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Farren, Naomi J. orcid.org/0000-0002-5668-1648, Schmidt, Christina, Juchem, Hannes et al. (6 more authors) (2023) Emission ratio determination from road vehicles using a range of remote emission sensing techniques. Science of the Total Environment. 162621. ISSN 0048-9697

https://doi.org/10.1016/j.scitotenv.2023.162621

Science of the Total Environment 875 (2023) 162621

Contents lists available at ScienceDirect

Science of the Total Environment

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

Emission ratio determination from road vehicles using a range of remote emission sensing techniques

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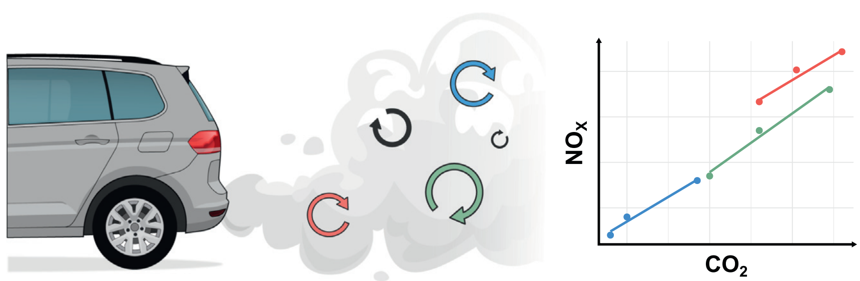
HIGHLIGHTS

- Plume dilution approach suitable for multiple remote emission sensing techniques.
- Emission ratios determined for a range of vehicles under different driving conditions.
- Effect of vehicle aftertreatment system tampering on emissions behaviour explored.
- Suitable for quantifying a suite of pollutant emission ratios in urban environments.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Philip K. Hopke

Keywords:
Vehicle emissions
Remote sensing
Plume chasing
Point sampling
Regression

ABSTRACT

The development of remote emission sensing techniques such as plume chasing and point sampling has progressed significantly and is providing new insight into vehicle emissions behaviour. However, the analysis of remote emission sensing data can be highly challenging and there is currently no standardised method available. In this study we present a single data processing approach to quantify vehicle exhaust emissions measured using a range of remote emission sensing techniques. The method uses rolling regression calculated over short time intervals to derive the characteristics of diluting plumes. We apply the method to high time-resolution plume chasing and point sampling data to quantify gaseous exhaust emission ratios from individual vehicles. Data from a series of vehicle emission characterisation experiments conducted under controlled conditions is used to demonstrate the potential of this approach. First, the method is validated through comparison with on-board emission measurements. Second, the ability of this approach to detect changes in NO_x / CO_2 ratios associated with aftertreatment system tampering and different engine operating conditions is shown. Third, the flexibility of the approach is demonstrated by varying the pollutants used as regression variables and quantifying the NO_2 / NO_x ratios for different vehicle types. A higher proportion of total NO_x is emitted as NO_2 when the selective catalytic reduction system of the measured heavy duty truck is tampered. In addition, the applicability of this approach to urban environments is illustrated using mobile measurements conducted in Milan, Italy in 2021. Emissions from local combustion sources are distinguished from a complex urban background and the spatiotemporal variability in emissions is shown. The mean NO_x / CO_2 ratio of 1.61 ppb/ppm is considered representative of the local vehicle fleet. It is envisaged that this approach can be used to quantify emissions from a range of mobile and stationary fuel combustion sources, including non-road vehicles, ships, trains, boilers and incinerators.

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http://dx.doi.org/10.1016/j.scitotenv.2023.162621
Received 27 October 2022; Received in revised form 27 February 2023; Accepted 28 February 2023
Available online 05 March 2023

1. Introduction

European emission standards were introduced in the early 1990s to regulate vehicular exhaust pollutants that have harmful health effects and negative impacts on air quality and the environment (EC, 1991; EC, 1993). Emissions legislation has since become increasingly stringent, in an attempt to progressively reduce the permissible levels of emissions. Over time, the suite of regulated pollutants has broadened and increasingly complex aftertreatment technologies have been developed, aiming to achieve compliance with the most recent Euro standards. The development of these technologies and advances in engines has considerably reduced emissions of many regulated pollutants, but there are still concerns that the reduction in emissions has not been as effective as anticipated. For example, over the last decade on-road emissions of NO_x ($NO + NO_2$) from diesel vehicles have been found to be substantially higher than emissions measured during type approval tests (Ligterink et al., 2013; Yang et al., 2015). Furthermore, modern diesel engines rely on the use of oxidation catalysts to convert NO to NO_2 to improve the combustion of soot in diesel particulate filters. Whilst this may have a positive effect on particle emissions, the NO_x emissions that remain can consist of higher proportions of NO_2, the more harmful component (Carslaw, 2005; Feng et al., 2014; He et al., 2015). There is growing evidence that the use of new aftertreatment technologies has also led to additional complexity of the emissions that remain, due to the introduction of unintended byproducts such as NH_3 and N_2O, which are currently unregulated.

To address the discrepancies between laboratory and on-road vehicle emissions, technologies to perform on-road emission measurements under real driving conditions have been developed. Traditional cross-road remote sensing is a non-intrusive technique which directs infrared and ultraviolet light through individual plumes and uses absorption spectroscopy to quantify the concentrations of different pollutants present (Burgard et al., 2006a; Burgard et al., 2006b). Cross-road remote sensing measures the molar volume ratio of pollutants to CO_2, which can be converted to fuel-specific emission factors, i.e., a measure of the mass of pollutant emitted per mass of fuel burned. Large samples of snapshot emission measurements (typically 0.5 s) can be collected in a short space of time, and aggregated to generate emission factors for different vehicle groupings, such as vehicle type, fuel type, emissions standard and vehicle manufacturer. Remote sensing has been used extensively across Europe and further afield to monitor real-world emission factors for a range of vehicle types (Bernard et al., 2019), yet there are some limitations of the current commercially available technology. For example, the instrument is limited to a select group of pollutants and the instantaneous nature of the measurements prevents detailed information on the emissions behaviour of a single vehicle under a range of conditions from being obtained.

To address these and other challenges, the development of new remote emission sensing technologies (point sampling and plume chasing) has been a key focus in recent years. Point sampling uses fast-response air quality instruments deployed in a stationary position at the side of the road to perform extractive sampling of pollutants in dispersing vehicle exhaust plumes (Hak et al., 2009; Hallquist et al., 2013; Watne et al., 2018). This approach offers the potential to broaden the suite of pollutants, as theoretically any species can be measured, provided the accuracy and response time of the instrument is sufficient to resolve the transient emissions of passing vehicles. Plume chasing uses a vehicle equipped with fast-response instrumentation to follow a vehicle of interest and measure the emissions within its dispersing plume (Pöhler, 2020; Pöhler et al., 2020). The benefit of this approach is that a specific vehicle can be monitored over several minutes, to obtain detailed emissions behaviour information over a range of driving conditions. Similar to traditional cross-road remote sensing, both technologies rely on the measurement of atmospheric ratios of pollutants to CO_2 at high time resolution, ensuring fuel-specific emission factors can be calculated for individual vehicles. Another technique that can be used to measure vehicle emission ratios is mobile monitoring (Wagner et al., 2021). Mobile monitoring differs from plume chasing in that the sample inlet is not positioned to directly capture the exhaust plume of the vehicle in front, but to sample a mixture of fresh emissions from vehicle exhaust plumes in the nearby vicinity as well as dispersed plumes from vehicles and point sources further away. This means that mobile observations are representative of a range of combustion sources (e.g. integrated fleet characteristics rather than a single specific vehicle) in addition to other non-combustion sources.

