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TECHNICAL PAPER

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Interinstrument comparison of remote-sensing devices and a new method for calculating on-road nitrogen oxides emissions and validation of vehicle-specific power

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ABSTRACT
Emissions of nitrogen oxides (NOx) by vehicles in real driving environments are only partially understood. This has been brought to the attention of the world with recent revelations of the cheating of the type of approval tests exposed in the dieselgate scandal. Remote-sensing devices offer investigators an opportunity to directly measure in situ real driving emissions of tens of thousands of vehicles. Remote-sensing NO_2 measurements are not as widely available as would be desirable. The aim of this study is to improve the ability of investigators to estimate the NO_2 emissions and to improve the confidence of the total NOx results calculated from standard remote-sensing device (RSD) measurements. The accuracy of the RSD speed and acceleration module was also validated using state-of-the-art onboard global positioning system (GPS) tracking. Two RSDs used in roadside vehicle emissions surveys were tested side by side under off-carriageway conditions away from transient pollution sources to ascertain the consistency of their measurements. The speed correlation was consistent across the range of measurements at 95% confidence and the acceleration correlation was consistent at 95% confidence intervals for all but the most extreme acceleration cases. VSP was consistent at 95% confidence across all measurements except for those at VSP ≥ 15 kW t^{-1}, which show a small underestimate. The controlled distribution gas nitric oxide measurements follow a normal distribution with 2σ equal to 18.9% of the mean, compared to 15% observed during factory calibration indicative of additional error introduced into the system. Systematic errors of +84 ppm were observed but within the tolerance of the control gas. Interinstrument correlation was performed, with the relationship between the FEAT and the RSD4600 being linear with a gradient of 0.93 and an R^2 of 0.85, indicating good correlation. A new method to calculate NOx emissions using fractional NO_2 combined with NO measurements made by the RSD4600 was constructed, validated, and shown to be more accurate than previous methods.

Implications: Synchronized remote-sensing measurements of NO were taken using two different remote-sensing devices in an off-road study. It was found that the measurements taken by both instruments were well correlated. Fractional NO_2 measurements from a prior study, measurable on only one device, were used to create new NO_x emission factors for the device that could not be measured by the second device. These estimates were validated against direct measurement of total NO_x emission factors and shown to be an improvement on previous methodologies. Validation of vehicle-specific power was performed with good correlation observed.

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Introduction

Air pollution is increasingly being recognized as a significant health risk (COMEAP, 2015; IARC, 2013; World Health Organization [WHO], 2013; Kampa and Castanas, 2008). Vehicle emissions have been recognized as the dominant source of air pollution in urban environments (Colvile et al., 2001). Laboratory tests offer highly precise measurement of vehicle emissions under controlled conditions but are not always representative of the real world (Tzirakis et al., 2006). Laboratory-based tests are typically undertaken by professional or robotic drivers over a standardized testing procedure and drive cycle (Council of European Union, 2001) and used to type approve vehicles as legal (e.g., Council of European Union, 1999a, 1999b). They are able to produce highly precise and repeatable measurements of vehicle emissions. Factors including vehicle age, cold starts, vehicle payload, tire type and pressure, four-wheel-drive systems, enthusiastic amateur aftermarket modifications, and engine remapping are rarely tested due to the high cost of laboratory tests. Funding is often only available to test new vehicle technologies and power trains, rather than testing the in-service deterioration of vehicles

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and their emission controls at set time intervals. With the potential for significant aging and deactivating effects of catalyst systems (Carol et al., 1989; Forzatti and Lietti, 1999; Butt, 2012), long-term trends for individual vehicles or fleet subsections are not otherwise available. Alongside these rarely tested factors a number of other factors remain untestable. Off-cycle engine power demands such as those typical of real driving cannot be tested using current drive cycles. Even the recently developed World Harmonized Light Vehicle Test procedure (WLTP) drive cycle does not adequately account for all real driving styles, particularly more aggressive phases with prompt application of torque (Demuynck et al., 2012; Sileghem et al., 2014). Road gradient has been shown to be an important factor in vehicle emissions (Wyatt et al., 2014). Laboratory tests do not account for the increased load on the engine that road gradient introduces. The complexities of calculating and applying the additional resistance acting upon the vehicle that continuously changing road gradient introduces are difficult to accurately recreate. While not directly measurable by the remote-sensing device (RSD), these factors, if influential, will be naturally visible in the distribution of the measurements and accounted for in the statistical analysis.

Portable emissions measurement systems (PEMS) begin to address some of the real driving factors but they are still limited in the range of vehicles tested to date. Kousoulidou et al. (2013), for example, uses six vehicles over a 2-week period to study emissions factors, compared to a remote-sensing study that can study 20,000 or more vehicles per week. While work is being done to improve the number of vehicles studied using PEMS, with O'Driscoll et al. (2016), for example, testing 39 Euro 6 diesel vehicles for their nitrogen oxides (NOx) emissions, this number is still small compared to an RSD survey.

Unlike satellite-based remote-sensing measurements, a short-path, cross-road RSD can explain the influence of real driving environment factors. Portable RSD measurement campaigns offer the opportunity to measure the emission performance of large volumes (typically 200–500 operational vehicles per hour, depending on traffic conditions) of vehicles in situ with the measurements enhanced by the off-line sourcing of metadata such as the fuel, rated consumption, marque, Euro standard, and date of registration. Remote-sensing captures a clear snapshot of real-world driving dynamics and vehicle emission at the point of measurement, often giving results that differ from what would be expected in a laboratory (Smit and Bluett, 2011), especially in the field of high-emitting vehicles where multiple measurements are possible (Carslaw et al., 2013). As remote-sensing studies survey the emission performance of tens of thousands of vehicles, unlike PEMS or laboratory studies, their results can be used to contrast real driving environments (RDE) from different marques and models without fear of the results being distorted by a faulty vehicle, test, or bad batch of fuel.

