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Towards better air quality using the plume chasing method: validation studies of real driving NO_x emission measurements of vehicles

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Abstract

The reliability of Plume Chasing as a Remote Emission Measurement Technique in detecting NO_x emissions is investigated. It allows, for example, to identify high-NO_x-emitting vehicles with high precision. During a 5-day study of the CARES project, controlled Plume Chasing measurements of different types of vehicles were performed on a test track. The test track experiments included 21 different sessions with different driving properties and different test vehicles representative for a common vehicle fleet. During the experiments, the emission control systems were activated and deactivated in a blind comparison experiment. The Plume Chasing method showed excellent correlation with the averaged reference SEMS NO_x data. The main cause for deviations was found to be situations when emissions are significantly influenced by plumes from high emitting vehicles driving ahead.

Keywords: Plume Chasing; SCR Manipulation; CARES; EU research; NOx Emissions; High emitter;

1. Introduction

NO_x emissions from vehicles are a major cause of bad air quality in urban areas. In order to improve the air quality, vehicles must comply with increasingly stringent emission values. The emissions per vehicle are regulated by the

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Nomenclature

BG	Background
CARES	City Air Remote Emission Sensing
CO_2	Carbon dioxide
CPC	Condensation Particle Counter
DPF	Diesel Particulate Filter
ERC	Emission Re-Circulation
HDV	Heavy duty vehicle
LDV	Light duty vehicle
NO_x	Nitrogen Oxide
PN	Particle Number
SCR	Selective Catalytic Reduction
SEMS	Smart Emission Measurement System
PEMS	Portable Emission Measurement System
SMPS	Scanning Mobility Particle Sizer

EURO Norm (e.g. HDVs EURO V: 2000 mg/kWh, EURO VI: 460 mg/kWh). Not all vehicles meet the EURO Norms under real driving conditions applying ERC and/or SCR technology. This can be due to an insufficient emission reduction system from the manufacturer, or due to a defect or manipulation by the owner. The detection of these vehicles should be a main goal to significantly improve air quality. Existing possibilities to measure whether the vehicles comply with the regulations over their lifetime (e.g. PEMS) are rare and costly. Within the framework of the EU H2020 project CARES (CARES, 2020), different remote emission sensing techniques and instruments are further developed and evaluated. They offer the ability to measure the emissions of vehicles without direct installation on the vehicle, and can therefore be easily applied in real environments. We investigate the Plume Chasing method and present an extensive validation study showing that this method compares very well with the SEMS / PEMS emission measurements for different types of vehicles. For HDVs several validation studies of Plume Chasing against the established PEMS have already previously shown excellent agreement between the observed emission values (Janssen & Hagberg, 2020; Roth, 2018).

2. Methods

2.1. Instrumental setup

The Plume Chasing method uses a measurement vehicle equipped with different instruments to capture the emission plume of vehicles (see Fig. 1, left). The sampled air from the diluted exhaust plume is analysed in real-time by these instruments. Assuming that the ratio between pollutant and CO_2 does not change, no matter how much the exhaust gas is diluted, the specific emissions can then be calculated from the ratio in the sampled air, e.g. NO_x to CO_2 ratio converted to NO_x emissions in g/kWh or g/km, using assumptions of the CO_2 emissions. In our study, two ICAD NO_x-CO_2 instruments (Airyx GmbH) were installed together with a LICOR CO_2 sensor and several particle instruments in a measurement vehicle from TNO (Utrecht, Netherlands). An ultrasonic anemometer measured the wind velocity and direction on the roof of the chasing vehicle, while a radar determined the distance to the chased vehicle. A dashboard camera took pictures of the chase to provide additional information for interpreting the collected data. The used instruments allow fast (1 s time resolution) and simple measurements with high accuracy (sub ppb for

Fig. 1. Sketch of Plume Chasing method (left) and Plume Chasing vehicle (right).

NO_x) and wide measurement range (0–5000 ppb for NO_x). For more details, see Table 1. Teflon tubing was used for sampling the gases, stainless steel and conductive tubing for the particles. The inlet sampling position was about 30 cm above the road and 10 cm in front of the bumper of the van (see Fig. 1, right). The left and right inlets were 150 cm apart and merged to one central sampling line before reaching the instruments. This yields a more reliable measurement of the plume, being less sensitive to the position of the exhaust on the chased vehicles and also to meteorological effects such as side winds. The inlet positions and the choice of measurement instruments were optimised in previous experiments during the CARES project.

Table 1. Instruments used for Plume Chasing.