The advancement of technologies that can be used to monitor increasingly complex emission sources at high temporal and/or spatial resolution will undoubtedly produce large data sets for which the analysis is non-trivial. The use of a complementary, robust data processing method is essential for accurate data interpretation and maximising the outcomes of the analysis. Existing data processing methods for high resolution point sampling, plume chasing and mobile monitoring data often derive emission ratios by subtracting background concentrations from the average pollutant concentrations associated with a particular emission event. Ideally the background concentration would be quantified at the same location and time as the emission event, but since this is physically impossible, the background level must be estimated in some way. Several 'background subtraction' approaches have been developed to address this challenge and applied to point sampling, plume chasing, and mobile monitoring data. Examples include the calculation of low percentile concentrations within a rolling window centred around the time of the measured emission event, (Padilla et al., 2022; Schmidt et al., 2022) the application of hourly adjustment factors based on ambient concentrations from a nearby fixed-site monitor, (Apte et al., 2017) and the use of the inflection points of concentration distributions (Vojtisek-Lom et al., 2020). Several point sampling studies have also used geometric peak fitting methods to quantify emissions from individual vehicles and ships (Hak et al., 2009; Jonsson et al., 2011). Overall these approaches provide useful insight into vehicle emissions behaviour, but the definition of the background value is somewhat arbitrary. Background subtraction methods can perform well for low traffic flows or scenarios where other sources of emissions are minimal, but they are less applicable to urban settings with higher traffic flows and increased source complexity.

Plume chasing, point sampling and mobile monitoring all generate high resolution time series data, yet different data processing methods are often implemented. A single method to quantify emissions from local combustion events such as diluting vehicle exhaust plumes, that does not require background subtraction and can be applied to any of the measurement techniques, would be incredibly valuable. In this study, we present a single 'plume dilution' rolling regression approach that is first applied to plume chasing and point sampling measurements for the determination of NO_x / CO_2 and NO_2 / NO_x emission ratios for individual vehicles under real driving conditions. The use of regression allows for combustion events to be identified based on observed strong positive correlations between the pollutant and the combustion tracer. The method does not require background subtraction, which is a significant advantage over current methods. The ability of the approach to detect vehicle aftertreatment system tampering is demonstrated and the method is validated against on-board tailpipe emission measurements conducted simultaneously to the plume chasing measurements. Next, the method is applied to urban mobile monitoring data collected in the city of Milan (Italy) in 2021, to quantify the spatiotemporal variability in NO_x / CO_2 emission ratios. Overall it is envisaged that the described approach can be used for a vast range of air quality applications, providing insight into a variety of pollutant emission ratios from different combustion sources.

2. Methods

2.1. Vehicle emission characterisation experiments

The main objective of the vehicle emission characterisation experiments was to bring together newly developed instrumentation for plume chasing and point sampling with a range of commercially available measurement systems to characterise the performance of the remote emission sensing technologies. The data generated from these experiments is ideal for developing the new data processing approach presented in this study, due to the controlled conditions under which the measurements were conducted.

2.1.1. Measurement location

Controlled characterisation experiments were performed on a 2.8 km circuit test track at Rijkdienst voor het Wegverkeer Test Centre Lelystad (TCL) in the Netherlands, as part of the Horizon 2020 City Air Remote Emission Sensing (CARES) project (CARES, 2023; Farren et al., 2022a). The experiments were carried out between 21st and 25th June 2021. TCL is located approximately 15 km southeast of Lelystad (52.45701, 5.51437) in an isolated location away from major emission sources. This environment was ideal for characterisation experiments due to the stable background and lack of major emission sources in the local area. The measurements were performed in daylight hours during periods of dry weather.

2.1.2. Experimental design

A program of experiments was designed to evaluate the performance of the different remote emission sensing technologies under a wide range of driving conditions. Six test vehicles were driven around the track and an instrumented plume chase vehicle followed one of the test vehicles during each lap. Point sampling instruments and an Opus AccuScan cross-road remote sensing device were deployed on an additional test lane on the inner side of a straight section of the circuit. On each lap, the test vehicles and the plume chase vehicle diverted off the main track to drive past the point sampling instruments and remote sensing device.

Vehicle tests were divided into several sessions conducted under different driving conditions. Constant speed tests were performed at speeds of 30, 50, 80, 100 and 120 km h^{-1}. Acceleration tests were carried out, in which a convoy of test vehicles waited either 8 or 30 m before the point sampling instruments and remote sensing device, and accelerated from standstill past the instruments before continuing around the rest of the track. The drivers were asked to either accelerate normally or aggressively. These tests were defined as ‘normal’ and ‘sporty’ acceleration and the median acceleration values measured by the remote sensing device for all test vehicles were 2.0 and 3.7 m s^{-2} respectively. All of the test vehicles participated in the different sessions, with a variety of driving conditions and order of vehicles. Repeat laps for each set of test conditions were performed. There were 1435 vehicle passes through the point sampling and remote sensing instrumentation and 231 plume chases in total. A small proportion of measurements (approximately the first lap of the track for each vehicle at the start of each day) were conducted when the engine was cold. For the remaining time, the engine was warm and conditioned by the prevailing ambient conditions. A detailed record of the test conditions allows for a comparison of the cold start and warm engine emissions under the same driving conditions.

2.1.3. Test vehicles

The test vehicles used for the characterisation experiments were selected to be representative of wider vehicle fleets and the vehicle technical information is provided in Table 1. The order of the test vehicles was varied systematically for different sessions and the time delay between each vehicle was approximately 20 s for the measurements considered in this study.

Table 1
Vehicle technical information for the six test vehicles. TWC = three way catalyst. DOC = diesel oxidation catalyst. SCR = selective catalytic reduction system. DPF = diesel particulate filter.

Description	Category	Make/model	Fuel type	Euro class	Aftertreatment
Scooter	L3	Yamaha NMAX	Gasoline	5	TWC
Motorcycle	L3	Yamaha MT-07	Gasoline	5	TWC
Car	M1	VW Touran	Gasoline	5	TWC
Van 1	M1	VW Transporter	Diesel	6	DOC-SCR-DPF
Van 2	N1	VW Caddy	Diesel	6	DOC-SCR-DPF
Truck	N3	Ford F-MAX	Diesel	VI	DOC-SCR-DPF

2.1.4. Aftertreatment system tampering

The selective catalytic reduction (SCR) systems in the heavy duty truck and two light duty diesel vehicles (Van 1 and Van 2) were switched on and off periodically between sessions, to assess the ability of different measurement techniques to detect vehicle emission tampering (Farren et al., 2022b).