In studies where repeated measurements have been performed on the same or similar vehicles, a variance in the distribution of measurements is often observed beyond the stated error of the instrument (e.g., Carslaw and Rhys-Tyler, 2013b). The source of this variance is either the natural variance of the instrument measurement or the natural variance of the amount of pollution emitted by the fleet in off-cycle dynamic ranges. It is important to understand the source of the observed variance when making conclusions about vehicle emissions. Measuring the variance of the instrument in a controlled environment using gas with known concentrations allows the system variance to be defined rigorously and confirms that any other variance observed in measurements of vehicles is a result of the dynamic ranges of the vehicles observed.

Remote-sensing has been used in multiple studies using different iterations and equipment configurations in the United Kingdom (Carslaw et al., 2011, 2013; Carslaw and Rhys-Tyler, 2013b). However a direct comparison between devices has only been performed in off-road studies measuring heavy-duty vehicles at weigh stations in the United States (Bishop et al., 2009). Bishop et al. (2009) reported a linear nitric oxide (NO) correlation of $R^2 > 0.78$ but with the RSD4600 instrument underreporting by a factor of 27–30%. The vehicles in Bishop et al. (2009) were never able to exceed a speed greater than 8 km hr^{-1} and as such their power demands are not comparable with real driving. A comparison study of smaller, cleaner vehicles more relevant to general road use, with power demands more representative of real driving environments, is required, and this study aims to fill this knowledge gap.

The RSD4600, a commercially available instrument, cannot measure nitrogen dioxide (NO_2) directly, and inferences need to be made about the fraction of primary NO_2 emitted by the vehicle (fNO_2), and hence total NOx present in the exhaust plume. Measuring NO is important and worthwhile; however, due to the additional negative health outcomes associated with NO_2 and the expression of the vehicle emissions standards in terms of the total NOx it is desirable to measure both NO and NO_2. In calculating the total NOx accurate knowledge about the NO_2 content is required. The NO_2 content of vehicle exhaust plumes has been subject to change over recent years (Carslaw, 2005). The Fuel Efficiency Automobile Test (FEAT) system is the only RSD system equipped with the instrumentation required to measure NO_2 directly (Burgard et al., 2006b). In previous work the NO to NOx estimation has been performed using NO_2 values sourced from the literature derived using the Netcen roadside model based on the interactions of NO, NO_2, and ozone (O_3) (Grice et al., 2009; Abbott and Stedman, 2005), a method commonly applied in such calculations; however, being able to link the fNO_2 to measurements performed using the same equipment would be more robust. Validation of the NO measurements across two instruments with different specifications and calibrations not only would show that the measurement is consistent but would allow for fNO_2 measurements previously made by the FEAT system to be used in conjunction with the NO measurements made by the RSD4600, leading to more accurate estimates of the total NOx emitted by vehicles on the road.

The aims of this paper are to understand and quantify the accuracy of the RSD4600 measurements for NO under controlled conditions and to test the consistency of measurements across different equipment. Successful correlation between equipment will allow on-road measurements of fNO_2 by the FEAT system to be combined with NO measurements by the RSD4600 to better estimate total NOx by the more widely used and commercially available RSD4600 instrument. Steps have been taken to ascertain the accuracy of the speed and acceleration measurements under controlled vehicle driving conditions. This was undertaken to increase the confidence of the vehicle-specific power (VSP) measurement often used when analyzing vehicle emissions in previous (Carslaw and Rhys-Tyler, 2013a) and in future studies.

Instrumentation and methodology

Remote-sensing instrumentation

Initially developed in 1989 as part of the U.S. clean air program (EPA, 1990) to measure carbon monoxide (CO) (Bishop et al., 1989), remote-sensing device technology has been developed further to include hydrocarbons (HC) (Popp et al., 1999) and NO, with prototype FEAT devices able to record ammonia NH3 and NO_2 (Burgard et al., 2006a, 2006b). Measurements of the abundance of these species are made by infrared (IR) and ultraviolet (UV) photometry at frequencies where the species are known to have absorption lines (Bishop and Stedman, 1996). Measurement of NO and NO_2 is especially problematic as there are other species with strong absorption lines at frequencies similar to those used to measure NO and with a high potential for interference. The most noticeable source of interference in NO measurements is water (H_2O). Water can cause interference from both the ambient concentration and from water vapor present in the exhaust plume; however, the high spectral resolution of the instrumentation allows the impact of interference to be minimized (Jimenez-Palacios, 1999).

The Denver FEAT instrument uses a nondispersive infrared (NDIR) laser system and a dispersive ultraviolet laser system. The systems consist of a dual-element light source (silicon carbide gas drier igniter and a xenon arc lamp) and a separate detector unit with four nondispersive infrared detectors that provide an infrared (IR) reference (3.9 μm) and measurements of carbon dioxide (CO_2, 4.3 μm), as well as channels for CO and HC measurements not used in this paper. The detector unit is connected by fiber-optic cable to two dispersive ultraviolet spectrometers that measure NO, sulfur dioxide (SO_2), and ammonia (NH_3) between 200 nm and 226 nm, and NO_2 between 430 nm and 447 nm (Carslaw et al., 2015). In this instance the two units are powered by two petrol generators located approximately 5 m downwind of their respective instrument on each side of the road to decrease the risk of contamination of the emissions measurements by transient sources of pollution.

