ELSEVIER

Contents lists available at ScienceDirect

# Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv



# Research Paper

# Identification of high emitting heavy duty vehicles using Plume Chasing: European case study for enforcement

Christina Schmidt a,b, Denis Pöhler a,b, Markus Knoll b, Yoann Bernard b, Thomas Frateur, Norbert E. Ligterink e,1, Michal Vojtíšek f,g, Alexander Bergmann, Ulrich Platt a

- <sup>a</sup> Institute of Environmental Physics, Heidelberg University, INF 229, Heidelberg, 69120, Germany
- <sup>b</sup> Airyx GmbH, Hans-Bunte-Str. 4, Heidelberg, 69123, Germany
- c Institute of Electrical Measurement and Sensor Systems, Graz University of Technology, Inffeldgasse 33/I, Graz, 8010, Austria
- d ICCT International Council on Clean Transportation, Fasanenstr. 85, Berlin, 10623, Germany
- <sup>e</sup> Department of Sustainable Transport and Logistics, TNO, Anna van Buerenplein 1, The Hague, 2595 DA, The Netherlands
- f Department of Vehicles and Ground Transport, Czech University of Life Sciences, Kamycka 129, Praha-Suchdol, 165 21, Czech Republic
- EDepartment of Automotive, Combustion Engine and Railway Engineering, Czech Technical University of Prague, Technicka 4, Prague, 166 07, Czech Republic

#### ARTICLE INFO

Editor: Paylos Kassomenos

Keywords:
Remote emission sensing
Plume Chasing
High-emitter detection
Real world driving emissions
Vehicle emission screening
Air pollution

#### ABSTRACT

The real-world emissions of almost 5000 heavy-duty vehicles (HDVs), determined by the Plume Chasing method in different European countries from 2016 to 2023, are presented. NO, emission factors are provided for all assessed vehicles, while solid particle number (PN) and black carbon (BC) emission factors are reported for over 1100 HDVs. A real-time processing software, used in most campaigns, classified vehicles as suspect or high emitters according to Euro emission standards, usually in less than a minute. Several of the identified suspicious and high emitters were stopped and inspected by authorities, revealing causes of the high emissions such as tampering, defects, or cold SCR. The inspections provided further evidence of the merits of the Plume Chasing method to authorities for a reliable pre-identification of malfunctions of the emission systems. We found that average emissions are highly dependent on the country, the national emissions enforcement and the year of measurement. In 2023, in Slovakia and Czechia the share of suspicious and high emitters was substantial at 34% and 41% respectively, while in Germany, only 8% of HDVs were identified as such. Looking back to 2019, around 28 % of HDVs measured on German highways were identified as suspicious or high emitters. In countries with strict regulations and more inspections, such as Denmark, the proportion is lower than in other countries over the same period (16% in 2020). The impact of high emitters on the overall fleet performance was assessed and we show that high emitters are responsible for more than a doubling of the average HDV NO<sub>v</sub> emissions, while for particulate matter emissions the impact of high emitters is even greater. 4-14 % of the dirtiest vehicles are responsible for 50 % of the BC and PN emissions.

# 1. Introduction

Nitrogen Oxides ( $\mathrm{NO_x}$ ) and Particulate Matter (PM) emissions from vehicles are still a major cause of poor air quality in urban areas. Both pollutants are known to have adverse effects on human health (World Health Organization, 2021; Oberdörster et al., 2005; Brook et al., 2010). In order to improve air quality, vehicles around the world must comply with increasingly stringent emission regulations.

The emissions per vehicle are regulated in Europe by the Euro emission standard, which is updated at regular intervals by European Union directives that provide for the gradual introduction of increasingly stringent standards. Euro VI is currently in force for new heavy duty vehicle (HDV) registrations. For example, the type approval limits for Euro V and Euro VI HDVs are  $2000\,\mathrm{mg\,kWh^{-1}}$  and  $460\,\mathrm{mg\,kWh^{-1}}$  for  $\mathrm{NO_x}$  respectively. To comply with emission standards, vehicles are equipped with emission abatement technologies such as Exhaust Gas Re-circulation (EGR), Selective Catalytic Reduction (SCR), Lean  $\mathrm{NO_x}$  Traps (LNTs), Diesel Particulate Filters (DPFs), Diesel Oxidation Catalysts (DOC) and ammonia slip catalysts. Throughout their lifespan, the vehicle emission system may not be properly maintained, defective

<sup>\*</sup> Corresponding author at: Institute of Environmental Physics, Heidelberg University, INF 229, Heidelberg, 69120, Germany. E-mail address: cschmidt@iup.uni-heidelberg.de (C. Schmidt).

 $<sup>^{\</sup>rm 1}\,$  currently at Dutch Human Environment and Transport Inspectorate.

parts are not correctly repaired/left unrepaired, or may be intentionally tampered with, resulting in significantly higher emissions than allowed.

Different tampering strategies are known and affect the concentrations of pollutants emitted from the tailpipe. One tampering type targets to stop AdBlue® usage by means of an AdBlue® emulator. AdBlue®, also called Diesel Exhaust Fluid, is injected before the SCR to reduce NO<sub>x</sub>. Some tampering strategies target complete shutdown of EGR and SCR and even removal of SCR and DPF (van den Meiracker and Vermeulen, 2020a,b). For the scenario with an AdBlue® emulator, EGR would still reduce NO<sub>x</sub>. Vermeulen et al. (2021), van den Meiracker and Vermeulen (2020b) observed for two tested HDVs a range between 5000 and 6000 mg kWh<sup>-1</sup>. However, 3500 mg kWh<sup>-1</sup> are also achievable with EGR-only systems, as is the case for Euro IV HDVs.  $NO_x$  emissions can become somewhat higher about  $7000 \,\mathrm{mg}\,\mathrm{kWh}^{-1}$ according to Majewski (2023) in case of the 'SCR only' concept where no EGR is used (Scania and Iveco). For the scenario of ECU (Electronic Control Unit) flashing, where aside from SCR also EGR can be deactivated, NO<sub>x</sub> levels could reach level of engine out, typically over 12 000 mg kWh<sup>-1</sup> (Majewski, 2023; van den Meiracker and Vermeulen, 2020b). These defective vehicles therefore have emissions that are often several times higher than those of vehicles with properly functioning emission reduction systems and, while representing a small fraction of the fleet, produce a major share of the total fleet emissions (Vojtisek-Lom et al., 2020; Boveroux et al., 2019). The detection and subsequent repair or de-registration of these vehicles should be a main goal of efforts to significantly improve air quality.

Conventional methods of measuring compliance over the lifetime of a vehicle during the Periodical Technical Inspection (PTI) do not yet cover the  $\mathrm{NO}_{\mathrm{x}}$  emissions due to difficulties in achieving operational temperature of the SCR at standstill and realizing a sufficient engine load in a garage test (Franzetti et al., 2023, 2024). For particles, solid particle number (PN) inspections during PTI for HDV's have been introduced in three European countries in the last two years and high emitting vehicles have been successfully identified, but PTI is mostly applied annually and can potentially be circumvented. Additional monitoring of emissions during normal driving is required to gain insight into the emission patterns of the fleet. Other methods, such as real driving emission tests (RDE) with a portable emission measurement system (PEMS), are too time consuming and expensive to be applied to many vehicles.

A more efficient and cost-effective way to measure emissions over the lifetime of a large number of vehicles is through Remote Emission Sensing (RES). RES offers the ability to measure vehicle emissions without direct installation on the target vehicle and even without being noticed by the persons inside the target vehicle. RES techniques include open-path spectroscopy measuring across the road (Bishop et al., 1989), the Point Sampling (PS) technique, where the diluted exhaust of the passing vehicles is sampled and analyzed by stationary instruments (Hansen and Rosen, 1990; Hak et al., 2009), and the here applied Plume Chasing technique, where an instrumented vehicle chases a target vehicle and analyses its diluted exhaust (Vogt et al., 2003; Pirjola et al., 2004; Canagaratna et al., 2004). These techniques and tools have been further developed in recent years. Examples include projects such as the EU project CARES (City Air Remote Emission Sensing) (EU Horizon, 2020; Farren et al., 2023; Knoll et al., 2024a; Schmidt et al., 2025) and contributions from different research groups all over the world during the last three decades (Leinonen et al., 2023; Tong et al., 2022; Vojtisek-Lom et al., 2020; Ježek et al., 2015; Lau et al., 2015; Kwak et al., 2014; Dallmann et al., 2013; Wang et al., 2012; Liggio et al., 2012; Ban-Weiss et al., 2009; Zavala et al., 2006).

Plume Chasing has a smaller sample size compared to other RES techniques and requires additional resources, such as a vehicle. However, its ability to collect measurements from individual vehicles over an extended period of time is what sets Plume Chasing apart. The longer and more accurate measurement makes the identification of high emitters more precise and reliable (avoiding false positives), which is

highly relevant for authority use. Plume Chasing captures emissions under typical driving conditions with a warm engine, largely avoiding problematic emission scenarios that occur at low speeds or during acceleration. Additionally, unlike cross-road RES, Plume Chasing functions effectively in almost all weather conditions, including rain. The Plume Chasing system can be easily integrated into an inspection vehicle operated by a trained police officer/authority, minimizing the need for extra resources beyond the measurement instruments. Furthermore, unlike stationary RES techniques, Plume Chasing is mobile and can be quickly relocated, making it difficult for drivers to evade detection by e.g. altering their route or slowing down at the measurement point. A number of validation studies (Janssen and Hagberg, 2020; Roth, 2018; Farren et al., 2022; Schmidt et al., 2025) have demonstrated the high accuracy and reliability of Plume Chasing through excellent agreement with PEMS and SEMS (Smart Emission Measurement System).

Using the Plume Chasing method, we determined on-road emission factors for 4969 HDVs. The introduced system features an automated measurement setup with a short preparation time (< 10 min), enabling rapid deployment. Its simple hardware design can be installed in any vehicle and powered from the vehicle's 12 V system, ensuring ease of use. The accompanying software performs real-time automatic emission calculation and classification, eliminating the need for post-processing. This setup is capable of measuring emissions from typically up to 200 HDVs per day, making the Plume Chasing method a practical and effective solution for large-scale, real-world emission studies. We also compare our results with the RES techniques PS (Knoll et al., 2024a; Zhou et al., 2020) and open-path RES (OPUS RSE) (Hooftman et al., 2020; Cha and Sjödin, 2022; Bernard et al., 2023) as well as with other Plume Chasing studies (Olin et al., 2023; Vojtisek-Lom et al., 2020; Lau et al., 2015).

## 2. Methods

#### 2.1. Measurement setup

The Plume Chasing method uses a measurement vehicle equipped with instruments that can include different gas and particle analyzers in addition to  $\mathrm{CO}_2$  to measure the emissions from vehicles (Fig. 1). Two different Plume Chasing setups were used in the campaigns.