2.1.5. Plume chasing and point sampling

A range of both newly developed and commercially available instrumentation was used for point sampling and plume chasing, to perform fast-response remote emission sensing measurements of particles and a range of gaseous pollutants. The point sampling measurements were made on a continuous basis. A photograph of each passing vehicle was taken to capture the registration plate and a light barrier was used to measure vehicle speed for individual vehicles. The registration plate and the corresponding timestamp allowed for individual vehicles to be matched to measured point sampling emission events. The width of the test lane for the point sampling measurements was 5.75 m. The sample inlet was placed on the surface of the road. Two sample inlet positions were tested for the point sampling measurements: 0.34 m and 2.40 m inward from the edge of the test lane. The sample inlet of the plume chase vehicle was installed on the front bumper. The typical duration of the plume chases were 3–4 min, i.e., 1 lap of the test track.

2.1.6. ICAD NO_2 / NO_x /NO analyser

Plume chasing and point sampling measurements conducted using the Iterative Cavity Enhanced Differential Optical Absorption Spectrometer (ICAD) are a particular focus of this study. The ICAD was developed by Airyx and uses optical absorption spectroscopy in an optical cavity in the spectral range between ≈ 430 to 465 nm to provide a direct measurement of NO_2 (Airyx, 2023). Before a second optical cavity, NO is converted to NO_2 by gas phase titration with a NO_x -free ozone source, and a measure of total NO_x concentration is obtained. NO concentration is calculated by subtracting the NO_2 concentration measured in the first optical cavity from the total NO_x concentration. The instrument is installed with an infrared sensor for parallel measurements of CO_2. Further information on the ICAD approach is described in detail elsewhere (Horbanski et al., 2019).

The instrument offers 1 s time resolution, sub-ppb precision, and fast response time ($t_{90} < 2$ s), thus lending itself to the detection of individual dispersing vehicle exhaust plumes. The ICAD is suitable for field measurements as it is lightweight (<10 kg) and has low power requirements (<30 W at 12 V). The spectral fitting algorithm separates the absorption structure of NO_2 from overlapping absorptions such as water vapour and glyoxal. The instrument is practical to use for mobile applications since consumable gases are not required for instrument operation, it has a short warm up time, and it is insensitive to mechanical vibrations and temperature variations. Species are measured at ambient humidity as the instrument is not fitted with a sample dryer. Instrument drift is negligible (<0.1 ppb $month^{-1}$) due to regularly automated reference measurements. The NO_2 measurements are time-aligned to the corresponding NO_x and CO_2 measurements by applying a constant time offset of 3 s.

2.1.7. On-board emission measurements

On-board emission measurements provide a measure of pollutant concentrations at the point of emission, i.e., the vehicle tailpipe. Emission ratios should remain approximately constant as the plume dilutes, therefore on-board emission measurements can be useful for validating remote emission sensing measurements. On-board emission measurements for the truck, Van 1 and Van 2 were performed by installing Smart Emissions Measurement System (SEMS) equipment. The SEMS relies on a zirconium dioxide multi-layer sensor mounted on the exhaust pipe of the vehicle for the measurement of NO_x concentrations at the point of emission (Yu et al., 2021). A Global Positioning System is used to determine vehicle speed and an on-board diagnostics module is responsible for the operation of the sensors, data storage and power supply of the equipment. The on-board diagnostics data is also used to determine the CO_2 concentrations.

2.2. Urban mobile measurements

An instrumented mobile laboratory was used to conduct mobile measurements in the city of Milan, Italy. The purpose of these measurements was to demonstrate that the data processing method is not limited to the analysis of measurements collected under controlled test track conditions and that it is robust enough to be applied to data collected on busy urban roads where there is a complex mix of emission sources. Another key objective of these measurements was to demonstrate that the approach is not limited to point sampling and plume chasing techniques, and can be applied to other remote emission sensing technologies that generate high resolution time series, such as mobile monitoring.

The measurements were collected using fast-response air quality instrumentation installed in the back of a Nissan NV400SE. Ambient air is sampled from a front-facing inlet positioned 2.25 m above the ground. The setup is designed to capture a mixture of local, freshly emitted vehicle exhaust plumes in addition to more dispersed plumes from vehicles and point sources further away. Further information on the mobile laboratory can be found in in the literature (Wagner et al., 2021). In this work we consider the NO_x and CO_2 measurements obtained using an ICAD analyser.

There are a series of ring roads surrounding the city of Milan which form an expanding circle from the centre (Area C) outwards into Area B. Area B is a Low Emission Zone (LEZ) with no access for the most polluting vehicles and Area C is a combined Urban Toll Road and LEZ (Urban Access Regulations, 2022). The measurement route was designed to capture a 'slice' of the circular ring roads, to capture potential changes in vehicle fleet, congestion levels and other emission sources. Mobile measurements were conducted on 30th September and 1st October 2021 during daytime hours (09:00–15:00 local time). Seven repeat circuits were sampled; each circuit was 13.2 km in total and took approximately 1 h–1 h 15 min to complete, depending on the traffic conditions.

Latitude and longitude data was available for every NO_x and CO_2 measurement, which enabled the data to be linked to the road network (downloaded from OpenStreetMap using overpass turbo) (OpenStreetMap, 2023; Overpass turbo, 2023). The road network was divided into 30 m segments and a mean NO_x / CO_2 ratio was calculated for each segment.

2.3. Rolling regression method

2.3.1. Concept

Here we introduce the short-term rolling regression method used in this study by considering a scatter plot produced from simultaneous measurements of NO_x and CO_2 concentrations in a dispersing plume emitted from a single vehicle exhaust emission event (Fig. 1). The blue data points are generated from three consecutive measurements at 1 s time resolution. Fitting a regression line through these points and determining the value of the slope provides a NO_x / CO_2 emission ratio for the 3 s interval. The NO_x / CO_2 ratio simply represents the change in the concentration of NO_x (Δ NO_x) divided by the change in the concentration of CO_2 (Δ CO_2). The red and green data points and their associated regression lines are generated from measurements of the same plume, made at different 3 s intervals. The slopes of the three regression lines are approximately the same, which is shown visually as parallel regression lines on the scatter plot. This is expected for measurements of a dispersing plume emitted from a single vehicle exhaust emission event, as the ratio of the co-emitted species should be representative of the ratio at the point of emission, and remain constant as the plume dilutes. A 3 s interval is selected as a minimum of 3 data points are required for the regression; this takes 3 s to obtain using an instrument with 1 Hz time resolution. The interval is kept to the minimum width required, to retain as much temporal resolution and information on plume dynamics as possible.

The coefficient of determination (R^2) provides a measure of how close the data points are to the regression line. A high R^2 value means that the NO_x / CO_2 ratio remains constant within the 3 s interval, providing strong evidence that the measurements made within that time frame are from the same source. The length of the regression lines gives an indication of how much the pollutant concentrations change within a particular interval. For example the red regression line shown in Fig. 1 is shorter than the blue and green lines and is associated with smaller Δ NO_x and Δ CO_2 values. Δ CO_2 is a useful parameter since a CO_2 enhancement is required to identify a combustion event.

