driving Emissions (RDE) regulations lower nitrogen oxides (NO_x) emissions by 86% relative to pre-RDE Euro 6b diesel-powered cars. This is the first study to apply the chasing method over a decade to a representative sample of

European vehicles and to report real-world BC emission factors for Euro 5b–6d compliant cars. Lorenz curve analysis reveals a growing influence of high-emitting vehicles ("super-emitters") on total fleet emissions, suggesting that targeting super-emitters could yield substantial reductions in traffic-related pollution. Despite methodological variability, the results are consistent across campaigns and align with major regulatory milestones, thus confirming the chasing method's utility as a robust tool for super-emitter identification and fleet-wide emissions monitoring. By providing real-world evidence of regulatory effectiveness, this work supports scientific understanding and confidence in emissions policy.