For most campaigns, a simple setup with only an ICAD (Iterative CAvity enhanced DOAS (Differential Optical Absorption Spectroscopy, Platt and Stutz 2008)) instrument in Plume Chasing configuration was used for  $\mathrm{NO_x}$  high-emitter identification of HDVs (Fig. 1). The ICAD measures  $\mathrm{NO_x}$ ,  $\mathrm{NO_2}$ ,  $\mathrm{CO_2}$  as well as a GPS for locational information (Airyx GmbH) (Horbanski et al., 2019). This simple setup has the advantage of being easy for authorities to use.

In recent campaigns (Table S2), the simple measurement system was extended to measure the particle metrics solid  $PN_{23}$  and black carbon (BC). Solid  $PN_{23}$  includes only non-volatile particles larger than  $23\,\mathrm{nm}$ . Only the solid fraction of  $PN_{23}$  is measured under the Euro emissions legislation to improve the reproducibility and repeatability of the measurement (Giechaskiel et al., 2018). For solid  $PN_{23}$  measurement, a Counter (AVL DiTEST GmbH) (Schriefl et al., 2019) based on advanced diffusion charging was used in order to relate the calculated emissions to official limits of the Euro emission standard. BC was measured using a custom developed Black Carbon Tracker (Graz University of Technology) (Knoll, 2024).

The exhaust was sampled via two inlets, one at the left and one at the right front (about 150 cm apart), typically about 25 cm above the ground. Both sampling lines are merged into a central sampling line before reaching the instruments. Teflon tubing was used to transport the sample gas to the ICAD to measure  $\rm CO_2$  and  $\rm NO_x$  without observable losses. Tygon or stainless-steel tubing was used to transport the sample gas to the PM instruments. The tubing was installed through the vehicle to the measuring instruments without any sharp bends to minimize



Fig. 1. Setup of Plume Chasing. The tablet is used to display real-time data and is connected to the ICAD instrument via WIFI.

particle losses. Diffusional particle losses were less than  $3\,\%$  at  $23\,\mathrm{nm}$  (higher losses for smaller particles).

Prior to the campaigns, the instruments were calibrated. The ICAD is intrinsically calibrated using internal absorption spectra and a quantified light path resulting in an accuracy of  $\sim 2\%$ . The BCT was calibrated against a factory calibrated Aethalometer AE33 (Magee Scientific) ( $R^2 > 0.98$ ; sensitivity  $< 2~\mu g~m^{-3}$ ) using soot generated by a miniCAST (Jing Ltd., Model 6204 Type B). The Counter was calibrated against a condensation particle counter (Model 3775, TSI Incorporated) that has a counting efficiency of one in the size range of interest with NaCl generated by an atomizer (ATM220, Topas GmbH) in the size range between 23 and 200 nm (linearity (polydisperse):  $R^2 > 0.97$ ; sensitivity (polydisperse):  $< 1.000 \, \text{fm}^{-3}$ ). During the campaigns, the instruments were regularly checked for functionality by means of typical operating parameters (BCT: flow, optical power; Counter: flow, corona and electrometer self-tests, zero air self-test).

During the campaign in Czechia in 2022 a more complex scientific setup was used, which had different instruments for particle and gas measurements installed in a measurement van from TNO (Appendix, Figure S1 and Table S1). This setup was optimized in the EU Horizon 2020 project CARES and used in several validation studies (e.g. Schmidt et al., 2025).

After 2019, a new emission software from Airyx GmbH was used to indicate with a traffic light system (see Fig. 1, top center) in real-time whether the vehicle under examination was a low (green light), suspicious (yellow light) or high (red light)  $NO_x$  emitter (Airyx GmbH, 2022; Schmidt and Pöhler, 2023). This allowed simple recommendations to the authority for further inspections. The thresholds for low/suspicious/high emitters are listed in Table 1 and further discussed in Section 3.2.

### 2.2. Measurement surveys

The data were collected in 11 different measurement campaigns from 2016 to 2023 in 6 different European countries. In 2016 in Germany (G2016), in 2018 in Austria (A2018), in 2019 in Germany (G2019), in 2020 in Denmark (DK2020), in 2021 in Belgium (B2021), in 2022 in Germany (G2022) and Czechia (CZ2022) and Austria (A2022) and in 2023 in Germany (G2023), Czechia (CZ2023) and Slovakia (SK2023) (Table S2 in Appendix A). The measurements were mainly taken on highways (Fig. 2). The duration of the campaigns ranged from 2 to 18 days. Up to 163 HDVs were measured per day by a Plume Chasing vehicle. Only measurements with more than 5–10 valid data points (10 s of emission data) and where the Euro

emission standard was specified (98.5%) were included in the dataset. In total 4969 chases of HDVs are used in this study (Table S2). A detailed description of the individual measurement campaigns as well as the number of investigated vehicles and pollutants can be found in Appendix A.

#### 2.3. Data analysis

The Airyx Plume Chasing data analysis software determined  $\mathrm{NO}_x$  and  $\mathrm{NO}_2$  emission factors in real-time. For the Slovakian campaign the software was extended to additionally measure  $\mathrm{PN}_{23}$  and BC emission factors in real-time (Schmidt et al., 2023; Schmidt and Pöhler, 2023). The data of the different pollutants was time aligned. To account for the different response functions of the instruments compared to the ICAD  $\mathrm{CO}_2$  sensor, the time series data (e.g.  $\mathrm{NO}_x$ , BC) were smoothed with Gaussian filters so that the response functions were similar in time. The emission ratio  $R_E$  for a target vehicle measurement is calculated according to Eq. (1) with the average emitted concentration  $\mathrm{E}_{\mathrm{X}}$  of pollutant X (e.g.  $\mathrm{NO}_x$ ,  $\mathrm{NO}_2$ ,  $\mathrm{PN}_{23}$ , BC) and the average emitted  $\mathrm{CO}_2$  concentration  $\mathrm{E}_{\mathrm{CO}_2}$ .

$$R_E = \frac{E_X}{E_{CO_2}} = \frac{\sum_{t=0}^{T} ([X(t)] - [X_{BG}(t)])}{\sum_{t=0}^{T} ([CO_2(t)] - [CO_2BG(t)])}$$
(1)

 $CO_2$  is measured in ppm,  $NO_x$  and  $NO_2$  in ppb, PN in #/cm³ and BC in  $\mu g/m^3$ . The background (BG) values are determined as the smallest  $CO_2$  value within a time interval of  $120 \, \mathrm{s}$  before each individual measurement point at time t and its associated pollutant value X, with  $X \in (NO_x, NO_2, PN_{23}, BC)$ . For further descriptions and analysis of the used Plume Chasing technique, see Schmidt et al. (2025).

To ensure that only emissions from the plume of the chased target vehicle are taken into account, only those data points of the time series were considered valid where the  $\mathrm{CO}_2$  concentration was more than 30 ppm (campaign CZ2022, 20 ppm) above the BG. When HDVs were driving close together, a possible contamination from the HDV in front of the currently measured HDV is possible. Therefore, both HDVs were measured one after the other. If an HDV in front of another had significantly higher emissions, the HDV in the rear was discarded as contaminated and not used for further analysis (Schmidt et al., 2025).

The fuel-based emission factors of pollutant *X* can be calculated via

$$EF_{kg\ fuel}(X) = R_E \cdot \frac{\delta_x \cdot \omega_c}{M(C)}.$$
 (2)

 $\delta_X$  is dependent on the pollutant type X:  $\delta_X = M(X)$  for  $X \in (NO_X, NO_2)$  and  $\delta_X = \frac{T \cdot R}{p}$  for  $X \in (PN, PM)$  with M(X) the molar mass of pollutant

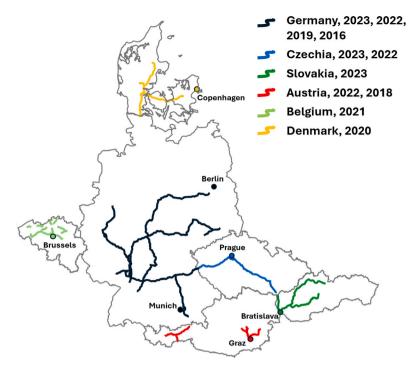


Fig. 2. Measurement locations of Plume Chasing campaigns in Germany, Czechia, Slovakia, Austria, Belgium and Denmark between 2016 and 2023.

Table 1 Different thresholds for  $NO_v$  and BC in  $mg\,kWh^{-1}$  and for  $PN_{33}$  in  $\#kWh^{-1}$  for Euro V and Euro VI HDVs.

	NO <sub>x</sub>		ВС		PN <sub>23</sub>	
	Euro V	Euro VI	Euro V	Euro VI	Euro V	Euro VI
low (up to)	≤ 2500	≤ 1200	≤ 54	≤ 18		$\leq 2.6 \cdot 10^{13}$
suspicious (up to)	≤ 3500	≤ 2200	≤ 72	≤ 24		$\leq 3.5 \cdot 10^{13}$
high (above)	> 3500	> 2200	> 72	> 24		$>3.5\cdot 10^{13}$
Euro emission limit	2000	460	PM: 30	PM: 10		$0.6 \cdot 10^{12}$
			(BC: 18)	(BC: 6)		
RDE conformity factor		1.5				1.63

X, T the instrument specific standard temperature, p the standard pressure and R the ideal gas constant. M(C) is the molar mass of carbon and  $\omega_c$  is the carbon mass fraction in fuel (Tong et al., 2022) and depends on the type of fuel. For petrol and diesel, a  $\omega_c$  of 0.86 is used in this study, for CNG, 0.71 and for LPG, 0.82 (Prussi et al., 2020). To compare emission factors to the Euro standards, the fuel-based emission factors are converted to an energy-specific emission factor via

$$EF_{kWh}(X) = EF_{kg\ fuel}(X) \cdot FC_{\text{CO}_2\ \text{WHTC}}$$
(3)

with the WHTC (World Harmonized Transient Cycle) fuel consumption  $FC_{\rm CO_2~WHTC}$  dependent on the engine displacement (Kazemi Bakhshmand et al., 2022). If the engine displacement is not given, a  $FC_{\rm CO_2~WHTC}$  of 0.211 kg fuel kWh<sup>-1</sup> was assumed (corresponding to a 40 % motor efficiency). This is close to the optimal motor efficiency and may result in slightly underestimated emission values.

#### 2.4. Thresholds for suspicious and high emitter

The emission thresholds for the classification of high and suspicious emitters are summarized in Table 1 and vary between the Euro emission standard of the HDV. The  $\mathrm{NO}_{\mathrm{X}}$  thresholds were derived and optimized from Plume Chasing emission measurements on highways combined with inspections (Pöhler, 2021). The thresholds are based on type-approval limits with safety margins, including the variability of the emission system and uncertainties due to the measurement approach, such as the influence of interfering plumes.