Generating multiple short-term, overlapping regressions from 1 Hz time series data of two co-emitted pollutants and filtering by high R^2 values provides a way to detect dispersing plumes from local combustion events and quantify the associated emission ratios. Regressions are run over short time periods and it is not necessary to account for varying background concentrations. The regressions represent an increment above local background levels and the timescale of changes in background concentrations are negligible relative to the regression timescale (3 s). Subtraction of background NO_x and CO_2 concentrations would slide the position of a regression line on the x-y coordinate plane, but the slope value (i.e. the emission ratio) would remain unchanged.

In principle, this short-term continuous regression approach is suitable for multiple pollutant combinations and a range of air quality applications. In this work, we use the method to identify dispersing plumes from local vehicle exhaust emission events. A single approach is applied to 1 Hz pollutant time series data collected using three different techniques: point sampling, plume chasing and mobile monitoring.

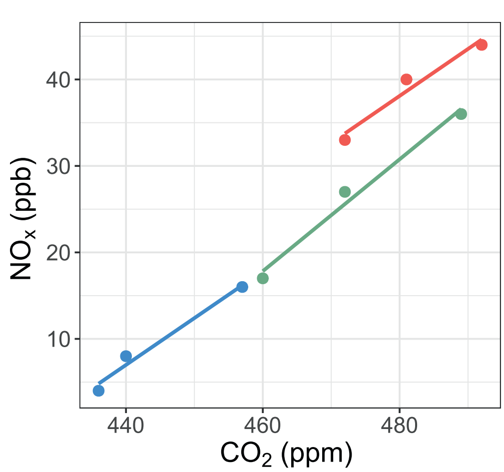

Fig. 1. NO_x vs CO_2 scatter plot generated from 1 Hz measurements of a dispersing plume emitted from a local combustion event, e.g. vehicle exhaust emissions. The different coloured data points represent measurements from three distinct 3 s intervals and the associated regression lines are shown.

2.3.2. Method description

As regression methods are sensitive to time alignment, the first step is to check that any pollutant time series data to be used as regression variables, such as 1 Hz NO_x and CO_2 measurements, are accurately time-aligned. Next, the regressions are performed for the selected variables using the *mobilemeasr* R package (Wilde, 2021). The regression lines are fit using ordinary least squares regressions and the size of the moving window (n) is set to 3 s. For the point sampling measurements, it is necessary to apply a time offset to correct for the lag time between the vehicle pass and the corresponding emission measurement. The time offset is specific to each measurement campaign and will depend on multiple factors including the length and position of the sample line and the sampling flow rate. Corresponding metadata such as vehicle pass or vehicle chase information can be merged with the regression results using the data timestamps.

To determine emission ratios of measured vehicle exhaust plumes, the regression lines are filtered by NO_x / CO_2 $R^2 > 0.95$, Δ $CO_2 > 10$ ppm and NO_x / CO_2 slope > 0. The Δ CO_2 filter ensures the observed high correlation is driven by real covariance. The positive slope filter checks that all high R^2 events are driven by positive correlation, since co-emitted species in a plume must simultaneously increase or decrease with time. The regression approach was also applied using NO_2 and NO_x as variables, to investigate the proportion of NO_2 in NO_x for the filtered emission events. In this instance, the NO_2 vs. NO_x regression results were merged (by date) with the NO_x vs. CO_2 regression results and four filtering criteria were applied: NO_x / CO_2 $R^2 > 0.95$, Δ $CO_2 > 10$ ppm NO_x / CO_2 slope > 0, and NO_2 / NO_x $R^2 > 0.90$. This ensured that the results were representative of when the plume was being sampled and also that there was good correlation between NO_2 and NO_x for the filtered emission events.

3. Results and discussion

3.1. Application to point sampling and plume chase measurements

The rolling regression method was applied to the NO_x and CO_2 point sampling and plume chasing measurements obtained during the characterisation experiments. Continuous regressions were run such that the first regression considered measurements made at 1, 2 and 3 s, the second regression at 2, 3 and 4 s, and so on. Filtering steps were applied ($R^2 > 0.95$, Δ $CO_2 > 10$ ppm, slope > 0), to extract multiple regression lines representative of diluting plumes from individual vehicle exhaust emission events.

Fig. 2 shows point sampling time series data for a 3 min period of NO_x and CO_2 measurements conducted at the test track. For this test, the vehicles waited in a convoy 8 m before the point sampling setup, and accelerated past at ≈ 20 s intervals. The order of the test vehicles is shown in Fig. 2. Each vertical line shows the time that the vehicle drove past the point sampling setup. Peaks in the NO_x and CO_2 concentrations are observed as individual vehicles drive past the point sampling instrumentation and the diluting plumes are measured. The truck NO_x and CO_2 measurements are plotted on a separate scale due to the higher concentrations. The different coloured data points in Fig. 2 show the NO_x and CO_2 measurements that meet the filtering criteria ($R^2 > 0.95$, Δ $CO_2 > 10$ ppm, slope > 0). The slope values of the regression lines generated from these data points are used to calculate emission ratios for individual vehicles. These results demonstrate the ability of the measurement technique to detect pollutants in diluting plumes from vehicle exhausts, and the ability of the data processing approach to quantify the emission ratios associated with the measured plumes. In this example, the NO_x and CO_2 concentrations measured when the scooter drives past do not meet the filtering criteria, due to the small plume generated by the vehicle under these driving conditions.

A drawback of point sampling is the challenge of distinguishing separate exhaust plumes for individual vehicles driving past in quick succession (typically several seconds). Plume mixing can occur and the measured emission ratio may be representative of emissions from more than one exhaust plume. The short-term rolling regression approach maximises the chances of isolating individual plumes, due to the high R^2 filter and the ability for the 3 s regressions to rapidly detect changes in emission ratios (slope values). Furthermore, using this approach removes the need to handle the complexities associated with background subtraction in busy traffic conditions.

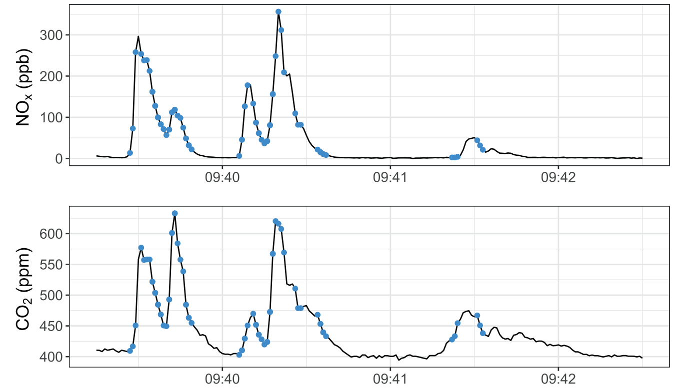

Fig. 3. NO_x and CO_2 plume chasing data collected at the test track at a time resolution of 1 s, whilst the plume chasing vehicle was measuring the diluting exhaust plume of Van 2 (VW Caddy). The blue data points show the NO_x and CO_2 measurements which meet the regression filtering criteria.

