King's Research Portal

DOI:
10.1016/j.scitotenv.2017.07.137

Document Version
Peer reviewed version

Link to publication record in King's Research Portal

Citation for published version (APA):
Ropkins, K., DeFries, T. H., Pope, F., Green, D. C., Kemper, J., Kishan, S., ... Stewart Hager, J. (2017). Evaluation of EDAR vehicle emissions remote sensing technology. *Science of the Total Environment*, *609*, 1464-1474. https://doi.org/10.1016/j.scitotenv.2017.07.137

Citing this paper
Please note that where the full-text provided on King's Research Portal is the Author Accepted Manuscript or Post-Print version this may differ from the final Published version. If citing, it is advised that you check and use the publisher's definitive version for pagination, volume/issue, and date of publication details. And where the final published version is provided on the Research Portal, if citing you are again advised to check the publisher's website for any subsequent corrections.

General rights
Copyright and moral rights for the publications made accessible in the Research Portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognize and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the Research Portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the Research Portal

Take down policy
If you believe that this document breaches copyright please contact librarypure@kcl.ac.uk providing details, and we will remove access to the work immediately and investigate your claim.

Download date: 25. Sep. 2020

Evaluation of EDAR Vehicle Emissions Remote Sensing Technology

Authors:

Karl Ropkins[1*]; Timothy H. DeFries[2]; Francis Pope[3]; David C. Green[4]; Jim Kemper[5]; Sandeep Kishan[2]; Gary W. Fuller[4]; Hu Li[6]; Jim Sidebottom[5,7]; Leigh R. Crilley[3]; Louisa Kramer[3]; William J. Bloss[3]; J. Stewart Hager[7].

[1] Institute for Transport Studies, Faculty of Environment, University of Leeds, Leeds, LS2 9JT, UK.

[2] Eastern Research Group Inc, 3508 Far West Boulevard, Suite 210, Austin, TX 78731, USA.

[3] School of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham, B15 2TT, UK

[4] Analytical & Environmental Sciences Division, King's College London, London, SE1 9NH, UK.

[5] Aurora Emissions Technical Center, Colorado Department of Public Health and Environment (CDPHE), Aurora, CO 80011, USA.

[6] School of Chemical and Process Engineering, Faculty of Engineering, University of Leeds, Leeds, LS2 9JT, UK.

[7] Hager Environmental & Atmospheric Technology (HEAT) LLC, 539 Milwaukee Way, Knoxville, TN 37932, USA.

[*] Corresponding Author Karl Ropkins, email: k.ropkins@its.leeds.ac.uk.

Highlights:

- EDAR is a new laser-based method for vehicle emissions remote sensing.
- Complementary blind EDAR evaluation trials undertaken in USA and UK.
- Simulated exhaust gas tests showed high sensitivity and low drift for EDAR.
- EDAR in good agreement with other real-world emissions measurements.

Abstract:

Despite much work in recent years, vehicle emissions remain a significant contributor in many areas where air quality standards are under threat. Policy-makers are actively exploring options for next generation vehicle emission control and local fleet management policies, and new monitoring technologies to aid these activities. Therefore, we report here on findings from two separate but complementary blind evaluation studies of one new-to-market real-world monitoring option, HEAT LLC's Emission Detection And Reporting system or EDAR, an above-road open path instrument that uses Differential Absorption LIDAR to provide a highly sensitive and selective measure of passing vehicle emissions.

The first study, by Colorado Department of Public Health and Environment and Eastern Research Group, was a simulated exhaust gas test exercise used to investigate the instrumental accuracy of the EDAR. Here, CO, NO, CH_4 and C_3H_8 measurements were found to exhibit high linearity, low bias, and low drift over a wide range of concentrations and vehicle speeds. Instrument accuracy was high (R^2 0.996 for CO, 0.998 for NO; 0.983 for CH_4; and 0.976 for C_3H_8) and detection limits were 50 to 100 ppm for CO, 10 to 30 ppm for NO, 15 to 35 ppmC for CH_4, and, depending on vehicle speed, 100 to 400 ppmC_3 for C_3H_8.