In order to check the compliance of  $PN_{23}$  and BC measurements with the Euro emission standards, we introduced thresholds for suspicious and high emitters similar to those for  $NO_x$ . There is no Euro emission limit for BC, only for PM mass. We introduced a threshold for BC which is 3 (suspicous) and 4 (high) times the Euro emission limit for PM mass (Table 1), assuming a BC fraction of PM mass of 60% for HDVs. Typical average BC/PM mass ratios for HDVs vehicles are between 0.4 and 0.6 (Bishop et al., 2015; Zheng et al., 2016), with higher BC/PM ratios measured when HD vehicles are under higher load, e.g. on highways (Zheng et al., 2016; Dallmann et al., 2014). For  $PN_{23}$  emissions, we chose thresholds for suspicious and high  $PN_{23}$  of approximately 27 (suspicous) and 36 (high) times the RDE Euro emission limit, respectively. In Section 3.2 the rationale behind the chosen thresholds is discussed.

# 2.5. Inspections

Inspections were carried out by authorities with varying levels of training. The type of authority involved depended on each country's regulations — specifically, who is responsible for traffic enforcement — such as police officers or traffic control authorities. The inspections distinguished between defects or faults, tampering (manipulation), software issues, cold SCR or no error. If the inspection did not distinguish between a defect/error and tampering (A2022 campaign), it is marked as 'Error (not specified)'. It is often difficult to differentiate between the two as the control electronics are often tampered with due to a defect in order to continue operating the truck without actually repairing the

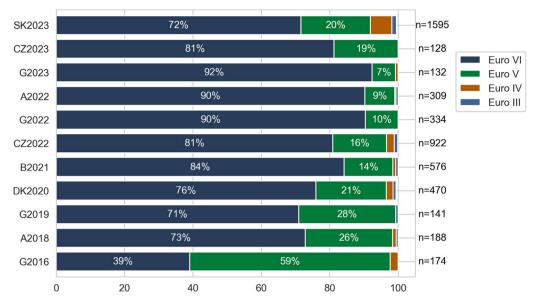


Fig. 3. Overview of Euro emission standards of investigated HDVs in the different Plume Chasing campaigns between 2016 and 2023.

defect (Pöhler, 2022a). In the other campaigns, if a defect/error and tampering were found, it was counted as tampering.

If an emulator could be found, it was evident that the truck was tampered. An emulator can be a hardware unit or software that is used to manipulate the original emission control system. It replaces or overrides signals from sensors or control units, to emulate a functioning emission control system when it is actually deactivated. For example, an AdBlue® emulator can be used to deactivate the SCR and bypass the emission control systems, resulting in increased NO<sub>x</sub> emissions. Finding an emulator requires a lot of training and knowledge of a truck's software parameters or where a hardware emulator might be hidden (e.g. under the trailer). Sometimes it takes a lot of time and a complete dismantling of the truck. This meant that it was highly dependent on the campaign, the people performing the inspection, the inspection tools used and the time available to investigate the cause of the high emissions. Therefore, the HDVs that were found to be defective/erroneous may also have been tampered and the percentage of tampered HDVs may be higher.

# 2.6. HDV fleet characteristic

In most of the campaigns, the majority of the HDVs analyzed are of the Euro VI category (>70%), followed by Euro V HDVs (Fig. 3, Table S3). The proportion of Euro VI HDVs has been increasing since 2016 in the different countries. For instance, in Germany, the fraction of Euro VI vehicles encountered rose from 39% in 2016 (G2016) to 92 % in 2023 (G2023). In Germany and Austria, the fraction of Euro VI HDVs encountered in 2023 (about 90%) was higher than in the Czech Republic in 2022 and 2023 (81 %). The high proportion of Euro VI HDVs in the A2018, G2022 and G2023 campaigns is probably due to lower tolls that apply to these vehicles and sectoral driving bans for Euro V and older vehicles that apply e.g. temporally on the route Brenner and Inntal highway. The significantly higher number of Euro V HDVs in the G2016 campaign compared to the G2019 campaign could be attributed not only to the normal replacement of vehicles over time, but also to the way Euro emission standards were assigned in the G2016 campaign. For 26% of the 174 HDVs, a Euro emission standard was assigned to the lowest possible Euro emission standard under the visible signs (e.g. HDVs with AdBlue® tanks were assigned to Euro V). As a result, Euro VI HDVs may therefore be incorrectly classified as Euro V.

The main brands of the investigated HDVs are Mercedes, Volvo, Scania, DAF and MAN, which make up more than 80% of the HDVs

(Figure S2). These brands have a relatively similar share. In Germany, Mercedes has the largest share of HDVs on the road (34% in 2023). In Denmark, the largest shares are held by Volvo and Scania (around 25% each in 2020).

The distribution of the country of origin of the HDVs investigated depends mainly on the country of investigation (Figure S3). For example, for the campaigns taking place in Germany, most of the trucks were registered in Germany (up to 50%) and surrounding countries such as Czechia or Poland; or in Czechia they were mainly Czech (about 36%), followed by trucks from surrounding countries such as Poland, Slovakia, Romania and Hungary.

#### 3. Results and discussion

#### 3.1. Emission statistics

An overview of the measured median and average  $NO_x$ ,  $NO_2/NO_x$ , BC and  $PN_{23}$  emissions from all the campaigns presented is given in Table 2. BC and  $PN_{23}$  were measured only in the most recent campaigns.

## 3.1.1. NO<sub>x</sub> emissions

Median  $\mathrm{NO_x}$  emissions were found to decrease with increasing Euro emission standard in each campaign (Fig. 4). Emissions decrease significantly from a range of  $1600-3600\,\mathrm{mg\,kWh^{-1}}$  for Euro V to  $400-900\,\mathrm{mg\,kWh^{-1}}$  for Euro VI HDVs (Table 2). This represents a reduction of approximately  $50-75\,\%$ .

In three countries (Germany, Austria, Czechia) at least two campaigns were carried out over the years. For these countries it is interesting to take a closer look at the emission trend. A decline in emissions is observed for Euro VI HDVs in Germany from 2016 to 2023 (852 to  $442\,\mathrm{mg\,kWh^{-1}}$ , see Table 2). Conversely, the median NO<sub>x</sub> emissions of Euro V HDVs exhibited an increase until 2022, followed by a decline. A comparable situation is evident in Austria between 2018 and 2022, characterized by a decline in Euro VI emissions accompanied by an increase in Euro V emissions (Table 2). This could be due to an increasing tampering rate, as well as due to deterioration of exhaust gas cleaning devices as Euro V vehicles get older. For Euro VI, the decrease seems counterintuitive at first glance, as in 2016 or 2018 all Euro VI HDVs are newer on average than in later campaigns. However, this could be explained by the fact that the Euro VI standard was made more stringent in 2019. None of the Euro VI HDVs in the G2016 or A2018

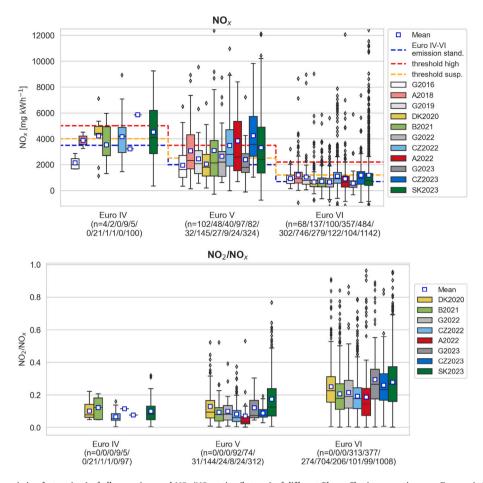


Fig. 4. Distribution of  $NO_x$  emission factors (top) of all campaigns and  $NO_2/NO_x$  ratios (bottom) of different Plume Chasing campaigns per Euro emission standard. The  $NO_x$  Euro IV to VI emission standards are marked with dashed blue lines. The thresholds for suspicious emitters are marked with dashed orange lines and the thresholds for high emitters are shown as dashed red lines. Below the Euro emission standard, the number of HDVs measured by the different campaigns is given in brackets (order as in the legend). Top whisker: 75th percentile + 1.5 interquartile range (IQR); bottom whisker: 25th percentile - 1.5 IQR; the diamonds represent the outliers.

campaign were Euro VI-D (in force from 2019). Euro VI-D HDVs have significantly lower emissions compared to Euro VI HDVs (Bernard et al., 2023) and their share has increased since 2019. The observed trends in Germany and Austria are different from those in Czechia. In Czechia, median Euro VI  $NO_x$  emissions are about the same in 2022 and 2023, while Euro V emissions are higher in 2023.

We compared  $NO_x$  emissions of HDVs between the different countries. In Denmark the average  $NO_x$  emission values for Euro V and Euro VI HDVs were among the lowest of all campaigns at  $1795\,\mathrm{mg\,kWh^{-1}}$  and  $529\,\mathrm{mg\,kWh^{-1}}$  in 2020, respectively. By contrast, the campaigns in Czechia found higher emission factors of Euro V HDVs compared to those in all the other countries that were investigated.  $NO_x$  Euro VI emissions are in Czechia in 2022 as high as Austria's or Germany's levels before 2019. In 2022, the median  $NO_x$  emissions on highways in Czechia are 27% (Euro V) and 74% (Euro VI), and in Austria 17% (Euro V) and 23% (Euro VI) higher than in Germany. In 2023, the difference is even greater when comparing emissions with measurements in Germany. The median  $NO_x$  emissions on highways in Czechia are 99% (Euro V) and 103% (Euro VI), and in Slovakia 34% (Euro V) and 75% (Euro VI) higher than in Germany.

The difference in emission levels between countries is particularly evident in the average values and less strongly pronounced in the median values. This illustrates the impact of a few high emitters, which can significantly raise the average emissions (see also Fig. 4, especially outliers of Euro VI). Thus, the different number of high emitters between the countries is probably responsible for the different averages. The number of very high emitters is particularly striking in the Slovakian campaign, where many HDVs with  $> 10\,000\,\mathrm{mg\,kWh^{-1}}$ 

were measured. These are  $\mathrm{NO}_x$  values at the level of engine out, which are an indication of ECU flashing, where EGR can also be affected in addition to SCR.

# 3.1.2. NO<sub>2</sub>/NO<sub>x</sub> emission ratios

The ratio of  $NO_2/NO_x$  (Fig. 4, bottom) was found to increase with higher Euro emission standard from on average 9.4%  $NO_2$  for Euro IV and 13.1% for Euro V to about 23.5% for Euro VI HDVs. The increase is comparable with the findings in other studies (Lau et al., 2015; Hooftman et al., 2020; Zhou et al., 2020) (see Table 3). This development is presumably due to more effective aftertreatment systems under the newer emission standards, suggesting that they reduce NO more effectively than  $NO_2$  (Zhou et al., 2020; Rodríguez et al., 2019). This shift has been a cause for concern in recent years, as  $NO_2$  is reduced less than  $NO_x$  (see also Table 2) and therefore direct exposure to noxious  $NO_2$ , as well as to ground-level ozone concentrations, is reduced less.

# 3.1.3. PM emissions

Average and median PM emissions were found to decrease with increasing Euro emission standard (Fig. 5, Table 2). The median BC emissions show a 80 % reduction in emissions from Euro V to Euro VI HDVs in the CZ2022 and SK2023 campaigns. Similarly, a 65 % reduction in  $\rm PN_{23}$  emissions was observed in the G2023 and CZ2023 campaigns. Similar to the findings for  $\rm NO_x$  emissions, PM emissions in Germany are significantly lower than in Czechia or Slovakia (PN $_{23}$ : 15–55 % for Euro V and 50–67 % for Euro VI; BC: 53 % for Euro V and 67 % for Euro VI). The reduction in BC and PN emissions from Euro V to Euro VI is due to the widespread introduction of DPFs to meet the newly introduced Euro VI PN limit.