Fig. 3 shows example plume chasing time series data for NO_x and CO_2 concentration measurements made during a ≈ 3 min plume chase of Van 2. The same regression approach was applied, and the blue data points show the NO_x and CO_2 measurements that meet the regression filtering criteria. The blue data points clearly correspond to when NO_x and CO_2 concentrations are elevated above background levels, which shows that this method is highly effective at identifying when the plume chasing vehicle is sampling the exhaust plume of the vehicle of interest.

3.2. Method validation using on-board emission measurements

To validate the data processing method, on-board emission measurements conducted at the test track using SEMS were compared to the corresponding plume chasing measurements. On-board emission measurements provide a measure of pollutant concentrations at the point of emission, i.e., the vehicle tailpipe. Theoretically the NO_x / CO_2 ratio should remain approximately constant from the point of emission (measured by SEMS) to the diluted plume (measured by plume chasing in this case), since both species are affected similarly by dilution.

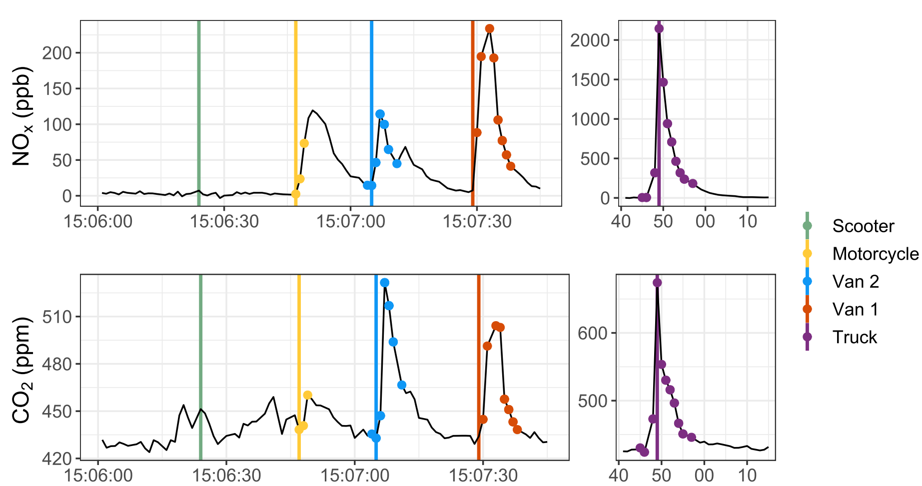

Fig. 2. NO_x and CO_2 point sampling data collected at the test track at a time resolution of 1 s. The vertical lines show vehicle pass timings and the highlighted data points show the NO_x and CO_2 measurements which meet the regression filtering criteria. The colour of the vertical lines and data points indicate which vehicle has travelled past the point sampling setup. The truck measurements are plotted on a separate scale due to the higher NO_x and CO_2 concentrations.

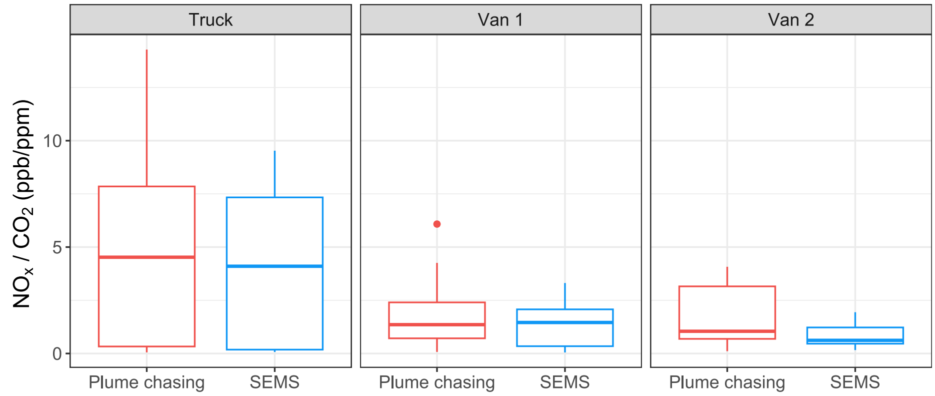

Fig. 4. Boxplots to show the distribution of the average NO_x / CO_2 emission ratios derived from each vehicle chase, calculated using the SEMS measurements and the plume chasing measurements. The bold horizontal line represents the median and the lower and upper hinges correspond to the 25th and 75th percentiles respectively. The whiskers represent 1.5 multiplied by the inter-quartile range and outlying data points are plotted individually.

The SEMS equipment was installed in the truck, Van 1 and Van 2. NO_x concentrations were measured at 1 s time resolution as each vehicle was driven around the test track and CO_2 concentrations were determined using the on-board diagnostics data. Only SEMS data collected when the vehicles were being measured by the plume chasing vehicle was considered, to ensure emission ratios were compared for simultaneous measurements of the same vehicles operating under the same driving conditions. Mean NO_x / CO_2 ratios were calculated using the SEMS data and the ICAD plume chasing data for each vehicle chase conducted during the characterisation experiments. The mean emission ratios for the plume chasing data were derived using the rolling regression approach and are based on measurements that meet the three filtering criteria outlined in Section 2.3.2. A comparison of the distribution of the mean NO_x / CO_2 emission ratios for each vehicle, based on the SEMS measurements vs. the plume chasing measurements, is shown in Fig. 4.

Overall the median values and inter-quartile range determined from each measurement technique are in good agreement for all three vehicles, with the exception of the 75th percentile for Van 2. The same trend in NO_x / CO_2 ratios is observed for the two techniques, i.e., truck >> Van 1 > Van 2. The boxplot whiskers represent 1.5 multiplied by the inter-quartile range and generally show a wider distribution for the plume chasing data. However this is not an exact like-for-like comparison and the observed differences may be attributed to the different instruments used for SEMS and plume chasing, in addition to the fact that SEMS does not measure CO_2 concentrations directly. Nevertheless, the results provide good evidence that the rolling regression data processing method is suitable for the identification and quantification of local diluting plumes from combustion sources such as vehicle exhaust emissions.

3.3. Effect of aftertreatment system tampering on NO_x / CO_2 ratios

SCR systems are used to minimise NO_x emissions from vehicles by using ammonia (NH_3) to convert NO_x to nitrogen gas and water. NH_3 is generated from the decomposition of urea present in diesel exhaust fluid. SCR emulators are illegal manipulation tools that are used to turn off SCR systems; they are used for several reasons, such as saving on the costs associated with refilling the vehicle with diesel exhaust fluid or replacing the SCR system after a certain mileage. During the characterisation experiments, the SCR systems in the truck, Van 1 and Van 2 were switched off for some of the test sessions, to replicate the effects of using a SCR emulator. Here we apply the rolling regression method to the point sampling data collected during the characterisation experiments to detect and quantify changes in emissions associated with aftertreatment system tampering.