The RSD4600 uses a single NDIR spectrometer operating at the same frequency windows as the FEAT system but without the facility to measure NO_2, SO_2, or NH_3. Unlike the FEAT system, a corner cube mirror (CCM) is deployed for the RSD4600 system such that the sensing beam is reflected back and the path length is doubled, hence increasing the number of interactions with beam photons and the signal-to-noise ratio, potentially improving the accuracy of the measurement. Having the source module and the detector module on the same side of the road with the same power supply and connected directly to the control unit means that the system as a whole is more stable. Contamination of the measurement by local transient sources is mitigated through the use of a dedicated van and a large portable battery power supply. The battery packs mounted in the van guarantee a consistent power supply throughout long periods of measurement and allow for easier deployment.

The RSD4600 is calibrated as required based on environmental conditions but always halfway through the day if no previous calibrations have been needed. Environmental conditions that would necessitate a recalibration include rapid changes in temperature due to variations in the intensity of the sun, obscuring of the laser beams by resuspended dust, or disturbance of the equipment alignment. Internal gas cells are used to perform the calibration. To ensure that the calibration is still valid the instrument is audited every hour using a "gas puff" method (as described in Bishop et al. [1989] along with details of the calibration process) with bottles of control gas containing a blended mix of molecules with known relative abundances similar to a petrol engine exhaust plume (ESP, 2005). The control gas is then released into the sensing beam to simulate a passing vehicle exhaust plume. The process is automatically controlled by the instrument and is initiated from the central computer.

RSD 4600 NO measurement validation

Initial validations of NO measurements along with hydrocarbon and carbon monoxide measurements were performed as part of the factory calibration of the instrument. Over time the signal strength can change, and repeated measurements to validate the initial accuracy have been undertaken to ensure that the equipment is still performing to the initial level of accuracy. Use of a control gas test, where the relative abundances of molecules remain constant, eliminates the unknown variance introduced on road tests from the vehicles. To ensure that the previous factory assessment remained valid and to assess the variance of the remote-sensing device the RSD4600 source and detector module (SDM) and corner cube mirror (CCM) were set up away from any roads in a sheltered location and calibrated so effects due to wind and background sources were minimized. Path lengths of 7 m, 8 m, and 9 m, short but representative of single lane deployment, were tested. The audit procedure described previously was executed 30 times, as rapidly as the control system would allow, over 15-min periods. After each 15-min period of testing, the CCM was moved closer to the SDM, to test the impact of path length on signal strength and to see whether significant attenuation was observed. Each movement of the CCM was followed by a full recalibration of the instrument.

Cross-instrument validation and vehicle data collection

To cross-validate the two instruments, multiple synchronized measurements of equivalent gas emissions are required. To achieve this the two different remote-sensing devices were set up adjacent to each other as shown in Figure 1, with the sensing beams approximately 1 m apart, as close as was practical. The equipment positions were marked and photographed to ensure consistency between different days of measurements. The RSD4600 and FEAT system were operated independently and used different control gasses and a different calibration process. At no time was any effort made to synchronize any measurements other than the time stamp for data entry. The equipment was controlled by separate computers and there was no communication between the devices or control systems. No vehicle emissions data was shared prior to completion of the experiments.

Selected vehicles were driven repeatedly through the RSD4600 and FEAT equipment. At the same time the high time resolution global positioning system (GPS) data were being collected from a subsample of vehicles

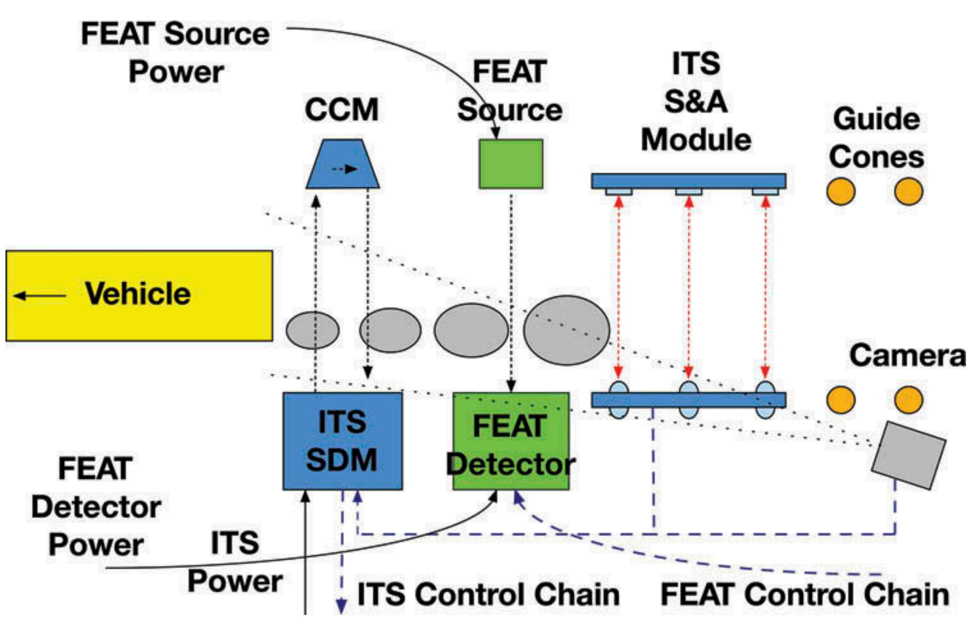

Figure 1. Schematic of the RSD4600 and FEAT remote-sensing devices deployed in tandem.