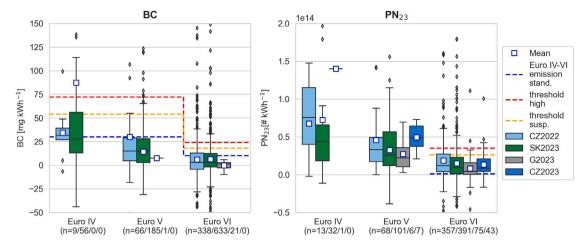


Fig. 5. BC and  $PN_{23}$  emissions of HDVs measured by Plume Chasing for different Euro emission standards in the CZ2022, SK2023, G2023 and CZ2023 campaign. The PM mass (not BC) Euro V and VI and the PN Euro VI emission standard are marked with dashed blue lines. The thresholds for suspicious emitters are marked with dashed orange lines. The thresholds for high emitters are shown as dashed red lines. Below the Euro emission standard, the number of HDVs measured by campaign is given in brackets. Top whisker: 75th percentile + 1.5 IQR; bottom whisker: 25th percentile - 1.5 IQR; the diamonds represent the outliers.

**Table 2**Overview of the median and average emission factors (in mg or # per kWh) of HDV Plume Chasing measurements segregated by Euro emissions standards and measurement

Pollutant X	Campaign	$EF_{kWh}$					
		Euro IV		Euro V		Euro VI	
		Median	Average	Median	Average	Median	Average
NO <sub>x</sub>	SK2023	4087	4511	2396	3312	772	1185
[mg kWh <sup>-1</sup> ]	CZ2023			3557	4232	941	1174
	G2023	5851	5851	1787	2386	442	567
	A2022	3213	3213	2579	3835	640	903
	CZ2022	4193	4156	2782	3484	904	1109
	G2022			2198	2641	519	657
	B2021	2703	3521	2142	3094	583	703
	DK2020	4423	4212	1795	2044	529	654
	G2019			2079	2444	786	1039
	A2018	3887	3887	2328	3069	913	1201
	G2016	2006	2134	1635	1952	852	924
NO <sub>2</sub>	SK2023	328	385	310	405	179	269
[mg kWh <sup>-1</sup> ]	CZ2023			286	326	227	269
	G2023	448	448	203	258	113	149
	A2022	367	367	106	179	51	96
	CZ2022	249	249	181	220	144	194
	G2022			146	215	90	119
	B2021	292	384	165	203	98	109
	DK2020	340	390	150	190	110	140
BC	SK2023	28	87	15	14	3	6
[mg kWh <sup>-1</sup> ]	G2023			7	7	1	0
	CZ2022	32	35	15	30	3	6
PN <sub>23</sub>	SK2023	44	72	26	33	9	15
$[# \cdot 10^{12} \text{ kWh}^{-1}]$	CZ2023			49	49	8	13
	G2023	140	140	22	27	4	8
	CZ2022	76	68	33	46	12	19

## 3.2. High-emitter identification with Plume Chasing

It was found that between 8% (G2023) and 41% (CZ2023) of the measured HDVs had high or suspicious NO<sub>x</sub> emissions (Fig. 6).

However, the percentage of high or suspicious emissions increased for Euro V HDVs (Figure S5, Appendix), indicating that this Euro emission standard had a larger share of high emitters compared to Euro VI (e.g. 33 % (G2023) or 75 % (CZ2023) for Euro V compared to 4 % (G2023) or 33 % (CZ2023) for Euro VI). The percentage of high and suspicious  $\mathrm{NO_x}$  emitters for Euro VI HDVs varied strongly between different countries and years. The campaigns in Germany, Belgium and Denmark found a smaller share for Euro VI HDVs after

**Table 3** Comparison of average  $NO_x/NO_2$  ratio of this study with selected Remote Emission Sensing and Plume Chasing literature data for different Euro emission standards.

Study	NO <sub>2</sub> /NO <sub>x</sub> [%]				
	Euro IV	Euro V	Euro VI		
SK2023	10	17	28		
CZ2023		9	26		
G2023	8	12	29		
A2022	11	7	19		
CZ2022	7	8	19		
G2022		10	21		
B2021	12	9	21		
DK2020	10	13	25		
Lau et al. (2015)	28	40			
Hooftman et al. (2020)	10.2	10.3	36.4		
Zhou et al. (2020)	2.7	6.0	22.5		

2020 compared to the pre-2020 campaigns in Austria and Germany and also to the more recent campaigns in Czechia and Slovakia (Figure S5, top, Appendix). A substantial decrease was observed in the number of high emitters in Germany. In 2019, 22 % of Euro VI HDVs were high emitters, but this went down to 11 % in 2022 and 4 % in 2023. For Euro V HDVs, the proportion remained relatively stable across different years: approx. 29-41 % for Germany, Belgium and Denmark and approx. 48-52 % for Austria and Slovakia. Czechia stands out with a relatively high proportion of high and suspicious Euro V emitters of 59 % in 2022 and 75 % in 2023.

We also found that the share of high  $NO_x$  emitters depends strongly on the country of origin of the vehicle (Appendix, Figure S6, only countries with n>10 are shown). For Euro VI HDVs, a rate of 30 % or more of high and suspicious emitters was found for vehicles originating from the countries Slovakia (SK), Romania (RO), Bulgaria (BG), Serbia (SRB), Ukraine (UA), Italy (I), Bosnia and Herzegovina (BIH) and North Macedonia (NMK). For Euro V HDVs, a rate of 50% or more was found for the countries Poland (PL), Romania (RO), Serbia (SRB) and Ukraine (UA). Notably, there was thus a larger share of suspicious and high emitters for HDVs from some countries in the southeast region of Europe.

We did only find a small dependence of high and suspicious  $NO_x$  emitters on the brand of the HDV (Appendix, Figure S7). The share was between 23 % (Mercedes) and 39 % (Iveco).

Thresholds for high  $PN_{23}$  and BC emitters have been introduced in this study (Section 2.4). The safety margins for the set thresholds used to identify suspicious and high emitters are larger for BC and  $PN_{23}$  than for  $NO_x$ . This is because, unlike  $NO_x$ , there have been no technical

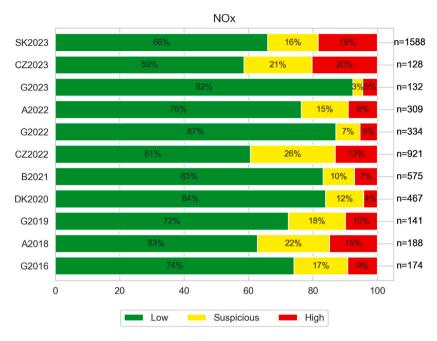


Fig. 6. Percentage of low (green), suspicious (yellow) and high (red) NO<sub>x</sub> emitters of investigated HDVs for the different measurement campaigns.

inspections to validate the BC and  $PN_{23}$  thresholds, including aspects of high deterioration of the DPF over the life-cycle. Additionally, the Euro emission standards for  $PN_{23}$  are significantly tighter than those for PM mass, necessitating higher safety margins for  $PN_{23}$  compared to BC to achieve a similar threshold. To establish  $PN_{23}$  thresholds based on the set BC thresholds, we analyzed BC and  $PN_{23}$  emission data from hundreds of HDVs in our campaigns (Fig. 12). The derived conversion factor of  $1.46 \times 10^{12} \, \text{mmg}^{-1}$  is within the range of the BC-to-PN $_{23}$  relationship found by Arndt (2013) ( $1 \times 10^{12} - 6 \times 10^{12} \, \text{mmg}^{-1}$ ). Applying this conversion factor, the e.g. Euro VI BC threshold of  $18 \, \text{mg} \, \text{kWh}^{-1}$  gives a  $PN_{23}$  threshold for suspicious emitters of  $2.6 \times 10^{13} \, \text{kWh}^{-1}$ .

With the chosen thresholds for BC and PN23 (see Section 2.4), we found about 14-26% of suspicious and high PN23 emitters in the campaigns in Slovakia and Czechia, as well as 11 % in Germany (Fig. 7, top). For BC, the share of suspicious and high emitters was around 15-18 % for Slovakia and Czechia and 0% in Germany (Fig. 7, bottom). The differentiation by Euro emission standard is shown for BC in Figure S8 in the Appendix. As there are more suspicious and high BC emitters for Euro VI HDVs compared to Euro V, this could also be due to the choice of threshold. Perhaps the Euro V thresholds of 54 and 72 mg kWh<sup>-1</sup> are too high, or the Euro VI thresholds of 18 and 24 mg kWh<sup>-1</sup> are too low. Whether high emitters of PN correlate with high emitters of BC or NO<sub>x</sub> is investigated in Section 3.4. It should be noted that for PN23 and BC emissions, the introduced thresholds for high and suspicious emitters are only assumptions. They should be further optimized with technical inspections of the measured vehicles, as has already been done for NO<sub>x</sub> emissions.

### 3.3. Authority inspections

From the suspicious and high  $\mathrm{NO_x}$  emitting HDVs a total of 141 vehicles were inspected by the authorities within the campaigns performed in the different countries (Slovakia 2023, Austria 2022, Czechia 2022, Belgium 2021 and Denmark 2020) (Fig. 8, left). Approximately 40 % of the inspected vehicles in Slovakia, Austria and Czechia were Euro V and 60 % Euro VI HDVs. In Belgium and Denmark the inspected HDVs were around 60 % Euro V and 40 % Euro VI (Figure S9). The results of the inspections to determine the cause of the high emissions are shown in Fig. 9.

For the DK2020, the CZ2022 and SK2023 campaign, a reason for the high emissions was identified for all inspected vehicles (see Fig. 9).

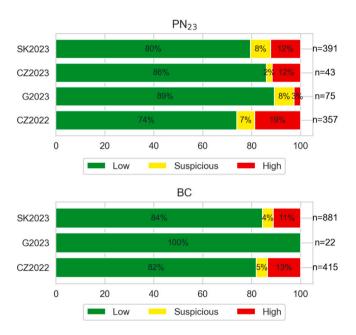


Fig. 7. Percentage of low (green), suspicious (yellow) and high (red)  $PN_{23}$  (top) and BC (bottom) emitters of investigated HDVs for the different measurement campaigns.



**Fig. 8.** Inspections of HDVs by Czech police identified as high  $NO_x$  emitters using the Plume Chasing method (left); example of a commercially available emulator found in the cable tree during the inspection of high  $NO_x$  emitters (right).