Fig. 5 shows the filtered regression lines from the NO_x vs. CO_2 regression results for the truck, Van 1 and Van 2 point sampling measurements. The regression lines are coloured according to the state of the SCR system. The analysis is based on vehicle emission measurements under a variety of driving conditions, including a range of constant speeds between 30 and 80 km h^{-1}, and at different levels of vehicle acceleration. The test sessions under consideration covered the same driving conditions and vehicle order for SCR system on vs. off, to ensure a like-for-like comparison of emission ratios. The patterns in the regression lines reveal important changes in emissions behaviour associated with SCR tampering; the increased NO_x / CO_2 ratios when the SCR system is tampered compared to operating normally is shown for all vehicle types by the steeper slopes of the red regression lines compared to the blue.

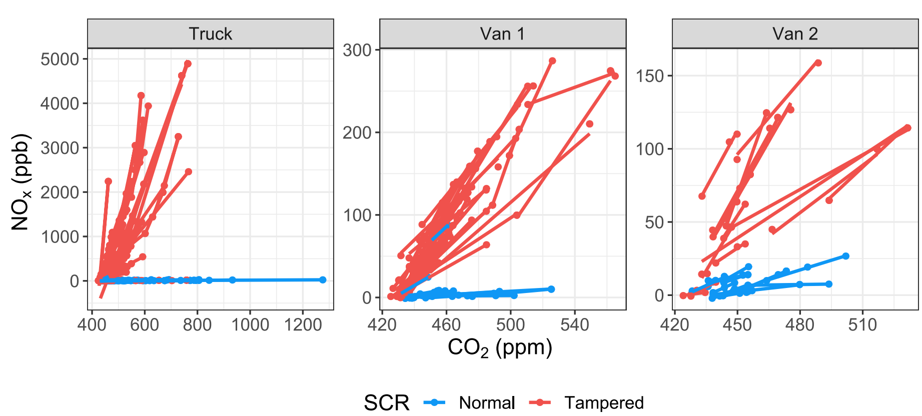

Fig. 5. NO_x vs. CO_2 scatter plots for the truck (Ford F-MAX), Van 1 (VW Transporter), and Van 2 (VW Caddy) point sampling measurements performed at Test Centre Lelystad. The filtered 3 s regression lines are coloured by whether the SCR system is switched on (operating normally) or switched off (tampered). Vehicles travelling at different constant speeds and levels of acceleration are considered.

Table 2
Mean NO_x / CO_2 ratios (ppb/ppm) and 95 % confidence intervals for the truck (Ford F-MAX), Van 1 (VW Transporter), and Van 2 (VW Caddy) point sampling measurements, grouped by the state of the SCR system.

Vehicle	NO_x / CO_2 (SCR on)	NO_x / CO_2 (SCR off)
Truck	0.29 ± 0.16	10.67 ± 1.22
Van 1	0.40 ± 0.15	2.59 ± 0.13
Van 2	0.45 ± 0.07	1.88 ± 0.26

The mean NO_x / CO_2 ratios for SCR system on vs. off are provided for each vehicle in Table 2. The average emission ratios were <0.5 ppb/ppm for all three vehicles when the SCR system was operating normally. For each vehicle, the observed increase in NO_x / CO_2 ratios when the SCR system was tampered was statistically significant, as denoted in Table 2 by the lack of overlap in the 95 % confidence intervals for the two SCR states. The largest increase in NO_x / CO_2 emission ratios due to SCR tampering was observed for the truck, and an average ratio of 10.7 ppb/ppm was observed when the SCR system was switched off. Overall this analysis demonstrates the ability of the regression approach to provide valuable insight into changes in emissions behaviour associated with aftertreatment system tampering. Whilst remote emission sensing measurements on their own may indicate a problem and potential evidence of aftertreatment system tampering, this can only be known for certain in practice through a vehicle inspection.

3.4. Detection of cold-start emissions

Vehicle exhaust emissions that occur in the first few minutes after ignition, before the engine and aftertreatment system have reached normal operating temperatures, are referred to as cold start emissions. Here we apply the regression approach to plume chasing truck measurements to reveal potential changes in emission behaviour associated with cold start emissions. Fig. 6 shows the filtered regression lines from the NO_x vs. CO_2 regression results obtained during truck plume chasing measurements. The data was collected during four subsequent vehicle chases, each conducted under the same driving conditions. The data obtained during the first plume chase, which took place at the start of the day, is shown by the blue regression lines. The red regression lines represent the remaining three plume chases. The gradients of the blue regression lines are considerably steeper, which equates to higher NO_x / CO_2 emission ratios. This is likely attributed to the fact that the truck aftertreatment systems had not reached optimum operating temperatures for the first few minutes of this vehicle chase; a cold SCR system has a limited ability to minimise NO_x emissions and therefore elevated emissions measured in the dispersing plume are expected. The mean NO_x / CO_2 ratio associated with the first plume chase was 4.52 ppb/ppm and the mean NO_x / CO_2 ratio associated with the next three chases was 0.65 ppb/ppm.

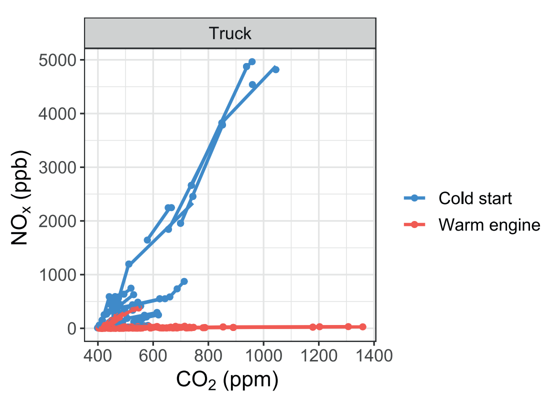

Fig. 6. NO_x vs. CO_2 scatter plot for the truck plume chasing measurements performed at the Test Centre Lelystad. The blue 3 s regression lines represent measurements conducted at the start of the day when the engine and aftertreatment systems were likely to have been below normal operating temperatures. The red 3 s regression lines represent subsequent truck plume chasing measurements when the engine has warmed up. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.5. Quantifying the fraction of NO_2 in NO_x

Increasingly stringent Euro standards have led to considerable reductions in permitted NO_x levels in vehicle exhaust emissions, yet an important consideration is the proportion of NO_x that is emitted as NO_2. NO_2 is considered the more harmful component of NO_x and is associated with adverse human health effects. The adoption of increasingly complex diesel emission control technologies has heightened concern over the fraction of total NO_x emitted as NO_2. Diesel oxidation catalysts (DOC) are used to deliberately oxidise exhaust gas components such as CO and hydrocarbons to less harmful products, but also oxidise NO to NO_2. Optimising the ratio of NO to NO_2 using an oxidation catalyst supports the performance of SCR systems and promotes passive regeneration of diesel particulate filters, both of which are positioned downstream of the DOC (Schaefer et al., 2009; Lowell and Kamakaté, 2012; Dimaratos et al., 2022).

The regression approach presented in this study is not restricted to NO_x / CO_2 ratios and any pollutants can be used as regression variables, provided that high time resolution measurements are available. Here we apply the regression approach to NO_2 and NO_x concentrations measured using the ICAD analyser installed in the plume chasing vehicle. This allows for quantification of the fraction of NO_2 in NO_x and an investigation of changes in NO_2 / NO_x ratios associated with SCR system tampering.