detailed in Table 1. Petrol- and diesel-powered vehicles were provided by members of the public, the University of Leeds, and Leeds City Council from their maintenance fleets and included a range of cars, vans, trucks, and buses. Different fuels, diesel, petrol, and compressed natural gas (CNG) were tested. The sulfur content of fuels was not measured but is regulated by Council of European Union (1998) and is required to be less than 10 ppm. As the vehicles used were taken from the in service fleet it is assumed that the European Union (EU) regulation value is representative of the sulfur content of the fuel. A summary of the vehicle tests are provided in Table 1.

To investigate the difference in engine power requirements, different driving styles were replicated. First, second, and third gear runs either cruising or accelerating were performed. Accelerating vehicles were required to stop at the guide cones and accelerate through the equipment, and cruising vehicles were driven through from a rolling start at a comfortable engine speed as determined by the driver. For higher gears the vehicles were accelerated from a rolling start. Limitations on space meant that gears beyond third were impossible as the vehicle was unable to pick up enough speed. Where possible the same driver was used for each vehicle. Measurements were repeated a minimum of 10 times per operating condition, and more where possible according to vehicle availability and time constraints. In some cases less than 10 valid measurements were recorded, for reasons such as obscuring by resuspended dust from the pavement surface, invalid calibration and audit measurements, and invalid gas measurements. These lost measurements are typical of any data set and are not viewed as a cause for concern.

Table 1. Description of test vehicles used in study.

Vehicle	Fuel	Euro class	Type	Synchronized RSD measurements	GPS measurements
1	Petrol	3	LCV ($N1_{III}$)	83	19
2	Petrol	4	Car	39	NA
3	Diesel	5	Car	22	NA
4	Diesel	4	Car	45	NA
5	Diesel	4	LCV ($N1_{I}$)	18	14
6	Diesel	4	Car	171	58
7	CNG	5	LCV ($N1_{II}$)	30	NA
8	Diesel	5	Car	23	23
9	Diesel	4	LCV ($N1_{I}$)	18	NA
10	Diesel	2	LCV ($N1_{III}$)	21	4
11	Diesel	4	LCV ($N1_{III}$)	26	26
12	Diesel	4	LCV ($N1_{III}$)	32	31
13	Diesel	V	HCV (rigid 2 axle)	17	12
14	Diesel	IV	Bus (single deck)	12	NA
15	Diesel	3	LCV($N1_{II}$)	109	71
16	Diesel	5	Car	25	19
17	Petrol	4	Car	7	NA
18	Diesel	IV	Bus (single deck)	19	NA
Total	—	—	—	717	277

Validation of speed and acceleration

The speed and acceleration measurements have been validated to increase the confidence in the calculated vehicle-specific power values often used in the analysis of vehicle emissions (Carslaw et al., 2013). The RSD4600 systems' easy-to-deploy, rapidly operating light gates calculate the speed and acceleration of the vehicle as it passes through. Deployed at tire level, the instrument uses multiple light gates with a fixed separation and calculates the acceleration based on the time differential. For wheel *n* breaking beam *m* at time *bn,m* and reconstituting the beam at time *rn,m* and where the beams have a constant separation *s*, the speed (v) is calculated as $b_{1,2} - b_{1,1}/_s$ and $.r_{2,2} - r_{2,1}/_s$. The time differential of these two measurements is used to calculate the acceleration (a).

The validation data of the speed and acceleration measurements were collected using a VBox global positioning system (GPS) that was able to measure the vehicle speed and heading with a sample rate of 10 Hz. The acceleration was then calculated in postprocess analysis by taking the differential of the speed measurements with a frequency of 10 Hz. VSP is calculated using the speed (miles per hour) and acceleration (miles per hour per second) of the vehicle empirically through eq 1 using a user-supplied road gradient (θ) in degrees and parameters specified in Tate (2013). The VSP calculation is performed in postprocess analysis at 10 Hz for the GPS data and individually for RSD4600 measurements with valid speed and acceleration.

$$\begin{aligned} \mathrm{VSP} = (0.2\mathrm{va}) + (4.39\mathrm{v}\sin(\theta)) + \left(95.4 \times 10^{-3}\mathrm{v}\right) \\ + \left(27.2 \times 10^{-5}\mathrm{v}^3\right) \end{aligned} \tag{1}$$

To best estimate the vehicle speed as it passed through the RSD speed and acceleration module, only GPS measurements that were taken coincidental with the vehicle passing through the RSD4600 equipment were selected for further analysis. The selection criterion was based on selecting a GPS coordinate window that matched the center of the experiment. The central point of the RSD4600 was recorded and a circular selection window with a diameter equivalent to the separation of the light gates and the reflectors was used to define the window. All GPS points that fell within this window were selected. To ensure no further unwanted measurements were included through GPS measurement error, a further check against the two instruments' time stamp was performed. A 5-sec window was used for further exclusion of GPS data that did not represent the RSD4600 data. Through this process, simultaneous measurements of the wheel speed through the light gate and the GPS were taken for a range of different vehicle speeds and accelerations, allowing the RSD4600 speed and acceleration values to be validated.