Parameter	Instrument	Measurement range	Accuracy	Power
NO_x, NO_2, CO_2	ICAD-NOx-150DE-M	0 ... 5000 ppb	0.15 ppb (1 σ @ 30 s)	< 30 W
CO_2	LI-COR 7000	0 ... 3000 ppm	1 % nominally	< 40 W
PN (D_{50} of 2.5 nm)	TSI 3776 CPC	up to $3{\cdot}10^5$ particles/cm^3	$1/\sqrt{n}$	< 335 W
PN (d_p = 90 nm)	SMPS (Electrostatic Classifier 3082 and TSI 3775 CPC)	up to $5{\cdot}10^4$ particles/cm^3 photometric mode up to 10^7	$1/\sqrt{n}$	< 535 W
Location, Speed	Navilock Multi GNSS u-blox 8		2.5 m CEP	< 0.2 W
Number Plate	ARVOO ANPR camera (DUO12-35/25m35/25c780)			
Distance	Continental ARS300 radar	0.25 ... 200 m	1.5 % @ > 1 m	< 7 W
Wind speed, Wind direction	Vaisala WTX530 series, model 536	0 ... 60 m/s 0 ... 360 °	±3 % @ 10 m/s ±3.0 ° @ 10 m/s	< 9.7 W

2.2. Measurement procedure

In June 2021 the controlled Plume Chasing measurements of different types of vehicles (see Table 2) representative for a common vehicle fleet were performed, as part of the EU CARES project. During 5 days on the test track (see Fig. 2) of the Rijkdienst voor het Wegverkeer Test Centre Lelystad, Netherlands, different remote emission techniques were compared. The chased vehicles were equipped with SEMS and PEMS to be able to validate the measurements. In 21 different sessions each vehicle was followed by the Plume Chasing vehicle for at least one round (about 2.8 km) before switching to the next vehicle. The distance to the chased vehicle was kept constant at about 8 to 35 m, depending on the velocity of the vehicle. In the different sessions the emission control systems (SCR and DPF) of the test vehicles were activated and deactivated in a blind comparison experiment, resulting in low and high emissions. In addition, the driving conditions were changed, e.g. the velocity, the distance between the vehicles or their driving order, to investigate strengths and weaknesses of the different remote emission sensing techniques in identifying high and low emitters.

Fig. 2. Selection of test vehicles at RWE test track, Lelystad, Netherlands, Session 16, chasing vehicle at 4th position. Motorcycle and Scooter not shown in the picture.

Table 2. Vehicle information for the three test vehicles investigated in this study.

EU Category	Brand	Type	Fuel type	Euro class
N3	Ford F-Max	Truck	Diesel	VI
N1	Volkswagen	Caddy	Diesel	6
M1	Volkswagen	Transporter	Diesel	6
M1	Volkswagen	Touran	Gasoline	5J
L3	Yamaha	MT-07 Motorbike	Gasoline	5
L3	Yamaha	NMAX Scooter	Gasoline	5

2.3. Data analysis

The raw data was first cleaned from corrupted and duplicate data within the datasets, then time-shifted with the help of response time tests to account for the residence time in the tubing and the time shifts between the different instruments. To account for different instrument response functions in comparison to the ICAD CO_2 sensor, the signals were also smoothed with a Gaussian filter. In this study we compare the average NO_x/CO_2 ratios determined by Plume Chasing with the reference ratios of the SEMS. The ratio R of NO_x to CO_2 for Plume Chasing is calculated according to Eq.1,

$$R = \frac{E_{NO_x}}{E_{CO_2}} = \frac{\sum_{t=0}^{T}(NO_x(t) - NO_x^{BG}(t))}{\sum_{t=0}^{T}(CO_2(t) - CO_2^{BG}(t))} , \quad (1)$$

with the average emitted NO_x concentration E_{NOx} and the average emitted CO_2 concentration E_{CO2}. The background values of CO_2, CO_2^{BG}, and of NO_x, NO_x^{BG}, are determined from the 5 s moving average of the time series of the measured signals. The BG values are determined as the smallest CO_2 value within a time interval of 120 s before each individual measurement point and its associated NO_x value.

To ensure that only emissions from the plume of the chased vehicle are taken into account, a threshold of 20 ppm was imposed for CO_2. Only if the CO_2 concentration was more than 20 ppm above the BG, the Plume Chasing measurements were considered valid. For the same reason two speed filters were applied. The measurements were only considered valid if the Plume Chasing vehicle speed was higher than 10 km/h and if the difference between the wind speed measured at the roof of the vehicle and the vehicle speed was smaller than the vehicle speed (to ensure that the own plume is not measured). The same speed filters were also applied in the calculation of the SEMS NO_x/CO_2 ratios. For the calculation of the SEMS NO_x/CO_2 ratios, the mass flows of NO_x and CO_2 are averaged over the chasing time before building the ratios. Only CO_2 and NO_x values with an associated CO_2 level above 1 % CO_2 concentration are included in the average, since in situations with minor emissions the plume is not detectable with Plume Chasing.