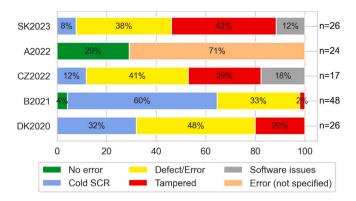


Fig. 9. Results of HDV inspections together with the authorities. The colors indicate whether 'no error' (green), a specified defect or error ('Defect/Error'; yellow), 'Software issues' (gray) such as missing mandatory software updates, a 'Cold SCR' (blue), a tampering such as an emulator ('Tampered'; red) or an unspecified error ('Error (not specified)'; orange) was found.

The inspections in CZ2022 were carried out by the Police of Czechia and by the Czech Road Transport Inspection. Martin Kristensen, a  $\mathrm{NO_x}$  Consulting staff representative and a specialist in this field, provided expert technical guidance during the inspections, especially in identifying causes of high  $\mathrm{NO_x}$  emissions and locating emissions tampering devices. Martin Kristensen was also involved in the DK2020 campaign.

Both the A2022 and B2021 campaigns with less experienced inspectors highlight the need for well-trained specialists for emission inspections with specialized technical equipment: In A2022, 5 HDVs showed constantly very high emissions, most probably originating from a non-functioning SCR system, but inspections could not identify any defect. In B2021, most suspicious and high emitters were categorized by the inspectors as 'Cold SCR', meaning the SCR system was below its working temperature and therefore not reducing NO<sub>x</sub> emissions sufficiently (Majewski, 2023). This is unlikely to be the case here, especially for Euro VI (Figure S9), since the time to reach the highway measurement locations should have allowed sufficient warm-up time (Pöhler, 2022b). A significant number of HDVs classified as 'Cold SCR' had continuously high emissions, which would imply the unlikely situation that the SCR did not warm-up at all over time. This is in contrast to a cold SCR, which warms up and reduces NO<sub>x</sub> emissions over time. A more likely explanation is misclassifications during inspection, as tampering can pretend a cold SCR simply by manipulating the temperature sensor — one of the easiest tampering methods — causing a non-functioning SCR system. Inspectors require extensive experience to identify these false temperature values as tampering and not wrongly as 'Cold SCR'. Also, some of the HDVs classified as 'Cold SCR' in the B2021 campaign showed a clear warm-up period in the data. The clearly decreasing emissions can easily be observed by a trained person on the Plume Chasing time series data displayed in the chasing vehicle's cockpit. In future, it would be worth trying to use thermal cameras to independently check cold SCR systems. In B2021 two high emitting HDVs were classified as 'No error' where one had a diesel powered refrigeration unit with high emissions (not regulated), the other also showed a clear SCR warm-up where measurements were misinterpreted due to insufficient experience of inspectors. For more details on SCR function and the influence of driving conditions on its efficiency, see Appendix A (Section 6).

Overall, several emulators (i.e. tampering devices) could be found throughout the campaigns (see e.g. Fig. 8, right). A software issue was found in Volvo Euro VI trucks in the CZ2022 campaign due to a missing mandatory update of OEM software, which reveals a lack of legislation on mandatory updates if trucks are not maintained at a manufacturer workshop. It was observed that vehicles classified as high emitters were more likely to be tampered (CZ2022: 38 % and DK2020:

27%) compared to those with only suspicious emissions (CZ2022: 0% and DK2020: 10%) (Appendix, Figure S10). It was also found that the number of trucks and the proportion of high emitters were lower when fixed inspection points were used. Truck drivers are well connected and likely avoid inspections or activate the emission control system in this area. The use of Plume Chasing measurements on different roads with varying inspection locations makes it more difficult to evade controls and police inspections. It was found in the campaign in Czechia (CZ2022) that the share of HDVs with high or suspicious emissions is during the night (between 6pm and 6am) (70%) higher than during the day (39%). Police checks took place as usual during the day. This may indicate that defective or tampered HDVs are used more during night travel to avoid controls.

## 3.4. Impact of high-emitters

As inspections have demonstrated that Plume Chasing is reliable in identifying illegally defective and tampered  $\mathrm{NO}_x$  systems of HDVs, the impact of these vehicles on total emissions was statistically analyzed. Example histograms of occurrence together with the cumulative distribution for the CZ2022 campaign can be found in the Appendix (Figure S11 and S12).

The increase of HDV NO<sub>x</sub> emissions due to high and suspicious emitters is shown in Fig. 10 (Appendix, Table S4). It is assumed that these vehicles would normally have the same emission level as the average low emitters in the same Euro class. For Euro V this results in an increase of the average fleet emissions by +52 % to +149 %. Euro VI emissions increase by +23% to +121%. This shows that defective and tampered HDVs cause in many countries more than a doubling of HDV NO<sub>x</sub> emissions. Fig. 11 shows that 50% of the total NO<sub>x</sub> emissions are caused by only 13 to 24% of the most polluting HDVs. The share of emissions caused by a small proportion of HDVs is even more pronounced for PM (BC,  $PN_{23}$ ), where 4 to 14 % cause 50 % of the emissions. These findings are consistent with the study by Olin et al. (2023). They found that the most emitting 20% of vehicles contribute to over 80 % of total PN (>23 nm, including volatile particles) emissions (in our study: 56-68 %). Zhou et al. (2020) found that the highest 10%of HDVs were responsible for 65 % of total PM, 70 % of total PN (larger than 5.6 nm) and 44 % of total  $NO_x$  (in our study: 44-66 %, 35-50 % and 28-45 %, respectively).

To see whether high emissions of one pollutant correlate with high emissions of another metric, we plot BC against PN23 emission factors (Fig. 12). The color bar indicates whether the HDV is a low, suspicious or high  $\mathrm{NO}_{\mathrm{x}}$  emitter. High  $\mathrm{PN}_{23}$  or BC emissions do not correlate strongly with high  $NO_x$  emissions (see also Appendix, Figure S13), but high BC emission factors are well correlated with high PN23 emissions (Fig. 12). We found that  $75\,\%$  of the suspicious and high BC emitters are also high PN23 emitters, but only 43% are high NOx emitters. Only 47% of the suspicious and high PN23 emitters are also high BC emitters. This is generally expected. If BC emissions are high, PN emissions, if measured in the size range of BC, must also be significant. However, high PN emissions are not necessarily associated with high BC emissions, as the emissions may be dominated by organic aerosols or feature a different size distribution, e.g. cracked channels in the DPF can lead to high PN, but still relatively low PM mass (Park et al., 2023; Borken-Kleefeld et al., 2023; Bernard et al., 2021). As mentioned above, the share of BC in PM is also highly dependent on the load of the HDV (Zheng et al., 2016), resulting in a lower correlation between BC and PN, as well as in the need to consider the engine load for the BC threshold. In the case of  $\mathrm{NO_x}$ , only 32 % and 21 % of the suspicious or high NO<sub>x</sub> emitters are suspicious or high BC and PN<sub>23</sub> emitters, respectively (Appendix, Figure S13).

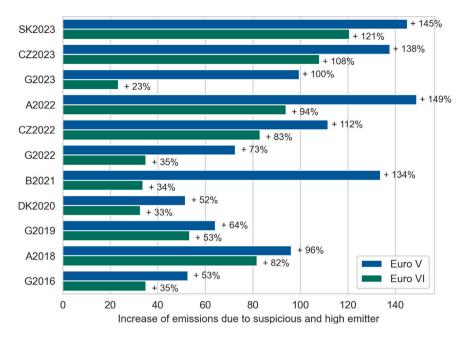


Fig. 10. Increase of average NO<sub>x</sub> emissions due to suspicious and high emitters.

Table 4
Comparison of average emission factors of this study with selected Remote Emission Sensing and Plume Chasing literature data for different Euro emission standards of diesel HDVs. Either BC or PM mass is shown (see footnotes). For the calculation from per kg fuel to per kWh a WHTC fuel consumption of 0.211 kg fuel kWh<sup>-1</sup> is used.

Study	Pollutant	$EF_{kWh}$		
		Euro IV	Euro V	Euro VI
This study Plume Chasing studies <sup>7,8</sup> PS studies <sup>1,5</sup> Open-path RES studies <sup>3,4,6</sup>	NO <sub>x</sub> [mg kWh <sup>-1</sup> ]	2134–5851 5106–6503 4178–5402 4184–6210	1952–4232 4418–4663 3249–4684 2802–4994	567–1201 1987 654–1350 574–1667
This study Plume Chasing studies <sup>2</sup> PS studies <sup>1,5</sup> Open-path RES studies	PN (·10 <sup>12</sup> ) [#kWh <sup>-1</sup> ]	64-140 <sup>c</sup> 80 <sup>e</sup> , 650 <sup>c</sup> 97 <sup>c</sup> -184 <sup>d</sup>	27-49 <sup>c</sup> 45 <sup>e</sup> , 200 <sup>c</sup> 110 <sup>c</sup> -205 <sup>d</sup>	8–19 <sup>c</sup> 5.5 <sup>e</sup> , 25 <sup>c</sup> 18 <sup>c</sup> –179 <sup>d</sup>
This study Plume Chasing studies <sup>8</sup> PS studies <sup>1,5</sup> Open-path RES studies <sup>4,6</sup>	BC, PM [mg kWh <sup>-1</sup> ]	37-87 <sup>a</sup> 253 <sup>a</sup> 36 <sup>b</sup> - 81 <sup>a</sup> 45-262 <sup>b</sup>	$7-30^{a}$ $211^{a}$ $31^{b} - 61^{a}$ $8-44^{b}$	$0-6^{a}$ $1^{b} - 9^{a}$ $-15-2^{b}$

<sup>&</sup>lt;sup>1</sup> Knoll et al. (2024a), <sup>2</sup> Olin et al. (2023), <sup>3</sup> Bernard et al. (2023), <sup>4</sup> Cha and Sjödin (2022), <sup>5</sup> Zhou et al. (2020), <sup>6</sup> Hooftman et al. (2020), <sup>7</sup> Hooftman et al. (2020), <sup>8</sup> Lau et al. (2015)

# 3.5. Comparison with other remote emission sensing studies

Table 4 compares the average emissions of selected Euro emission standards from this study with previous Plume Chasing, PS and openpath RES studies. Average BC emission factors are compared with BC emission factors (Knoll et al., 2024a; Lau et al., 2015) and with PM mass emission factors (Hooftman et al., 2020; Cha and Sjödin, 2022; Zhou et al., 2020). Average  $PN_{23}$  emission factors are compared either with  $PN_{23}$  (Knoll et al., 2024a; Olin et al., 2023) or with  $PN_{5.6}$  (particles larger than 5.6 nm) (Zhou et al., 2020). PN values from open-path RES studies are not used for the comparison as only rough estimates can be obtained from this technique (Knoll et al., 2024b). The emission values of the (Vojtisek-Lom et al., 2020) study were corrected to take into account different calculation factors (see Appendix, Section 8).