The second study, by the Universities of Birmingham and Leeds and King's College London, used the comparison of EDAR, on-board Portable Emissions Measurement System (PEMS) and car chaser (SNIFFER) system measurements collected under real-world conditions to investigate *in situ* EDAR performance. Given the analytical challenges associated with aligning these very different measurements, the observed agreements (e.g. EDAR versus PEMS R^2 0.92 for CO/CO_2; 0.97 for NO/CO_2; *ca.* 0.82 for NO_2/CO_2; and, 0.94 for PM/CO_2) were all highly encouraging and indicate that EDAR also provides a representative measure of vehicle emissions under real-world conditions.

Keywords:

Vehicle Emissions; Remote Sensing; VERSS, EDAR; PEMS; car chaser; SNIFFER.

1. Introduction:

Open path optical instruments, spectrophotometers that incorporate separate light sources and analyzers, and measure the absorption of light in ambient air between the two, have been widely used in environment applications since the 1970s. However, at that time source-to-analyzer path lengths were typically of the order of hundreds of meters or more.

The first successful demonstration of an absorption technique as a viable across-road Vehicle Emissions Remote Sensing System (VERSS) was probably by Don Stedman, Gary Bishop and Colleagues at the University of Denver and the Ford

Motor Company in the late 1980s (Bishop *et al.*, 1989; Stephens & Cadle, 1991). Their success where others before them had failed reflected their focus on the stabilisation of the instrument reference beam (Burgard *et al*., 2006a), a step that allowed them both to operate at a path length of *ca.* 10 meters and to account for air disturbance by passing vehicles (an effect that was termed ‘shimmering’ in some earlier publications on this topic, see e.g. Hoshizaki *et al.*, 1973). That first instrument was a liquid nitrogen cooled non-dispersive infrared (NDIR) that only measured CO and CO_2, but they actively worked to refine it over the next two decades, removing the need for liquid nitrogen cooling (Burgard *et al.*, 2006a), adding Hydrocarbon (HC), H_2O and NO channels to their NDIR system (Stedman *et al.*, 1994, 1995; Guenther *et al.*, 1995), integrating an ultraviolet (UV) spectrophotometer (Zhang *et al.*, 1996) and, using that and modifications thereof, providing improved NO measurement (Popp *et al.*, 1999) and additional NO_2, NH_3 and SO_2 channels (Burgard *et al.*, 2006b). The Denver group and industrial partners, Environmental Systems Products (ESP), also commercialized one variant of their system, known as Fuel Efficiency Automobile Test or FEAT, as the Remote Sensing Device (RSD) series of instruments, and provided some of the earliest comments on across-road particulate measurement (see e.g. Stedman & Bishop, 2002; ESP, 2010).

Other remote sensing systems, typically based on different spectrometric approaches, have been applied to passing vehicle emissions, see e.g. LIDAR (Moosmüller *et al.*, 2003), TILDAS (Jiménez, 1998; Jiménez, *et al.*, 1999.), and alternative light sources and/or detector system combinations (Jack *et al.*, 1995; Wang *et al.*, 2000; REVEAL, 2002), and several of these have been commercialized as for example the Smog Dog and the REVEAL. However, none of these have been widely adopted. As a result, the Stedman and Bishop FEAT and RSD series of instruments are responsible for the collection of the majority of the remote sensing data currently available (see e.g. Zhang *et al.*, 1995; Sjödin & Andréasson, 2000; McClintock, 2011; Bishop *et al.*, 2012; Chen & Borken-Kleefeld, 2014). FEAT and RSD systems have also been applied to a wide range of emissions measurement applications, quite literally planes, trains and automobiles. Furthermore, the FEAT is the prototype for the classic across-road design that most other VERSSs adopted, and what most researchers picture when they think of a VERSS.