Results

NO measurement validation

To assess the accuracy of the NO value reported by the RSD4600, repeated observations of equivalent gas plumes are required. Some natural variance is expected from both the instrument and the NO source; however, the "gas puff" audit procedure described in te first section provides the most repeatable measurements with the minimum variance possible. The gas puff procedure was repeated 90 times, 30 measurements over 7 m, 8 m, and 9 m beam path length. Signal attenuation was not found to be affected by the difference in separation of the SDM and CCM. A two-sample Kolmogorov-Smirnov (KS) test was applied to each data set with the hypothesis that the two samples came from the same distribution rejected for $p < 0.05$. The p values are presented in Table 2, with all values being greater than the minimum p value confirming that they were likely to be equivalent. The data from each path length were therefore combined to form a single data set. This is in agreement with what would be expected for a coherent beam, as described in previous studies (Jimenez- Palacios, 1998). This confirms that instrument separation does not impact the results for on-road studies where the instrument is deployed with different path lengths between sites.

The central limit theorem (Rice, 1995) shows us that any variation from a normal distribution would be indicative of some underlying dominant factor influencing the measurement. It is hypothesized that the NO:CO_2 measurements taken using the audit procedure described previously follow the normal distribution. The state of the relative abundance in the control gas is known and can be modeled as a normal distribution using the mean value, which is given by the supplier, and the error, which is given by the specifications of the instrument. The distribution of the measurements was fitted to a normal distribution using the fitdistrplus package in R (Delignette-Muller and Dutang, 2015; R Core Team, 2015). The two distributions are then compared to see whether they are the same and what form the distribution of measurement takes. The fitting algorithm gives a mean and standard error of 1118.2 ± 10.3 and 97.6 ± 7.3 with an Anderson–Darling statistic of 0.51. This shows that the normal distribution is a good fit for the distribution of NO:CO_2 measurements, as would be expected for a system without any internal bias.

A positive systematic offset is observed in the measured data compared to the gas bottle value. The offset from the controlled gas bottle value may be a calibration issue for the RSD4600 or may be an error introduced by the bottle itself, as the relative abundances are subject to an error of 5%. The standard error for a normal distribution Δ is calculated for NO as $\Delta NO = \sigma NO/\sqrt{n}$ where σNO is the standard deviation and n is the number of NO measurements. In this case σNO = 58ppm and n = 90 so ΔNO = 6.1 ppm. The audit gas was therefore measured as 1118 ± 6.1 ppm by the RSD4600. The stated value is 1034 ± 52ppm. These values are consistent with each other to greater than 98% confidence. For NO the systematic error of the mean is +84 ppm and the 2σ (≈ 98%) confidence interval is ±196 ppm. The correction falls within the error of the measurement; however, the percentage error is 18.9%, higher than the 15% quoted in the manual (ESP, 2005) but within the error of the abundances of the gas bottle.

The confidence intervals and distribution shape of the RSD4600 instrumentation have been measured using repeated measurements under controlled conditions with calibration gas. The instrument was found to be performing at an acceptable level; however, the results suggest that there may be some systematic drift in the NO measurements that may be due to an error in calibration, onboard calculation of the emission factor, or an error in the gas bottle concentration. Despite the observation of a small systematic error, all the errors are within acceptable limits of the equipment and the tolerance of the control gas. The source of any further changes in the distribution observed for vehicles is the exhaust plume measured, rather than the instrumentation system.

Table 2. Matrix of KS test p values.

Path length (m)	7	8	9
7	1	0.594	0.808
8	0.594	1	0.594
9	0.808	0.594	1

Cross-instrument validation

It is important to ensure consistency between instruments if further work is to be performed using

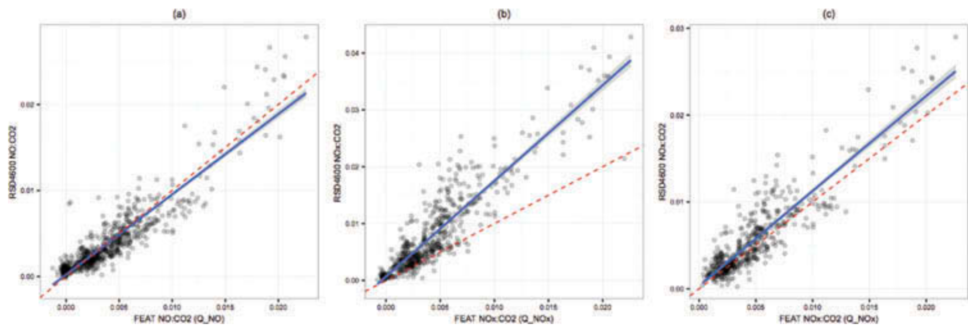

Figure 2. (a) Correlation of instruments. (b) Old method for calculating total NO_x using RSD4600 compared to measured NO_x. (c) New method for calculating total NO_x using on-road measurements of fNO_2 using the FEAT system and RSD4600 NO measurements.

comparative measurements taken using the two instruments, such as extrapolating fractional NO_2 values from the FEAT instrument to the RSD4600. The aligned measurements of NO, RSD4600 and the prototype FEAT system, for each vehicle pass through, as detailed in Table 1, were plotted against one another and a linear model was fitted for each pair to show the measured relationship along with a 95% confidence interval in Figure 2. For comparison the one-to-one equivalence line is also plotted along with the error associated with it. The results of this analysis are presented in the following.