3. Results and discussion

The average ambient air temperature, wind speed, wind direction and relative humidity for the different days during the measurement campaign can be found in Table 3. The temperature was similar on all measurement days (~ 16 °C), whereas the wind speed was stronger during the first two days (~ 11 m/s) than during the remaining three days (~ 7 m/s). There was no precipitation on the days of measuring and the relative humidity was about 70 to 90 %.

Table 3. Weather conditions during the measurement campaign. Averages are calculated for the time of measurements.

Date	Temperature [°C]	Wind speed [m/s]	Wind direction	Relative humidity [%]
21.06.2021	15.0	10.7	NE	89.2
22.06.2021	16.0	11.3	NE	68.6
23.06.2021	15.8	7.1	NE to SE	69.3
24.06.2021	16.2	6.1	NE to SE	70.7
25.06.2021	15.5	7.8	SW	80.1

An example time series of time-aligned NO_x and CO_2 data of both SEMS and Plume Chasing is shown in Fig. 3. The Plume Chasing vehicle was following a LDV (VW Caddy) for one round with three accelerations, two from standstill and one from 30 km/h. The red vertical line indicates the time when the point sampling location was passed.

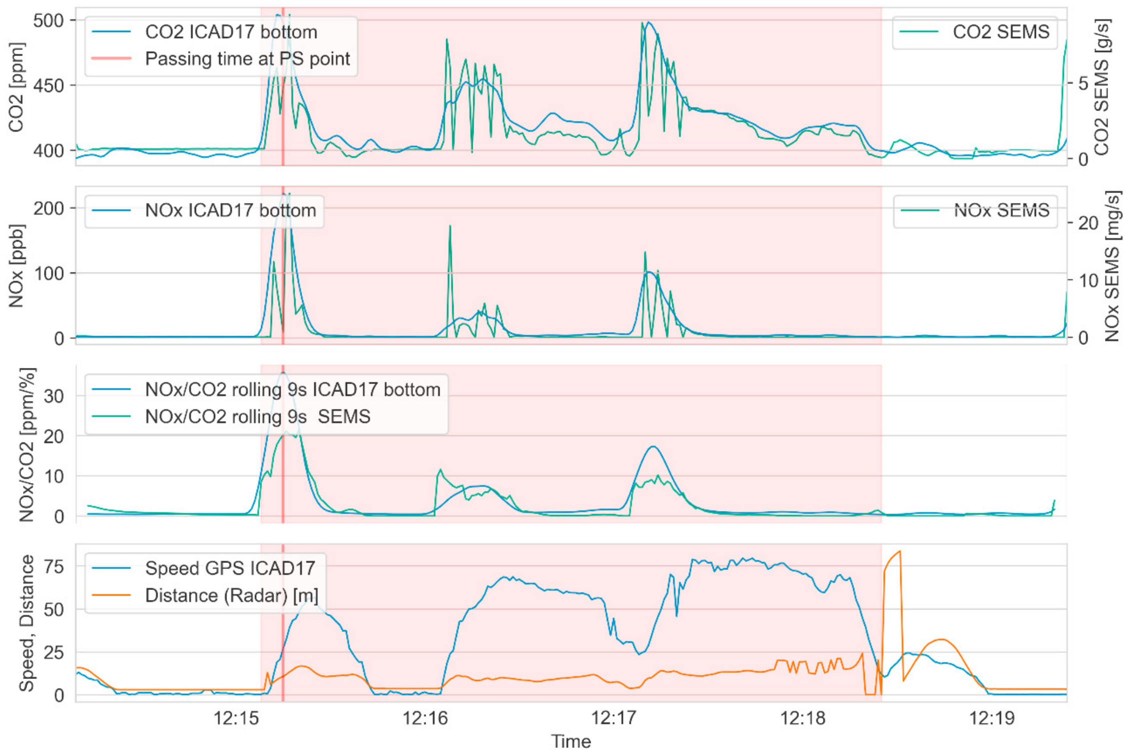

Fig. 3. Time series of NO_x, CO_2 and ratio NO_x/CO_2 of SEMS and Plume Chasing; velocity and distance to chased vehicle (VW Caddy), 24.06.2021, Session 15a.