The sampling strategy does, however, have its limitations (Frey & Eichenberger, 1997; Franco *et al.*, 2013). Emission measurement is based on the absorption of light from a single beam projected across the monitored vehicle lane. This means results are highly sensitive to exhaust position and degree of plume/light beam intersection. Exact absorption coefficient assignment is also subject to some uncertainty, see discussion of path length and plume properties in e.g. Jiménez (1998), although arguably some work has been done to address such issues (see e.g. Full, 2009).

Alternative sampling strategies have been proposed for some hard-to-measure vehicle types, but these typically employ active sampling methods, e.g. the On-road Heavy-duty Vehicle Emissions Monitoring System (OHMS) system developed by Bishop, Stedman and Colleagues for Heavy Duty Vehicles with higher cab-mounted exhausts (Bishop *et al.*, 2015).

More recently, Hager Environmental & Atmospheric Technologies (HEAT LLC) introduced the Emission Detection And Reporting (EDAR) system, an infrared laser based VERSS that incorporates several novel features that make it a particularly interesting option for vehicle emissions-based applications (Hager, 2015).

Firstly, it uses a patented variation on Differential Absorption LIDAR (DiAL), a technique pioneered by Measures and Pilon (1972) and previously applied in the NASA Activity Sensing of CO_2 Emissions Nights, Days and Seasons (ASCENDS) satellite program (Abshire *et al.*, 2010). DiAL is widely reported to have both greater sensitivity and greater resolving power by comparison to conventional absorption spectroscopy-based remote sensing systems (Ambrico *et al.,* 2000; Menzies & Tratt, 2003; Abshire *et al.*, 2010; Hager, 2015). In DiAL, laser pulses are transmitted at two wavelengths, one an analyte absorption line, and another nearby but not an absorption line for that species. If the wavelengths are sufficiently close signal scattering associated with instrumental noise, sensor drift and interference from other species such as water vapor (and particulates if analyte is gaseous) are assumed to be equal for both wavelengths, and the difference between the two is regarded a function of analyte concentration alone.

Secondly, the approach also allows EDAR to be readily tuned for novel applications, e.g. measurement of for individual hydrocarbons, a capability already demonstrated in recent EPA-instigated work where the EDAR was used to measure evaporative fuel losses from US vehicles (Hart *et al.*, 2015, Stanard *et al.*, 2014). But, similarly, the PM measurement method used in the UK study reported herein, which is based on principles described in Mazzoleni *et al.* (2010), was developed recently and the NO_2 measurement was specially commissioned for the same study at short notice.

However, perhaps most importantly, the EDAR also employs a down-facing, single-unit camera configuration (Figure 1) that potentially offers a number of practical advantages over the conventional across-road, single-beam arrangement of traditional VERSSs. Because the EDAR is an above-road unit that employs a whiskbroom scanning approach (side to side across one or more lane multiple times as a vehicle passes), it takes a down-facing image of a passing vehicle and its exhaust plume. The use of this plume image means not only that the approach is likely to be less sensitive to factors such as vehicle lane position, exhaust position and wind speed but also a potential source of novel information about vehicle emissions dispersion. (See Supporting Information for further discussion.) The ‘up high’ deployment of the system also means that, once installed, it is likely to be much less disruptive to traffic flows and pedestrians and much less susceptible to system fouling, e.g. from road-level dirt resuspension and splash-back from passing vehicles, than conventional across-road systems that deploy light sources, analyzers and (if used) mirror boxes only a few inches above the road surface.

[Figure 1 about here]

This paper therefore presents key findings from recent work to evaluate the performance of the EDAR, and is intended to contribute to the knowledge base for this new-to-market technology and for policy makers considering future options for monitoring, managing and perhaps even policing air quality problems caused by traffic.