The upper panel of Fig. 7 shows the NO_2 vs. NO_x scatter plots for the truck, Van 1 and Van 2 plume chasing measurements. The filtered 3 s regression lines are shown and coloured by the state of the SCR system. The lower panel of Fig. 7 shows the mean NO_2 / NO_x ratios for each vehicle type, grouped by SCR on/off. The average NO_2 / NO_x ratio ranges from 0.25 to 0.26 (i.e. ≈ 25 % NO_2 in NO_x) across the three vehicles when the SCR system is operating normally. The measured NO_2 / NO_x ratios for Van 1 and Van 2 compare well with the suggested value of 0.2–0.3 for Euro 6 passenger cars and light commercial vehicles in the EMEP/EEA Emission Inventory Guidebook (Ntziachristos et al., 2019). However the suggested value for Euro VI heavy duty vehicles is 0.1, which is lower than the NO_2 / NO_x ratio measured for this particular truck during the characterisation experiments.

When the SCR system is switched off, the proportion of NO_2 in NO_x increases for each vehicle, as shown by the red data points in Fig. 7. For Van 1 and Van 2, the NO_2 / NO_x ratio increases to around 0.29, however the increase for the truck is more significant and a NO_2 / NO_x ratio of 0.44 is observed when the SCR is tampered. This is likely due to fact that the DOC is still in operation upstream of the tampered SCR, oxidising NO to NO_2 to support the performance of the SCR system.

The combination of increased total NO_x emissions and an increased fraction of NO_2 in NO_x associated with SCR tampering of the investigated truck is concerning. Plume chasing studies conducted in Germany, Switzerland and Austria found elevated NO_x emissions due to manipulated or defective SCR systems in up to 35 % of Euro V trucks and up to 25 % of Euro VI trucks, originating mainly from East and South Europe (Pöhler et al., 2019). Measurements of NO_x emissions from trucks on a Czech motorway predicted that there was no SCR functionality on 10–15 % of Euro VI trucks (Vojtisek-Lom et al., 2020). Further studies are required to determine the proportion of NO_2 in NO_x emitted from a wider range of trucks with tampered SCR systems, but the measurements in this study show that SCR tampering has the potential to not only increase total NO_x emissions, but also the proportion emitted as NO_2. This will have a direct impact on roadside NO_2 concentrations and human exposure levels to this more harmful component of NO_x.

3.6. Application of the regression approach to urban measurements

In this work we have so far demonstrated the applicability of the short-term, continuous regression approach to detect vehicle exhaust emissions

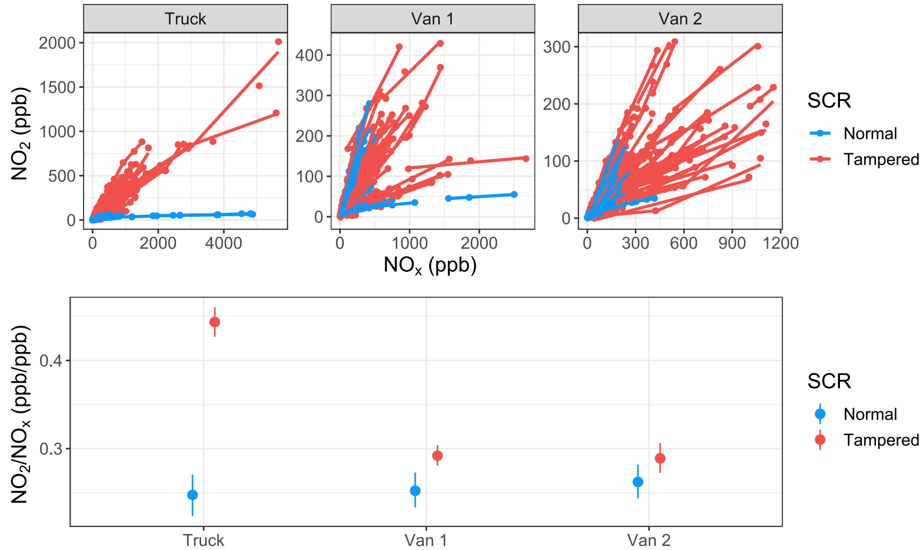

Fig. 7. NO_2 vs. NO_x scatter plots for the truck (Ford F-MAX), Van 1 (VW Transporter), and Van 2 (VW Caddy) plume chasing measurements carried out at the test track (upper panel). The filtered 3 s regression lines are shown and coloured by the state of the SCR system. Mean NO_2 / NO_x ratios for each vehicle type, grouped by SCR on/off (lower panel). The error bars represent the 95 % confidence intervals.

measured using plume chasing and point sampling techniques during controlled characterisation experiments. While the ability of the method to quantify changes in emissions associated with aftertreatment system tampering and cold start conditions has been demonstrated, ambient background concentrations at the test track were stable and the test vehicle exhausts were the main source of emissions. In principle, the presented method can be applied to high time resolution data in any setting, including urban environments where the source profile of emissions is complex and the background levels are not stable. This would provide the most valuable insight in terms of characterising diluting plumes from local combustion sources in real-world settings.

Fig. 8 shows the results obtained when the rolling regression approach is applied to the high spatiotemporal resolution measurements of NO_x and CO_2 collected during mobile monitoring in the city of Milan, Italy. The colour scale shows the variability in the NO_x / CO_2 ratios, calculated from the average ratios per 30 m road segment for each repeat circuit. The short term nature of individual regression lines (3 s) compared to background variations allows for fresh emissions from local combustion events such as vehicle exhaust emissions to be identified.

While the filtered data consist of only 13 % of the total data, importantly this data represents local plume dilution events. A low proportion of filtered data is expected in an urban environment because of the complexity of