We found that the  $NO_x$  emissions of our study are in good agreement with the results of other Plume Chasing (Vojtisek-Lom et al., 2020; Lau et al., 2015), PS (Knoll et al., 2024a; Zhou et al., 2020) and open-path RES (Bernard et al., 2023; Cha and Sjödin, 2022; Hooftman et al., 2020)

studies. The wide range of emissions in this study is due to the different years and countries in which the campaigns were conducted. There is only one exception, the 2016 campaign in Germany, which shows the lowest average  $\mathrm{NO}_{x}$  values shown in this study for Euro IV and Euro V HDVs. The discrepancy is likely due to the Euro classification in this campaign, where trucks were visually classified as the lowest expected Euro class. For Euro VI HDVs, the average is highly dependent on the proportion of Euro VI-D HDVs in the fleet. These vehicles have lower emissions, close to the Euro VI standard (Bernard et al., 2023).

PN emissions for HDVs found in other Plume Chasing (Olin et al., 2023) and PS (Knoll et al., 2024a; Zhou et al., 2020) studies are higher than those from our study. One reason for this is the different particle cut-off diameters and characteristics (with/without volatile particle remover) of the PN setup used. Zhou et al. (2020) measured both, solid and total (includes both solid and volatile fraction) PN larger than 5.6 nm and is therefore expected to measure higher emissions. While the average PN23 emissions found by Olin et al. (2023) were much higher than our values, the median emissions for Euro IV and Euro V are in a similar range. This is mainly due to the fact that Olin et al. (2023) measured total PN including volatile particles, and the statistics are limited by the small number of HDVs measured (5 HDVs for the Euro IV statistics, 6 for Euro V and 13 for Euro VI). One problem with volatile or total PN measurement, particularly in Plume Chasing, is the high uncertainty of the measurement. The high dilution and temperature change after the exhaust leaves the tailpipe causes various processes such as nucleation and condensation, where high concentrations of nanoparticles are formed (Kittelson, 1998; Uhrner et al., 2007). Especially in the sub 50 nm range the volatile particles often significantly exceed the solid particle fraction. This can also be particularly pronounced in highly polluted areas, such as highways, where exhaust gas concentrations are elevated (Wehner et al., 2004). Plume Chasing setups used to measure regulated emissions or to perform high emitter identification should use solid (non-volatile) PN measurement setups. Not only because of regulation, but also because the reproducibility is significantly improved.

The BC emission factors in our study compare well with the PM mass and BC emission factors of the PS technique for Euro IV and Euro VI. For Euro V HDVs, we found about 50% lower average emission factors compared to PS studies. This may be due to differences in fleet characteristics, such as the date of first registration of the HDVs or the mileage (deterioration of the exhaust aftertreatment system). Lau

 $<sup>^{\</sup>rm a}$  BC,  $^{\rm b}$  PM,  $^{\rm c}$  PN > 23nm,  $^{\rm d}$  PN > 5.6 nm,  $^{\rm e}$  Median value.

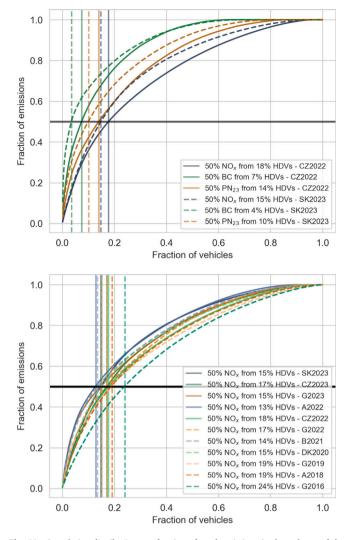


Fig. 11. Cumulative distributions — fraction of total emissions in dependence of the fraction of the vehicles ordered by emissions from highest to lowest. Negative emission values are set to zero (for a plot with negative values see Figure S14). Negative values may arise due to the methodology, especially the BG determination of the pollutants. Elevated pollution levels in the BG interval (e.g. due to other emission sources in the environment) can occasionally cause negative emission values.

et al. (2015) found much higher (3 to 15 times) BC emission factors for Euro IV and V using the Plume Chasing technique in China compared to our data. This is probably due to differences in the vehicle fleets, instruments and data analysis used in the different studies. Open-path RES studies found on average higher PM emission factors for Euro IV and V HDVs compared to our findings. Since BC is a subset of PM, this is expected. For Euro VI, the PM emissions of the open-path RES studies are lower than our findings, in parts even negative. The technique (smoke is measured by opacity and is a proxy for PM) may be too imprecise at low emission levels (Knoll, 2024; Knoll et al., 2024b).

# 4. Conclusions

This study presents the results of almost 5000 Plume Chasing measurements of HDVs from 2016 to 2023, by far the most extensive Plume Chasing study of its kind. While most previous studies on Plume Chasing looked at method improvement and on validation studies we have demonstrated the applicability of the Plume Chasing method for authorities to use with the focus on identifying non-compliant  $\mathrm{NO}_{x}$  emissions due to defects or tampering. This was implemented using an automated and user-friendly software solution which identifies high

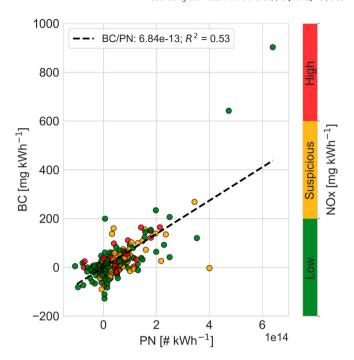


Fig. 12. The scatter plot shows a direct comparison between the  $NO_x$ ,  $PN_{23}$  and BC emission factors. The color bar indicates whether the HDV is a low, suspicious or high  $NO_x$  emitter.

emitters in real-time. The study provides an overview of HDV emissions on highways mainly for  $\mathrm{NO}_x$ , but also  $\mathrm{PN}_{23}$  and BC emissions, in different European countries and for different Euro emission standards. The average emission data from this study for  $\mathrm{NO}_x$ , BC and  $\mathrm{PN}_{23}$  are in good agreement with other HDV emission studies. Key findings and suggestions for the future use of Plume Chasing are as follows.

- A small fraction of HDVs is responsible for a large share of emissions: 50% of the total BC,  $PN_{23}$  and  $NO_x$  emissions are caused by 4 to 8 %, 10 to 14 % and 13 to 24 % of the most polluting HDVs.
- Average emissions are highly dependent on the country and the year of measurement. The share of HDVs with high  $\mathrm{NO_x}$  emissions varies from 8 % to 41 %. We found that Denmark already had a relatively low share of suspicious and high emitters in 2020 (34 % and 9 % for Euro V and VI, respectively). Denmark is one of the few countries with strong enforcement rules, high fines and many inspections. In contrast, most other European countries still lack legislation that enables the authorities to take effective action against illegal tampering. Countries without any enforcement (Slovakia or Czechia) have the highest rate of suspicious and high emitters (48–75 % and 29–34 % for Euro V and VI, respectively).
- Economic factors can also play a role in the rate of suspicious and high emitters. It is likely that old or malfunctioning vehicles in economically more affluent areas are sold to organizations in less well-off areas resulting in a higher share of these vehicles e.g. in southeastern Europe.
- Inspections proved that a cause could be found for the majority of the identified high or suspicious  $\mathrm{NO}_{\mathrm{x}}$  emitters. Most of these HDVs were found to drive illegally with a defective SCR system or with tampering. The latter often motivated by the desire to keep a defective truck in service.
- Identifying and fining high emitters more effectively would be a simple way to reduce air pollution without the need to further tighten emission standards, which can be costly for vehicle manufacturers and operators. Further it would avoid competitive advantages of non-compliant HDV operators.

- At the moment there is still a lack of  $NO_x$  and PN inspection during PTI for trucks at the EU level to identify high emitters. For  $NO_x$  this is difficult to achieve. The Plume Chasing method presented in this work is a potential solution.
- It was shown that there are gaps in the enforcement of mandatory HDV software updates when vehicles are not serviced in manufacturer-licensed workshops. A better system is needed that forces all vehicles to receive mandatory updates.
- We suggest thresholds for suspicious and high Euro V and Euro VI non-volatile  $PN_{23}$  (above 27 and 36 times the RDE Euro emission limit) and BC emitters (above 3 and 4 times the Euro emission limit for PM mass). As has already been done for  $NO_x$ , these thresholds need to be further optimized through technical inspections of the vehicles measured.
- Plume Chasing setups used to measure regulated PN emissions should use non-volatile (solid) PN measurement. For high emitter PM identification, either BC or PN can be used as the metric.

This study demonstrates that Plume Chasing is a well suited, simple tool for authorities to reliably and efficiently identify tampered and defective HDV SCR systems. This pre-selection of HDVs avoids the need for complex technical evidence of a defect or tampering on randomly selected HDVs. If suitable measurement criteria can be established to e.g. avoid false positives due to cold SCR and setting adequate emission thresholds and measurement times, Plume Chasing could achieve a 100 % hit rate in real-time high-emitter identification. This technology has the potential to become a standard screening tool, installed in the road inspection vehicles already in use, to identify HDVs with excess emissions without the need for further on-board diagnostics. This can ensure that excessive emitters are removed from the roads, e.g. through fines and subsequent repair or removal from the vehicle fleet, which has a direct impact on improving our air quality.

## CRediT authorship contribution statement

Christina Schmidt: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Conceptualization. Denis Pöhler: Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. Markus Knoll: Writing – review & editing, Validation, Investigation. Yoann Bernard: Writing – review & editing, Project administration, Funding acquisition. Thomas Frateur: Writing – review & editing, Investigation. Norbert E. Ligterink: Writing – review & editing, Resources, Project administration. Michal Vojtíšek: Writing – review & editing, Validation, Resources, Investigation. Alexander Bergmann: Writing – review & editing, Resources, Funding acquisition. Ulrich Platt: Writing – review & editing, Supervision, Resources, Funding acquisition.

# Declaration of competing interest

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

# Acknowledgments

This study was conducted under the European project City Air Remote Emission Sensing (CARES), which has received funding from the European Union's Horizon 2020 research and innovation program under grant agreement No. 814966. The Slovakian campaign was conducted on behalf of the TRUE Initiative. We acknowledge all project partners for their help during sampling and/or analysis.

## Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.scitotenv.2025.179844.

#### Data availability

Data will be made available on request.