2. Methods:

This paper presents the findings of two complementary EDAR evaluation projects. The first was implemented by the U.S. Environmental Protection Agency (EPA), and undertaken by the Colorado Department of Public Health and Environment (CDPHE), and Eastern Research Group (ERG) on Bandimere Speedway grounds near Morrison, Colorado in the US in September 2015. The second was undertaken by the Universities of Birmingham and Leeds and King’s College London at three roadside sites in the UK, Tyburn Road Birmingham, Marylebone Road Central London, and Blackheath Hill Greenwich, in February 2016 as part of work supported by the UK Department for Transport (DfT).

Both evaluations were ‘blind tests’. HEAT personnel acted solely as EDAR operators and technical advisors regarding the EDAR performance during the tests, and were not privy to evaluation method outputs prior to the reporting of their own results. Likewise, assessors were not privy to patent-pending or otherwise commercially sensitive information regarding EDAR functionality and treated associated analyses for these first-round evaluations as a ‘black box’ comparison where these were issues.

We present the combined findings of these two studies, referred to hereafter as CDPHE/ERG and UoB/UoL/KCL, respectively, together because they provide complementary insights into the performance of the EDAR.

2.1. CDPHE/ERG Simulated Exhaust Gas Study:

As part of the CDPHE/ERG study, the EDAR instrument underwent blind evaluation using simulated exhaust gas methods to assess accuracy, precision, detection limit and drift.

The EDAR system was setup to measure exhaust CO, NO, CH_4 and C_3H_8, (all as estimated ppm analyte and molar ratio analyte/CO_2) and CO_2 (estimated % CO_2) and mounted (*ca.* 5 meters) above the study site, a private roadway within the grounds of the Bandimere Speedway, on a purpose-built hydraulic boom for operation in its standard down-facing configuration. The boom was secured by guy-wires and mounted on a custom-built deployment trailer used previously in EDAR studies in, e.g., Connecticut, Arizona and Tennessee. Directly below the EDAR, 3M retro-reflective tape was attached to the road surface to reflect the laser infrared light back to the EDAR instrument, creating an analytical path length of *ca.* 10 meters. Small ramps were secured to the road prior to the retro-reflective tape to protect it from damage during testing.

A CDPHE RSD Audit Truck was used as the test vehicle for the study. The test vehicle was a conventional gasoline truck that was fitted with an extended exhaust pipe to divert actual engine exhaust gases *ca.* 3 meters away from its conventional release point, and a simulated tailpipe and gas release system that allowed the controlled release of bottled reference gas from that point to simulate an exhaust plume while the vehicle is in motion. The test vehicle was also equipped with a flow meter to regulate simulated exhaust gas flow rates.

Figure 2 includes both photographs of the EDAR in the trailer mounted configuration used in the CDPHE/ERG study and the Simulated Exhaust Gas Audit Truck, and a schematic of the Audit Truck.

[Figure 2 about here]

Four five-gas (CH_4, C_3H_8, CO, NO and CO_2) reference blends, formulated by Air Liquide and hereafter designated blends A-D, were used in the study. The concentrations and relative ratios of analytes in these, as summarized in Table 1, were selected to approximate stoichiometric gasoline combustion emissions from a range of vehicles including a number that failed conventional emissions tests such as IM240, the chassis dynamometer test the EPA recommends for in-use light duty vehicles inspection & maintenance (I&M) programs.

Two additional reference gas blends, designated Q and F, were used for instrument drift checking, quality assurance and instrument testing during setup work.

[Table 1 about here]

EDAR measurements were collected for test vehicle drive-throughs at various speeds (nominally 15, 30, 45, and 60 miles per hour or mph) with reference gas release rates of ca. 30 standard cubic feet per minute (SCFM), the release rate that CDPHE typically uses to evaluate other remote sensing instruments. Initially eight drive-through measurements were made with blend F (two at each vehicle speed) and the concentrations of this blend were made known to HEAT personnel so they could confirm proper operation of their system. Then a series of 160 test runs were undertaken (ten replicates each of all combinations of the blends A-D at the four vehicle speeds) and these were used to calculate performance statistics including precision, accuracy and detection limit. Nine further runs were also made with blend Q to investigate instrument drift. These were made across the study period, and associated measurements were collected using vehicle run-though speeds of 15 mph to maximize EDAR signal size.