The measurements from the two instruments should have a linear relationship if they are well correlated. A linear model with the form $y = c1x + c2$ is fitted to the data with the coefficients $c1 = 0.93$ and $c2 = 0.0$ being derived from the fit. The results are displayed in Figure 2a in blue with a 95% confidence interval shown in gray. The 1:1 relationship is identified with a red dashed line. The adjusted R^2 value is 0.85. These values are consistent with the two instruments being well correlated. Some natural variation is expected and observed. There is no evidence of the systematic decrease observed in Bishop et al. (2009), and the R^2 value suggests a better correlation than was previously reported. The RSD4600 NOx:CO_2 is calculated using eq 2. The fNO_2 values for each vehicle are estimated using two methodologies. Figure 2b uses the values derived by Grice et al. (2009) and Figure 2c uses the values measured by Carslaw and Rhys-Tyler (2013a) using the FEAT system but at an earlier date. In each case the vehicle type, fuel and euro class values displayed in Table 1 are used to estimate each vehicle's fNO_2 individually. The fNO_2 values are shown in Table 3.

$$NO_{X-RSD4600} = \frac{NO_{RSD4600}}{1 - f_{NO2}} \tag{2}$$

Both of the estimates for RSD4600 NOx:CO_2 were compared to the NOx:CO_2 measured by the FEAT system (referred to as QNOx in other literature). The calculated NOx ratio using the previously observed on-road measurements (Carslaw and Rhys- Tyler, 2013a) gave a significantly better correlation to the NOx ratio measured directly by the FEAT instrument than the previously used Grice method, which overestimates the NOx:CO_2 ratio. The coefficients derived from the linear model of the measured fNO_2 coefficient values are $c2 = 0.0$ and $c1 = 1.02$ with an adjusted R^2 value of 0.70, meaning that the use of fNO_2 values from the FEAT system with NO values from the RSD4600 system is a valid method for calculating total NOx and an improvement over the old method, especially for vehicles which have a high NOx emission ratio.

Intervehicle comparison

To understand the vehicle-to-vehicle differences when applying this model, each individual vehicle was examined to study any biases that might be present due to the different vehicle, fuel types, and emissions controls used. The results for vehicles described in Table 1 (except vehicle 17, where no comparison was possible) are presented in Figure 3 and Figure 4 along with a map of the ideal one-to-

Table 3. Fractional NO_2 values used to calculate total NOx measured using the FEAT instrument in Carslaw and Rhys-Tyler (2013a), with Grice et al. (2009) coefficients in parentheses.

Vehicle type	Euro 1	Euro 2	Euro 3	Euro 4	Euro 5
Petrol car	0.02(0.04)	0.02 (0.04)	0.03 (0.03)	0.04 (0.03)	0.08 (0.03)
Diesel car	0.14 (0.11)	0.08 (0.11)	0.16 (0.30)	0.28 (0.55)	0.20 (0.55)
Light commercial	0.11 (0.11)	0.09 (0.11)	0.11 (0.30)	0.26 (0.55)	0.25 (0.55)

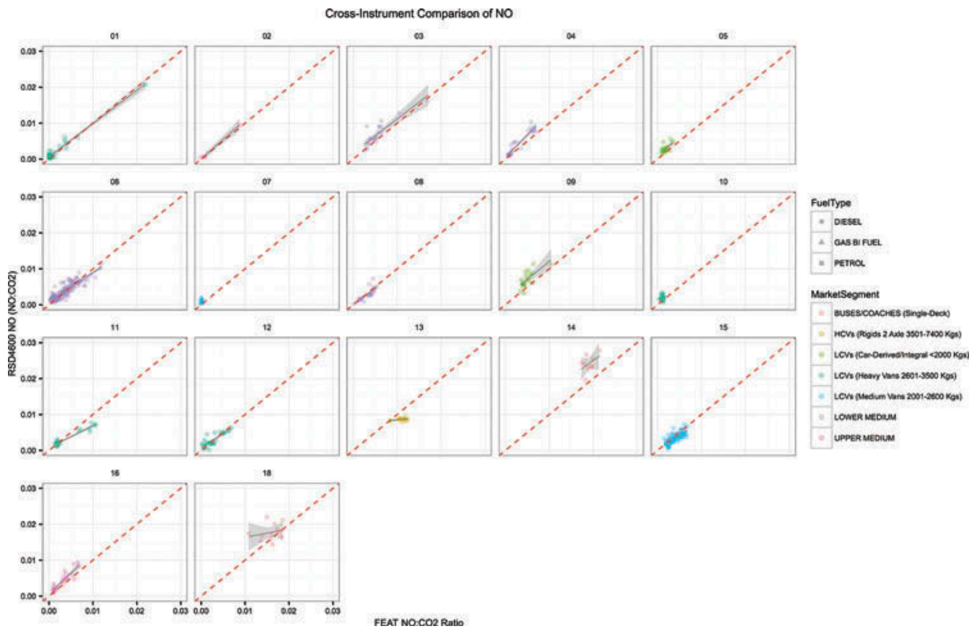

Figure 3. NO:CO_2 ratios for individual vehicles.

one relationship displayed as a dashed red line. Linear models are fitted to each vehicle's plot with a 95% confidence interval indicated by the shaded area.

The previously observed relationship between the RSD4600 and FEAT NO:CO_2 ratio emissions are confirmed and shown to be unbiased to any particular type of vehicle or fuel type, further corroborating the case that the instrument correlation is consistent across instruments with different specifications and different vehicle types. An overestimation is observed with vehicle 14; however, given the close grouping and small sample size, an accurate trend is difficult to establish from this data set and requires additional measurements to confirm. A vehicle with similar specifications (18) does not show this bias.