#### References

- Airyx GmbH, 2022. Real-time on-road NOx emission measurements, datasheet. URL: https://airyx.de/wp-content/uploads/2024/07/PlumeChasing\_Brochure\_20240702.pdf.
- Arndt, M., 2013. PN to PM Correlation. URL: https://www.avl.com/documents/10138/ 0/Arndt+-+PM+vs+PN.pdf/e2fa6032-7784-496e-b47d-0842fa574e6d.
- Ban-Weiss, G.A., Lunden, M.M., Kirchstetter, T.W., Harley, R.A., 2009. Measurement of Black Carbon and Particle Number Emission Factors from Individual Heavy-Duty Trucks. Environ. Sci. Technol. 43, 1419–1424. http://dx.doi.org/10.1021/ FS8021039
- Bernard, Y., Dallmann, T., Lee, K., Rintanen, I., Tietge, U., 2021. Evaluation of Real-World Vehicle Emissions in Brussels. Technical Report, TRUE Initiative, URL: https://theicct.org/publication/evaluation-of-real-world-vehicle-emissions-in-brussels/.
- Bernard, Y., Lee, K., Nepali, R., Tietge, U., Bedogni, M., Wagner, R., Farren, N., Carslaw, D., Knoll, M., Penz, M., Bergmann, A., Pöhler, D., Schmidt, C., Juchem, H., Casadei, S., Rossi, T., Moroni, S., Torres, F.C., Pedrini, R., Belmuso, S., Palomba, P., Migliavacca, G., Mock, P., Vojitsek, M., 2023. City Air Remote Emission Sensing (CARES) EU Horizon 2020 Project: Deliverable 3.4-Summary Report on Partner Cities' Measurements Campaigns. Technical Report, EU, URL: https://cares-project.eu/wp-content/uploads/2023/06/CARES-814966-D3.4-Summary-report-on-partner-cities-measurement-campaigns.pdf.
- Bishop, G.A., Hottor-Raguindin, R., Stedman, D.H., McClintock, P., Theobald, E., Johnson, J.D., Lee, D.W., Zietsman, J., Misra, C., 2015. On-road heavy-duty vehicle emissions monitoring system. Environ. Sci. Technol. 49 (3), 1639–1645. http://dx.doi.org/10.1021/ES505534E.
- Bishop, G.A., Starkey, J.R., Ihlenfeldt, A., Williams, W.J., Stedman, D.H., 1989. Ir long-path photometry: A remote sensing tool for automobile emissions. Anal. Chem. 61 (10), 671A–677A. http://dx.doi.org/10.1021/AC00185A002.
- Borken-Kleefeld, J., Bernard, Y., Farren, N., Hallquist, A., Ligterink, N., Pöhler, D., Rushton, Christopher Knoll, M., 2023. City Air Remote Emission Sensing (CARES) EU Horizon 2020 Project: D4.8-Final Report on High-emitter and Clean Vehicle Identification. Technical Report, EU, URL: https://cares-project.eu/wp-content/ uploads/2023/06/CARES-814966-D4.8-Final-report-on-high-emitter-detection.pdf.
- Boveroux, F., Cassiers, S., Buekenhoudt, P., Chavatte, L., Meyer, P.D., 2019. Feasibility Study of a New Test Procedure to Identify High Emitters of Particulate Matter during Periodic Technical Inspection. SAE Tech. Pap. 2019-01-11, http://dx.doi. org/10.4271/2019-01-1190.
- Brook, R.D., Rajagopalan, S., Pope, C.A., Brook, J.R., Bhatnagar, A., Diez-Roux, A.V., Holguin, F., Hong, Y., Luepker, R.V., Mittleman, M.A., Peters, A., Siscovick, D., Smith, S.C., Whitsel, L., Kaufman, J.D., 2010. Particulate Matter Air Pollution and Cardiovascular Disease. Circulation 121 (21), 2331–2378. http://dx.doi.org/10.1161/CIR.0B013E3181DBECE1.
- Canagaratna, M.R., Jayne, J.T., Ghertner, D.A., Herndon, S., Shi, Q., Jimenez, J.L., Silva, P.J., Williams, P., Lanni, T., Drewnick, F., Demerjian, K.L., Kolb, C.E., Worsnop, D.R., 2004. Chase Studies of Particulate Emissions from in-use New York City Vehicles. Aerosol Sci. Technol. 38 (6), 555–573. http://dx.doi.org/10.1080/02786820490465504.
- Cha, Y., Sjödin, A., 2022. Remote Sensing Measurements of Vehicle Emissions in Sarajevo. Technical Report, IVL, No. C 734, URL: http://ivl.diva-portal.org/smash/get/diva2:1748316/FULLTEXT02.pdf.
- Dallmann, T.R., Kirchstetter, T.W., Demartini, S.J., Harley, R.A., 2013. Quantifying on-road emissions from gasoline-powered motor vehicles: Accounting for the presence of medium- and heavy-duty diesel trucks. Environ. Sci. Technol. 47 (23), 13873–13881. http://dx.doi.org/10.1021/ES402875U.
- Dallmann, T.R., Onasch, T.B., Kirchstetter, T.W., Worton, D.R., Fortner, E.C., Herndon, S.C., Wood, E.C., Franklin, J.P., Worsnop, D.R., Goldstein, A.H., Harley, R.A., 2014. Characterization of particulate matter emissions from on-road gasoline and diesel vehicles using a soot particle aerosol mass spectrometer. Atmospheric Chem. Phys. 14 (14), 7585–7599. http://dx.doi.org/10.5194/ACP-14-7585-2014.
- EU Horizon, 2020. CARES | City Air Remote Emission Sensing. In: CARES Project No. 814966. http://dx.doi.org/10.3030/814966, URL: https://cares-project.eu/.
- Farren, N., Carslaw, D., Knoll, M., Schmidt, C., Pöhler, D., Hallquist, A., 2022. City Air Remote Emission Sensing (CARES) EU Horizon 2020 Project: Deliverable 1.1 Measurement Technology Intercomparison and Evaluation. Technical Report, EU, URL: https://cares-project.eu/measurement-tech-compare-d1-1/.
- Farren, N.J., Schmidt, C., Juchem, H., Pöhler, D., Wilde, S.E., Wagner, R.L., Wilson, S., Shaw, M.D., Carslaw, D.C., 2023. Emission ratio determination from road vehicles using a range of remote emission sensing techniques. Sci. Total Environ. 875, 162621. http://dx.doi.org/10.1016/J.SCITOTENV.2023.162621.
- Franzetti, J., Selleri, T., Ferrarese, C., Melas, A., Manara, D., Giechaskiel, B., Suarez-Bertoa, R., 2023. Assessment of a NOx Measurement Procedure for Periodic Technical Inspection (PTI) of Light-Duty Diesel Vehicles. Energies 16 (14), 5520. http://dx.doi.org/10.3390/EN16145520.

- Franzetti, J., Selleri, T., Fonseca González, N., Melas, A., Gioria, R., Suarez-Bertoa, R., 2024. Measuring NOx during periodic technical inspection of diesel vehicles. Environ. Sci. Eur. 36 (175), 1–19. http://dx.doi.org/10.1186/S12302-024-01002-8.
- Giechaskiel, B., Lahde, T., Suarez-Bertoa, R., Clairotte, M., Grigoratos, T., Zardini, A., Perujo, A., Martini, G., 2018. Particle number measurements in the European legislation and future JRC activities. Combustion Engines. Combust. Engines 174 (3), 3–16. http://dx.doi.org/10.19206/CE-2018-301.
- Hak, C.S., Hallquist, M., Ljungström, E., Svane, M., Pettersson, J.B., 2009. A new approach to in-situ determination of roadside particle emission factors of individual vehicles under conventional driving conditions. Atmos. Environ. 43 (15), 2481–2488. http://dx.doi.org/10.1016/j.atmosenv.2009.01.041.
- Hansen, A.D., Rosen, H., 1990. Individual measurements of the emission factor of aerosol black carbon in automobile plumes. J. Air Waste Manage. Assoc. 40 (12), 1654–1657. http://dx.doi.org/10.1080/10473289.1990.10466812.
- Hooftman, N., Ligterink, N., Bhoraskar, A., 2020. Analysis of the 2019 Flemish Remote Sensing Campaign. Technical Report, Vlaams Planbureau voor Omgeving, Vlaamse Overheid, Departement Omgeving, URL: https://repository.tno.nl/islandora/object/ uuid%3A7e96bc14-46e3-4e5a-8f06-e436c85160f7.
- Horbanski, M., Pöhler, D., Lampel, J., Platt, U., 2019. The ICAD (iterative cavity-enhanced DOAS) method. Atmospheric Meas. Tech. 12, 3365–3381. http://dx.doi.org/10.5194/amt-12-3365-2019.
- Janssen, J., Hagberg, N., 2020. Plume Chasing A way to detect high NOx emitting vehicles. In: AVL MTC Motortestcenter AB. Technical Report, https://www.danishroadtrafficauthority.dk/Media/638350311390689301/Plume Chasing - A way to detect high NOx emitting vehicles.pdf.
- Ježek, I., Drinovec, L., Ferrero, L., Carriero, M., Močnik, G., 2015. Determination of car on-road black carbon and particle number emission factors and comparison between mobile and stationary measurements. Atmospheric Meas. Tech. 8 (1), 43–55. http://dx.doi.org/10.5194/AMT-8-43-2015.
- Kazemi Bakhshmand, S., Mulholland, E., Tietge, U., Rodríguez, F., 2022. Remote Sensing of Heavy-Duty Vehicle Emissions in Europe. International Council on Clean Transportation, URL: https://theicct.org/wp-content/uploads/2022/08/remote-sensing-hdvs-europe-aug22.pdf.
- Kittelson, D.B., 1998. Engines and nanoparticles: a review. J. Aerosol Sci. 29 (5–6), 575–588. http://dx.doi.org/10.1016/S0021-8502(97)10037-4.
- Knoll, M.F., 2024. Point Sampling as Remote Emission Sensing Method to Screen Particulate Matter Emissions (Ph.D. thesis). (June), Graz University of Technology, URL: https://repository.tugraz.at/publications/esedq-gx203.
- Knoll, M., Penz, M., Juchem, H., Schmidt, C., Pöhler, D., Bergmann, A., 2024a. Large-scale automated emission measurement of individual vehicles with point sampling. Atmos. Meas. Tech. 17, 2481–2505. http://dx.doi.org/10.5194/amt-17-2481-2024.
- Knoll, M., Penz, M., Schmidt, C., Pöhler, D., Rossi, T., Casadei, S., Bernard, Y., Hallquist, A.M., Sjödin, A., Bergmann, A., 2024b. Evaluation of the point sampling method and inter-comparison of remote emission sensing systems for screening real-world car emissions. Sci. Total Environ. 932, 171710. http://dx.doi.org/10. 1016/J.SCITOTENV.2024.171710.
- Kwak, J.H., Kim, H.S., Lee, J.H., Lee, S.H., 2014. On-road chasing measurement of exhaust particle emissions from diesel, CNG, LPG, and DME-fueled vehicles using a mobile emission laboratory. Int. J. Automot. Technol. 15 (4), 543–551. http://dx.doi.org/10.1007/S12239-014-0057-Z.
- Lau, C.F., Rakowska, A., Townsend, T., Brimblecombe, P., Chan, T.L., Yam, Y.S., Močnik, G., Ning, Z., 2015. Evaluation of diesel fleet emissions and control policies from plume chasing measurements of on-road vehicles. Atmos. Environ. 122, 171–182. http://dx.doi.org/10.1016/j.atmosenv.2015.09.048.
- Leinonen, V., Olin, M., Martikainen, S., Karjalainen, P., Mikkonen, S., 2023. Challenges and solutions in determining dilution ratios and emission factors from chase measurements of passenger vehicles. Atmos. Meas. Tech. 16, 5075–5089. http: //dx.doi.org/10.5194/amt-16-5075-2023.
- Liggio, J., Gordon, M., Smallwood, G., Li, S.M., Stroud, C., Staebler, R., Lu, G., Lee, P., Taylor, B., Brook, J.R., 2012. Are emissions of black carbon from gasoline vehicles underestimated? Insights from near and on-road measurements. Environ. Sci. Technol. 46 (9), 4819–4828. http://dx.doi.org/10.1021/ES2033845.
- Majewski, W.A., 2023. Selective Catalytic Reduction. URL: https://dieselnet.com/tech/cat\_scr.php.
- Oberdörster, G., Oberdörster, E., Oberdörster, J., 2005. Nanotoxicology: An Emerging Discipline Evolving from Studies of Ultrafine Particles. Environ. Health Perspect. 113 (7), 823–839. http://dx.doi.org/10.1289/EHP.7339.
- Olin, M., Oikarinen, H., Marjanen, P., Mikkonen, S., Karjalainen, P., 2023. High Particle Number Emissions Determined with Robust Regression Plume Analysis (RRPA) from Hundreds of Vehicle Chases. Environ. Sci. Technol. 57, 8911–8920. http://dx.doi.org/10.1021/ACS.EST.2C08198.
- Park, G., Park, S., Hwang, T., Oh, S., Lee, S., 2023. A Study on the Impact of DPF Failure on Diesel Vehicles Emissions of Particulate Matter. Appl. Sci. 13 (13), 7592. http://dx.doi.org/10.3390/APP13137592.
- Pirjola, L., Parviainen, H., Hussein, T., Valli, A., Hämeri, K., Aaalto, P., Virtanen, A., Keskinen, J., Pakkanen, T.A., Mäkelä, T., Hillamo, R.E., 2004. "Sniffer" A novel tool for chasing vehicles and measuring traffic pollutants. Atmos. Environ. 38 (22), 3625–3635. http://dx.doi.org/10.1016/j.atmosenv.2004.03.047.