(See also DeFries (2016) and DeFries *et al.* (2017) for further details of this work.)

2.2. UoB/UoL/KCL Real-world Study:

As part of the UoB/UoL/KCL study, the EDAR instrument underwent blind evaluation under real-world conditions by *in situ* cross-comparison with other real-world methods (Portable Emissions Measurement System or PEMS and vehicle chaser or SNIFFER) as part of a series of more conventional roadside EDAR deployments.

During this study the EDAR was deployed at three sites in the UK, Birmingham Tyburn Road, London Marylebone Road and Greenwich Blackheath Hill. Birmingham Tyburn Road is on the A38 dual carriageway, a main arterial route into Birmingham (latitude 52.512194, longitude -1.830861). London Marylebone Road is on the A501 6-lane carriageway, a highly-congested roadway in central London (latitude 51.522530, longitude -0.154611). Greenwich Blackheath Hill is on the A2 on a steep incline (*ca.* 7%) on Blackheath Hill, a major arterial route in South London (latitude 51.472362, longitude -0.012113).

At each site the EDAR was deployed close to a conventional stationary air quality monitoring station. The stations provided fixed point air quality data at 1-hour and 15-minute resolutions that is routinely quality assured and used for regulatory air quality assurance. This, used in combination with local traffic flow and meteorological data, provided a means of characterizing conditions on the deployment days. However, co-location limited the choice of deployment sites, and meant that these sites were not optimal locations for EDAR (or any VERSS) deployment.

Two EDAR systems were deployed at all three sites for the UK studies. The first of these was setup to measure exhaust CO, NO and NO_2 (all as estimated ppm analyte and molar ratio analyte/CO_2) and CO_2 (estimated % CO_2). The NO_2 measurement channel was specially commissioned for this study. The second unit was setup to measure exhaust particulate matter (PM; reported as nanomoles/mole PM/CO_2).

At London Marylebone Road, the EDAR was mounted on the roof of the air quality monitoring station, while at Birmingham Tyburn Road and Greenwich Blackheath Hill it was mounted on scaffolding platforms setup adjacent to the local air quality monitoring station. One further compromise required for first-time UK deployment was that the EDAR units, although 5 meters above the road as in the CDPHE/ERG study, were near to, rather than directly over, the passing vehicles being monitored.

2.2.1. Portable Emissions Measurement System (PEMS) Comparison:

A vehicle fitted with a Portable Emissions Measurement System (PEMS), a specialist exhaust gas measurement system that provided a direct measure of the emissions of that vehicle, was run at the same site during the Greenwich Blackheath Hill EDAR deployment.

The PEMS system was purpose-built for this study and installed in a Ford Transit Connect Van (EURO 4 2.0L Diesel). It consisted of two gas benches, one NDIR-based for CO_2 and CO measurement and one UV-based for NO and NO_2 measurement, an ionization-based PM analyser, a Pitot-based exhaust flow measurement system, and dedicated exhaust sampling system. A zirconium sensor was also used to measure NO_x and O_2, and a secondary system, a parSYNC PLUS (supplied by 3DATX Inc) was used to provide confirmatory measures of CO_2, NO, NO_2 and PM exhaust concentrations, although the latter were not used directly in this study. Supporting vehicle, engine and GPS data were collected using a commercial logger. Associated data was aligned and emissions calculated using dedicated R code/methods (Ropkins, 2016).

A schematic of the PEMS vehicle installation is provided as Figure 3 Left.

[Figure 3 about here]

The PEMS test vehicle was run through the EDAR measurement area multiple times under a range of engine loads and in different gears with the objective of providing a broad range of emissions.