For the majority of vehicles the RSD4600 NO:CO_2 measurements alongside the NOx–FEAT method using measured fNO_2 values for calculating total NOx correlates strongly with the side-by-side FEAT measurements. A χ^2 goodness-of-fit test, a test that

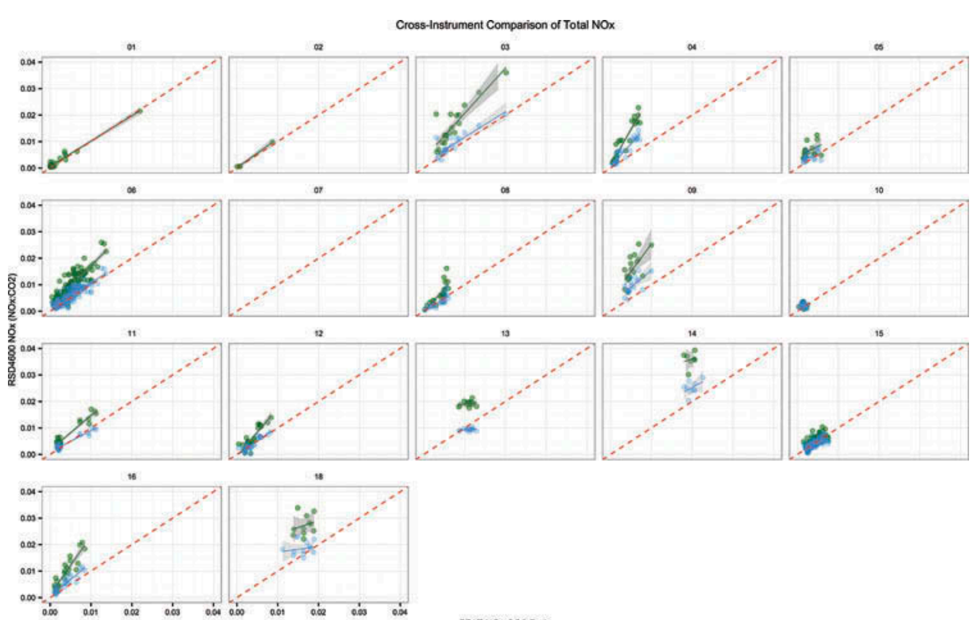

Figure 4. Individual vehicle comparison of both methods for calculating total NO_x, with the old method shown in green and the new method shown in blue.

compares the expected values of an experiment to the observed value with a lower score indicating a better match with expectation than a larger score, was applied to both the methods, giving results of χ^2model = 3.83 and χ^2FEAT = 0.371, suggesting that the FEAT method of deriving total NOx is superior to the previous methodology. By looking at the correlation plots it is clear that for most cases the FEAT method is an improvement over the NOx-model method using modeled fNO_2 values and at worst has an equivalent result. This is especially evident in higher emitting vehicles. In some cases a small systematic overestimation by the RSD4600 is observed such as is present for vehicle 4, vehicle 9, and vehicle 14; however, in comparison to the old method these still represent an improvement. Vehicle 1, a modified petrol light commercial vehicle (LCV), and vehicle 2 do not have fractional NO_2 values available and as such cannot be estimated using the FEAT method; however, in these cases the NOx-model method remains effective at predicting the total NOx and the results are included to reflect this. The cases of vehicles 13 and 4, rigid axle heavy commercial vehicles (HCVs), show that the Grice et al. (2009) methodology for estimating a vehicle's NOx emission is an underestimate by a factor of 2. The influence of commercial vehicles in total NOx emission is of special interest to many groups, and this result may prove very useful when calculating total NOx emission inventories moving forward.

Speed and acceleration validation

To validate the speed and acceleration measurements, the GPS data that met the selection criteria described in the first section of this paper were selected for analysis. In total 337,927 GPS points were recorded, with 1894 falling within the relevant selection window (0.56%). The selection window represents a very small portion of the loop driven for each pass through, and a small percentage selected for analysis is expected. From these points a further 186 GPS points (9.8% of the remaining points) were removed due to the driver not following instructions, leaving a total of 1708 GPS points for analysis. The valid GPS measurements correspond to 277 valid RSD speed and acceleration measurements across the range of vehicles.

The selected speed and acceleration data were plotted in Figure 5. The VSP was calculated using eq 1 for speed in miles per hour and acceleration in miles per hour per second and plotted alongside. A linear model of the form $y = c1x + c2$ was fitted to the speed and acceleration and a 95% confidence interval was calculated using the fitting algorithm. The coefficients for the speed and acceleration models are $c2$ = 0.61 and 0.47, $c1$ = 0.96 and 0.83, and adjusted R^2 values of 0.98 and 0.86, respectively. The linear model for the VSP has $c2$ = 0.52 and $c1$ = 0.88 and an adjusted R^2 value of 0.92.