- Platt, U., Stutz, J., 2008. Differential Optical Absorption Spectroscopy Principles and Applications (Physics of the Earth and Space Environments). Springer Berlin Heidelberg, p. 608, URL: https://link.springer.com/chapter/10.1007/978-3-540-75776-4 6
- Pöhler, D., 2021. Heavy Duty Vehicle (HDV) NOx Emission Measurement with Mobile Remote Sensing (Plume Chasing) and Subsequent Inspection of High Emitters, Study Performed for Danish Road Traffic Authority. Technical Report, Danish Road Traffic Authority Færdselsstyrelsen (FSTYR), https://www.danishroadtrafficauthority.dk/Media/638350311561191389/Heavy Duty Vehicle - NOx emission measurement with mobile remote sensing.pdf.
- Pöhler, D., 2022a. Detektion von hochemittierenden LKWs im fließenden Verkehr Eine Studie in der Steiermark , Österreich 2022 mit der Plume-Chasing Messmethode. Technical Report, Land Steiermark, URL: https://app.luis.steiermark.at/berichte/Download/Fachberichte/LKW\_Abgasmaipulation\_Projekt\_Airyx.pdf.
- Pöhler, D., 2022b. HDV (Heavy Duty Vehicles) NOx Emission Measurement with "Plume Chasing" and Subsequent Inspection of High Emitters A Study in Flanders (Belgium) November/December 2021, Study Performed for Flanders Environmental Agency. Technical Report, Flanders Environmental Agency, URL: <a href="https://airyx.de/wp-content/uploads/2024/02/Report\_PlumeChasing\_Belgium-Flanders2021">https://airyx.de/wp-content/uploads/2024/02/Report\_PlumeChasing\_Belgium-Flanders2021</a> v1.0.pdf.
- Prussi, M., Yugo, M., De Prada, L., Padella, M., Edwards, R., 2020. JEC Well-To-Wheels report v5. In: Report JCR, EUCAR and Concawe. Technical Report, Publications Office of the European Union, http://dx.doi.org/10.2760/100379.
- Rodríguez, F., Bernard, Y., Dornoff, J., Mock, P., 2019. Recommendations for Post-Euro 6 Standards for Light-Duty Vehicles. Technical Report, (October), International Council on Clean Transportation, pp. 1–54, URL: https://theicct.org/sites/default/ files/publications/Post\_Euro6\_standards\_report\_20191003.pdf.
- Roth, U., 2018. Optimierung und Validierung des Plume Chasing Verfahrens bei LKWs. Bachelorthesis, University of Heidelberg.
- Schmidt, C., Carslaw, D.C., Farren, N.J., Gijlswijk, N.R., Knoll, M., Ligterink, N.E., Lollinga, J.P., Pechout, M., Schmitt, S., Vojtíšek, M., Vroom, Q., Pöhler, D., 2025. Optimisation and validation of Plume Chasing for robust and automated NOx and particle vehicle emission measurements. Atmospheric Environ.: X 25, 100317. http://dx.doi.org/10.1016/j.aeaoa.2025.100317.
- Schmidt, C., Pöhler, D., 2023. City Air Remote Emission Sensing (CARES) EU Horizon 2020 Project: Deliverable 2.5: Final Version Standalone near Real-Time Plume Chase Analysis Software. Technical Report, EU, URL: https://cares-project.eu/wp-content/uploads/2023/06/CARES-814966-Deliverable-D2.5-Final-Plume-Chasing-Software.pdf.
- Schmidt, C., Pöhler, D., Bernard, Y., 2023. Emission Measurements With "Plume chasing" and Subsequent Inspection of Heavy Duty Vehicle (HDV) High Emitters A study in Slovakia September / October 2023. Technical Report, (October), The International Council on Clean Transportation, pp. 1–38, URL: https://airyx.de/wpcontent/uploads/2024/11/Report\_Slovakia\_2024\_11\_07.pdf.
- Schriefl, M.A., Bergmann, A., Fierz, M., 2019. Design principles for sensing particle number concentration and mean particle size with unipolar diffusion charging. IEEE Sensors J. 19 (4), 1392–1399. http://dx.doi.org/10.1109/JSEN.2018.2880278.
- Tong, Z., Li, Y., Lin, Q., Wang, H., Zhang, S., Wu, Y., Zhang, K.M., 2022. Uncertainty investigation of plume-chasing method for measuring on-road NOx emission factors of heavy-duty diesel vehicles. J. Hazard. Mater. 424, 127372. http://dx.doi.org/10.1016/j.jhazmat.2021.127372.
- Uhrner, U., von Löwis, S., Vehkamäki, H., Wehner, B., Bräsel, S., Hermann, M., Stratmann, F., Kulmala, M., Wiedensohler, A., 2007. Dilution and aerosol dynamics within a diesel car exhaust plume—CFD simulations of on-road measurement conditions. Atmos. Environ. 41 (35), 7440–7461. http://dx.doi.org/10.1016/J. ATMOSENV.2007.05.057.
- van den Meiracker, J., Vermeulen, R., 2020a. DIAS Smart Adaptive Remote Diagnostic Antitampering Systems EUROPEAN COMMISSION HORIZON 2020 LC-MG-1-4-2018 Grant Agreement ID: 814951 Deliverable No. D3.1, The Market of Cheating Devices and Testing Matrix with a Prioritization for Testing of Vehicle Tamp. Technical Report, EU, https://dias-project.com/sites/default/files/Deliverables/D3.1-Cheating devices and testing matrix\_0.pdf.
- van den Meiracker, J., Vermeulen, R., 2020b. DIAS Smart Adaptive Remote Diagnostic Antitampering Systems EUROPEAN COMMISSION HORIZON 2020 LC-MG-1-4-2018 Grant agreement ID: 814951 Deliverable No. D3.2, Status Quo of Critical Tampering Techniques and Proposal of Required New OBD Monitoring Functions. Technical Report, EU, https://dias-project.com/sites/default/files/Deliverables/D3.2 Status quo of critical tampering techniques and proposal of required new OBD monitoring functions.pdf.
- Vermeulen, R., Ligterink, N., Van Gijlswijk, R., Van Der Mark, P., Buskermolen, E., Verhagen, V., 2021. Dutch In-Service Emissions Testing Programme for Heavy-Duty Vehicles 2019–2020. TNO 2021 R10121. Technical Report, TNO, https://legacy.emissieregistratie.nl/erpubliek/documenten/05 Verkeer en vervoer/2021 (TNO) Dutch In-service emissions testing programme for heavy-duty vehicles 2019-2020.pdf.
- Vogt, R., Scheer, V., Casati, R., Benter, T., 2003. On-road measurement of particle emission in the exhaust plume of a diesel passenger car. Environ. Sci. Technol. 37 (18), 4070–4076. http://dx.doi.org/10.1021/ES0300315.

- Vojtisek-Lom, M., Arul Raj, A.F., Jindra, P., Macoun, D., Pechout, M., 2020. On-road detection of trucks with high NOx emissions from a patrol vehicle with on-board FTIR analyzer. Sci. Total Environ. 738, 139753. http://dx.doi.org/10.1016/J.SCITOTENV.2020.139753.
- Wang, X., Westerdahl, D., Hu, J., Wu, Y., Yin, H., Pan, X., Max Zhang, K., 2012. On-road diesel vehicle emission factors for nitrogen oxides and black carbon in two Chinese cities. Atmos. Environ. 46, 45–55. http://dx.doi.org/10.1016/J.ATMOSENV.2011. 10.033
- Wehner, B., Philippin, S., Wiedensohler, A., Scheer, V., Vogt, R., 2004. Variability of non-volatile fractions of atmospheric aerosol particles with traffic influence. Atmos. Environ. 38, 6081–6090. http://dx.doi.org/10.1016/j.atmosenv.2004.08.015.
- World Health Organization, 2021. WHO Global Air Quality Guidelines. Particulate Matter (PM2.5 and PM10), Ozone, Nitrogen Dioxide, Sulfur Dioxide and Carbon Monoxide. Technical Report, WHO, URL: https://www.who.int/publications/ i/item/9789240034228.
- Zavala, M., Herndon, S.C., Slott, R.S., Dunlea, E.J., Marr, L.C., Shorter, J.H., Zahniser, M., Knighton, W.B., Rogers, T.M., Kolb, C.E., Molina, L.T., Molina, M.J., 2006. Characterization of on-road vehicle emissions in the Mexico City Metropolitan Area using a mobile laboratory in chase and fleet average measurement modes during the MCMA-2003 field campaign. Atmos. Chem. Phys. 6, 5129–5142. http://dx.doi.org/10.5194/acp-6-5129-2006.
- Zheng, X., Wu, Y., Zhang, S., Baldauf, R.W., Zhang, K.M., Hu, J., Li, Z., Fu, L., Hao, J., 2016. Joint measurements of black carbon and particle mass for heavy-duty diesel vehicles using a portable emission measurement system. Atmos. Environ. 141, 435–442. http://dx.doi.org/10.1016/J.ATMOSENV.2016.07.013.
- Zhou, L., Liu, Q., Lee, B.P., Chan, C.K., Hallquist, A.M., Sjödin, A., Jerksjö, M., Salberg, H., Wängberg, I., Hallquist, M., Salvador, C.M., Gaita, S.M., Mellqvist, J., 2020. A transition of atmospheric emissions of particles and gases from on-road heavy-duty trucks. Atmospheric Chem. Phys. 20 (3), 1701–1722. http://dx.doi.org/10.5194/ACP-20-1701-2020.