The PEMS/EDAR data alignment strategy used here was a refinement of one previously employed to compare PEMS and RSD data in earlier work (Ropkins *et al.*, 2008) and summarised as follows: (1) The PEMS data was time and location filtered to provide ±20 seconds windows of data for the pass throughs. (2) Data within these windows was locally aligned by correlation lag-fitting using sets of six or more consecutive pass-throughs. (3) Paired PEMS and EDAR measurements were then filtered to remove cases where EDAR and PEMS data were unlikely to be comparable. PEMS logs data on a ‘per-second’ basis. EDAR interpolates vehicle emissions from plume images, resulting in measurements with a time resolution of about 10-100 milliseconds. For the shorter duration EDAR measurement to be broadly representative of the second of PEMS data it is encompassed by, the vehicle motion needs to be smooth throughout that second. So, cases where the PEMS vehicle trajectory were highly non-linear about the PEMS pass-through point ($R<0.8$ for 10Hz speed records, second before to second afterwards) were discarded prior to the analysis.

2.2.2. Car Chaser Comparison:

The University of Birmingham Mobile Air Monitoring Laboratory (MAML) was operated in car chaser or SNIFFER configuration to measure preceding vehicle emissions at the same site during the Birmingham Tyburn EDAR deployment.

The MAML test vehicle is a Ford Transit that was specially instrumented for this study with a NDIR CO_2 (LICOR LI-820), a chemiluminescence NO (TEI 42c), a chemiluminescence/Molybdenum NO_2 converter NOx (TEI 42i-TL) and a UV absorption O_3 (2B 202) analyzers, all sampling independently from a dedicated forward-facing inlet mounted on the vehicle roof.

A schematic of the SNIFFER vehicle installation is provided as Figure 3 Right.

The SNIFFER test vehicle was run through the EDAR measurement area multiple times following a range of other vehicles. In addition to chasing vehicles randomly selected from the passing fleet the SNIFFER test vehicle also ‘repeat chased’ a second test vehicle, a Vauxhall Zafira (Diesel 2.0 CDTI), to benchmark reproducibility.

The SNIFFER test vehicle was operated by a dedicated driver and journey documenter who recorded details of the chaser runs through the EDAR monitoring area, e.g. followed vehicle registration number, time followed, approximate time passing over EDAR reflect strip, etc. As SNIFFER vehicle measurements were of ambient air following the chased vehicle, background concentrations before/after identified plume events were subtracted to provide plume contributions. In cases where the analyte plume peak associated with a reactant trough indicating post-emission reaction (e.g. NO plume peaks were often seen alongside O_3 troughs indicating NO depletion), titration contributions were also accounted for by assuming e.g. NO emitted = NO observed + O_3 consumed. Finally, as ambient plumes were typically several seconds in duration, SNIFFER measurements were reported as averages with error bars to show measurement variability for the observed plume.

3. Results and Discussion:

The simulated exhaust gas study provided a highly standardizable and controllable point-of-reference for the evaluation of EDAR. In terms of assessing the instrumental accuracy, precision, limit of detection and degree of the drift, this approach is probably the most robust and confounder-free option for the assessment of EDAR instrument performance under routine operating conditions. However, it is also a relatively idealized point-of-reference by comparison to real-world vehicle exhaust emissions. Firstly, the gas blends are dry while exhaust gas is rarely moisture-free and, secondly, it is a very stable analytical reference while vehicle emissions are very dynamic.

By comparison, the PEMS and SNIFFER EDAR comparisons were more representative of on-road vehicle emissions. The reference methods provided real-world measures of the actual (wet, dusty and dynamic) emissions of in-use vehicles operating under conditions more typical of the conventional vehicle fleet. However, the associated references, PEMS and SNIFFER measurements, were less exact points of reference than the gas release set-points and the associated experiments were not as readily controllable. As a result, these point-of-references were more susceptible to measurement uncertainty.

Both EDAR and other VERSS manufacturers have made various claims about the (in)sensitivities of their systems to real-world confounders. The direct and unambiguous evaluation of such factors is arguably outside the scope of any current single test strategy. However, by reporting these complementary studies together, we aim to provide measures of both the absolute instrumental performance of the EDAR and the reliability of the real-world vehicle emissions data it generates in typical on-road applications.