The results show a strong positive correlation between the GPS measured speed and acceleration and the RSD4600 speed and acceleration module results and hence the derived VSP values. The vast majority of results fall within the error of the instrument. The 95% confidence interval derived from the linear model of the data is within the error for the equipment (±1 mph). Acceleration is more difficult to measure and as such the measurement error is greater than that of velocity (±0.5 mph sec^{-1}); however, the RSD4600 acceleration measurements still correlate very well with the GPS measurements. There is some evidence that the RSD4600 underestimates the acceleration at the most extreme values; however, those levels of acceleration are not typical of real-world driving and are still largely within the error of the instrumentation. The finding is

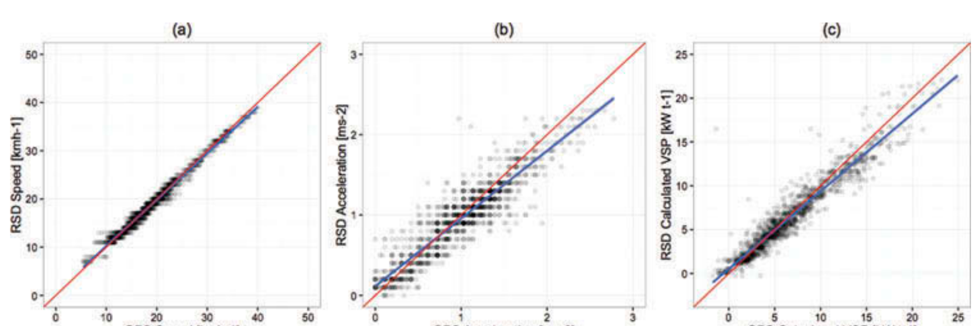

Figure 5. (a) Speed, (b) acceleration, and (c) VSP GPS to RSD4600 correlation plots. Gray to black data points show individual measurements, with darker areas having a higher density of data. A linear model is fitted to the data, shown as a blue line. The 95% confidence intervals are given for the model in gray and for the instrument in red.

that the speed and acceleration measured by the RSD4600 axre validated and therefore the calculated VSP values are considered reliable in studies such as Carslaw et al. (2013).

Conclusion

The NO:CO_2 measurement performance of the RSD4600 has been rigorously tested under controlled conditions and has also had its results validated. An unaccounted-for bias of 84 ppm has been found in the calibration or results of the controlled gas experiment; however, all results fall within the tolerance of the equipment and the materials used. At the current time, because the source of the bias cannot be isolated it is not treated as part of the systematic error of the instrument but as an error relating to the materials.

The NO measurements made by the RSD4600 of vehicles passing through the beam have been compared with the FEAT system on loan from the University of Denver. Side by side comparisons of a wide range of vehicles have been made with the RSD4600 and match well with an R^2 = 0.85. The strong correlation with no statistically significant offset suggests that the systematic error observed in the repeated controlled audit gas measurements to assess the instrument error is probably due to the gas bottle. Different calibration gas was used between the RSD4600 and the FEAT system, meaning that the consistency of their measurements is independent of the calibration, implying that this is not where the source of error is. While only one RSD4600 was available for testing, the observed consistency of measurements across two instruments that use subtly different measurement techniques means that benchmarking and calibrating other commercial RSDs is now easier. Any systematic error caused by uncertainties in the control gas abundances would be systematically carried through the distribution analysis, rather than adding to the measurement error. Repeating the control gas experiments with a different gas bottle would show whether or not the error is due to uncertainties in the control gas abundances or a calibration problem with the instrument.

The relationship between the instruments' NO emission and total NOx validation shows that the two different instruments are consistent. The results have shown no bias toward or against any particular vehicle type or fuel class, with consistently well-correlated results regardless of vehicle tested. The most significant implication of this result is that fractional N O2 (fNO_2) values measured in previous studies by the FEAT system, which the RSD4600 cannot measure, can be used to estimate the total NOx emitted by vehicles in RSD4600 studies. This is important, as there is only one operational FEAT system. There is significant logistical work and cost using this instrument, whereas multiple RSD4600 units are available, one of which is owned by the University of Leeds. Previous studies can also benefit from the improved estimated NO_2 fraction and hence total NOx.

A new method for calculating total NOx emissions using remotely sensed fractional NO_2 was created and validated against an instrument that could measure it directly. Validation of the fNO_2 measurements using this method has also been shown to be superior to methods using roadside models previously used, with the χ^2 goodness of fit decreasing by a factor of 10 when the new method is applied. The method has shown noticeable improvements in the correlation with heavier duty vehicles such as LCVs and HCVs, as well as with buses, high emitting vehicles that are of particular interest to policymakers. Measurements of the emission of LCVs and HCVs are rare except from RSD studies, and there is interest in their contribution to the total fleet NOx inventory. This study and other work using the FEAT system alone provide valuable information to policymakers to help them to make better informed decisions in the future.

The experiments detailed in the preceding set out to validate the measurements performed by remote-sensing device-based studies using the RSD4600. The speed and acceleration measurements were validated using a state-of-the-art GPS system, with the RSD4600 performing strongly. As the speed and acceleration module performed well, studies using the speed and this element of the RSD4600 to calculate vehicle-specific power as a diagnostic tool are validated in other works, such as Carslaw et al. (2013). The results are limited to vehicles operating in the lowest three gears; however, for urban driving these results represent the majority of operating conditions. Further investigation of higher vehicle speeds seems unnecessary for the purpose of validating the instrumentation.

In the future the RSD4600 can be used in urban settings to more accurately describe the total NOx emissions from a wide range of vehicles not limited to passenger cars but including light commercial vehicles, heavy commercial vehicles, and buses, with a high degree of confidence. The scope for remote-sensing studies has been improved from an estimation and has been shown to be very representative of actual emissions when used in combination with reported FEAT NOx and NO_2 measurements. These results continue to support the notion that use of remote-sensing in urban driving decision making is a valid and useful part of a holistic process aimed at reducing the total

NOx concentration in real driving environments, and they also validate previous work done in the field.

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