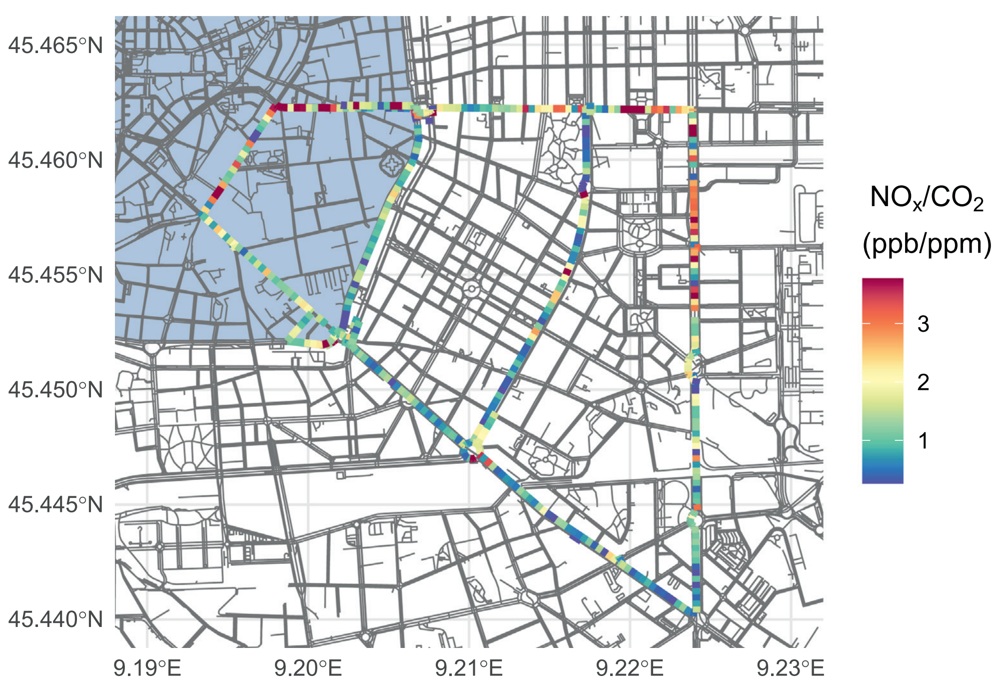

Fig. 8. Map to show the mobile monitoring route in the city of Milan, Italy. The route is divided into 30 m segments and coloured by the mean NO_x / CO_2 ratios (ppb/ppm) per segment. The colour scale spans from the 5th to the 95th percentile NO_x / CO_2 values. The NO_x / CO_2 values are derived from filtered 3 s NO_x vs. CO_2 regression lines. The blue shaded region shows Area C, the combined Urban Toll Road and Low Emission Zone. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

sources. However, we consider this aspect of the approach to be a considerable advantage: how to easily extract local dilution events from a time series of data affected by a multitude of sources, even though a lower proportion of the full dataset is used.

The average NO_x / CO_2 ratio calculated from the mean of every 30 m segment for every repeat circuit was 1.61 ppb/ppm, and 5 % of segments had an average NO_x / CO_2 ratio >4.77 ppb/ppm. The maximum average ratio for any single segment was 13.92 ppb/ppm. A key advantage of applying this method to spatial data is that potential emission hotspots are revealed, such as those shown on the map by the red data points. This highlights specific areas that would be useful to investigate further and obtain additional information to assess potential factors affecting observed emissions, such as traffic congestion levels, vehicle fleet composition, and other potential sources of emissions. In turn, this helps to inform policies and design more effective emission reduction strategies.

4. Conclusions

The short-term continuous regression approach presented in this study is designed to provide a single, reproducible method that can be applied to a range of high time resolution data sets for the quantification of emission ratios in diluting plumes from local combustion events. The development of this method has arisen from recent significant progressions in remote emission sensing technologies and the requirement for a complementary data processing method. The method can be used to quantify emission ratios for individual vehicles under real-world driving conditions. Background subtraction is not required, which significantly reduces the complexity of the data analysis, particularly in complex urban environments where background concentrations can vary over short spatial scales.

High time resolution plume chasing and point sampling data collected during vehicle emission characterisation experiments carried out under controlled conditions was used to show potential applications of the rolling regression approach. The method was validated by comparing derived plume chasing NO_x / CO_2 ratios to corresponding NO_x / CO_2 ratios from on-board emission measurements, for a range of vehicle types. The comparison was based on the principle that the NO_x / CO_2 ratios should remain approximately constant from the point of emission to the diluted plume, since both species are affected similarly by dilution.

Using point sampling data from the characterisation experiments, this approach was used to detect changes in NO_x / CO_2 ratios associated with aftertreatment system tampering. NO_x / CO_2 ratios were found to increase when the SCR systems of a range of diesel vehicles were switched off. The method was also applied to the plume chasing data and used to compare cold-start emissions from a heavy duty truck to emissions occurring when the engine had warmed up.

The approach is not limited to NO_x and CO_2 as regression variables. In this work we apply the method using NO_2 and NO_x plume chasing measurements and quantify the proportion of NO_x that is emitted as NO_2 for different vehicles. The NO_2 / NO_x ratio for the two light duty diesel vehicles is found to agree with the suggested values in the EMEP/EEA Emission Inventory Guidebook. However, the measured NO_2 / NO_x ratio for the truck considered in this study is approximately 15 % higher than suggested values in the guidebook. Furthermore, we see a significant increase in the NO_2 / NO_x ratio when the investigated truck SCR system is tampered. Further research to study whether this effect is observed for a wider range of trucks is important, given the prevalence of using SCR emulators in heavy duty vehicles.

Importantly, the method is not limited to measurements carried out under controlled conditions and is suitable for determining emission ratios in diluting plumes from local combustion events in environments with high source complexity, such as urban centres. We demonstrate this by using high spatio-temporal mobile measurements of NO_x and CO_2 conducted in the city of Milan, Italy. Only 13 % of the data meets the filtering criteria but this is considered an advantage of this approach - the method can extract local dilution events from a time series of data with a complex emission source profile. Local combustion events above the local/regional background are captured and the spatial variability in the associated NO_x / CO_2 ratios is shown.

In summary, this approach is considered highly robust and reproducible for quantifying emission ratios associated with diluting plumes from local combustion events. In theory, the method can be applied to any combination of pollutants, provided high time resolution measurements are available. The scope of this method is shown through the application of plume chasing, point sampling and mobile measurements used to measure on-road vehicle emissions, but it is anticipated that the method would be useful for a wide range of emission sources. This includes other mobile fuel combustion sources, such as construction and agricultural vehicles, ships, trains and aircraft, in addition to emissions from stationary combustion activities, i.e., industrial, commercial and residential combustion.

CRediT authorship contribution statement

Naomi J. Farren: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization, Project administration. **Christina Schmidt:** Validation, Investigation, Resources, Writing – review & editing. **Hannes Juchem:** Validation, Investigation, Resources, Writing – review & editing. **Denis Pöhler:** Validation, Investigation, Resources, Writing – review & editing, Project administration. **Shona E. Wilde:** Conceptualization, Methodology, Software, Formal analysis, Writing – review & editing, Visualization. **Rebecca L. Wagner:** Conceptualization, Methodology, Investigation, Resources, Writing – review & editing, Visualization. **Samuel Wilson:** Writing – review & editing, Visualization. **Marvin D. Shaw:** Investigation, Resources, Writing – review & editing. **David C. Carslaw:** Conceptualization, Methodology, Formal analysis, Writing – review & editing, Visualization, Supervision, Project administration, Funding acquisition.

Data availability

Data will be made available on request.

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.

Acknowledgement

This work has received funding from the European Union's Horizon 2020 research and innovation programme, as part of the CARES project under Grant Agreement No. 814966 (https://cares-project.eu/). Rebecca Wagner was supported by the NERC Panorama Doctoral Training Partnership (grant no. NE/S007458/1). We thank TNO and RDW (the Netherlands Vehicle Authority) and their involved staff for making the test track characterisation experiments happen. The authors would like to thank Quinn Vroom for the coordination of the vehicle emission measurements, and Jan Pieter Lollinga and Marcel Moerman for their pivotal role in building and running the plume chase vehicle. The point sampling expertise of Markus Knoll and Åsa Hallquist is gratefully acknowledged. We are grateful to the H2020 DIAS project (https://dias-project.com/) for helping to find a suitable truck for the test track experiments. Finally we thank Innovhub and AMAT and their involved staff for enabling the mobile measurements to take place in the city of Milan, Italy.

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