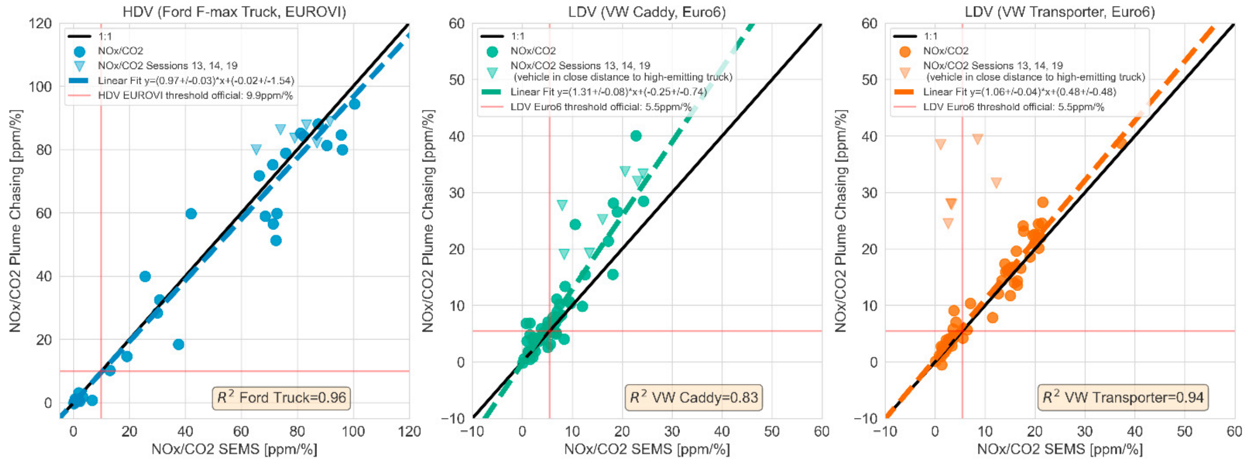

Fig. 4. Scatter plot NO_x/CO_2 Plume Chasing vs. SEMS of different vehicles; CO_2 threshold: 20 ppm, background interval: 120 s, velocity > 10 km/h, 21.–25.06.2021.

A scatter plot of the inferred NO_x/CO_2 ratios of Plume Chasing against those of SEMS is shown in Fig. 4 for the Ford truck (left), the VW Caddy (middle) and the VW Transporter (right). The Plume Chasing method shows excellent correlation with the averaged SEMS NO_x data of all three vehicles. The coefficient of determination is the highest for the truck with $R^2 = 0.96$. For the VW Transporter we find $R^2 = 0.94$ and for the VW Caddy $R^2 = 0.83$. While for the VW Transporter there is only a slight overestimation of the NO_x/CO_2 ratios by the Plume Chasing method, the overestimation for the VW Caddy is stronger.

The main cause for deviations from the reference data was found to be situations when emissions are significantly influenced by other plumes, e.g. a passenger car driving closely behind a very high-emitting truck with deactivated SCR system (Fig. 4, triangular data points of session 13, 14, 19). Possible ways to avoid interfering sources of pollutants would be to avoid busy roads for the measurements and not to measure LDVs with HDVs driving in front of them. Alternatively, a wrong classification could be avoided in this case by measuring both, the LDV and HDV.

However, the Plume Chasing method also slightly overestimated the NO_x/CO_2 ration in sessions without any high-emitting vehicle in front of the VW Caddy, i.e. without any interfering plumes. Another reason for deviations between the SEMS and Plume Chasing NO_x/CO_2 ratios could be a systematic underestimation of the NO_x/CO_2 ratios of the SEMS instrument. Deviations between SEMS and the established PEMS can reach several percent (Sato et al., 2020; Yu et al., 2021) as the SEMS uses a ZrO_2-based sensor, which has a NO_x measurement tolerance of +/− 20 ppm below 100 ppm. As the emissions of the VW Transporter are in a similar range as those of the VW Caddy, the underestimation must be specific for the SEMS installed in the Caddy or due to some vehicle properties. This could be checked in future campaigns by exchanging the SEMS between vehicles or using a PEMS.

In Fig. 4, the official HDV EURO VI and LDV Euro 6 thresholds are shown as red lines. The Plume Chasing thresholds for a tempering classification (not shown here) are about two (suspicious emitter) to three (high emitter) times higher than the official thresholds. These Plume Chasing thresholds have been optimised in previous studies in order to avoid false positive results on the one hand and still minimise false negative results on the other hand.

4. Conclusion and future works

By showing excellent correlation (R^2 between 0.83 and 0.94) with on-board SEMS measurements for a variety of vehicle types, this study further underlines the robustness and reliability of the Plume Chasing method in detecting NO_x emissions, thus allowing for example to easily identify high-emitting vehicles. While most previous studies focused only on HDVs, we have demonstrated the applicability of the Plume Chasing method also for LDVs. We also showed that interfering plumes from high emitters can lead to false positive detections and suggested strategies to avoid them.

First authorities in Europe have already started testing and implementing the Plume Chasing method for high NO_x emitter detection (Pöhler et al., 2022). For this purpose, the Plume Chasing system can be installed, for example, in regular enforcement vehicles.

In 2022 a city demonstration campaign will take place in Prague, which further evaluates the applicability of the Plume Chasing method in real urban measurement environments including potential interference from other vehicles and stationary emission sources.

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