3.1. Simulated Exhaust Gas Studies

At all speeds studied (15, 30, 45, 60 mph), EDAR measurements were found to be in good agreement with reported gas blend concentrations (See Figure 4).

[Figure 4 about here]

Several relatively high CO readings were observed while measuring the lowest CO reference gas levels. Although the exact source of these measurements was not identified, other on-site CO sources cannot be ruled out. CO results were therefore calculated with and without these possibly unrepresentative measurements to assess their influence. Linear regressions indicated small relative biases and intercept biases of +6% and *ca.* -29ppm, respectively, for CO in the range 30 to 30,000 ppm. Data scatter was <1% (as indicated by measurement/gas blend regression R^2 values of 0.992 or higher) and not majorly affected by the exclusion of the possibly unrepresentative measurements.

Conventional detection limits are not widely reported for VERSS systems, perhaps in part because measurements are typically expressed as molar ratios relative to CO_2 rather than absolute concentrations. For example, one approach used by Stedman, Bishop and colleagues in recent work with the FEAT uses Laplace factors and treats CO_2 as a dependent variable (see e.g. Bishop & Stedman, 2014) to provide a measure of noise associated with ratio-based outputs. However, here a more conventional measure, the EPA ‘Analysis of Pollutants’ guideline limit of detection method (US EPA, 2015) was used to estimate absolute values: 2.998 × standard deviation as determined by eight replicate analyte measurements at concentrations between one and five times the expected detection limit.

The EDAR detection limit for CO (estimated as 3 × standard deviation) was found to be *ca.* 50-100 ppm, or maybe slightly lower if the possibly unrepresentative measurements were removed.

For NO concentrations between *ca.* 40 and 500 ppm, both relative biases and intercept biases were also small, ca. -3% and -2 ppm, respectively, and data scatter was <1% (R^2 values of 0.998 or higher). The NO limit of detection, estimated as 3 × standard deviation (7 ppm), was about 10-30 ppm.

Performance statistics were also highly encouraging for both CH_4 and C_3H_8.

For CH_4 in the concentration range 0 to 210 ppmC, relative biases and intercept biases were about +4% and -19 ppmC, respectively, and although the data scatter was larger than seen for CO and NO (R^2 0.983) and, similarly, subject to no (or more strictly statistically negligible; no apparent trends, p for speed contribution << 0.05) speed dependency, the standard deviation was 5 to 12 ppmC, indicating a detection limit of about 15 to 35 ppmC.

For C_3H_8 in the concentration range 30 to 1300 ppmC_3, relative bias was +3 to -3%, intercept bias was 3 to 37 ppmC_3, R^2 was 0.993 to 0.952, and detection limit was 100 to 400 ppmC_3, although here it should be noted that a moderate speed dependency was observed for C_3H_8 during testing, and the results were subject to non-blind recalculation before final reporting which did improve the statistics.

Test vehicle runs using simulated exhaust gas Blend Q containing CO_2, CO, NO and C_3H_8 (Table 1) were made repeatedly alongside the main tests and regression analysis performed to provide a measure of instrument stability/drift. The results, summarized in Figure 5, indicated that the EDAR exhibited no significant drift for any of the emission species in Blend Q.

[Figure 5 about here]

A similar but smaller scale simulated exhaust gas audit was also undertaken on the University of Birmingham campus as part of quality assurance activities for the UoB/UoL/KCL study. This used an electric vehicle as the test vehicle, and, although not reported here, the results were highly consistent with those observed during the CDPHE/ERG study.

3.2. Portable Emissions Measurement System (PEMS) Comparisons:

The ability of PEMS to directly measure emissions across a wide range of driving activities (compare methods in Ropkins *et al.*, 2009 or Franco *et al.*, 2013) makes it the current front-runner for a real-world legislative emissions standard (Giechaskiel *et al.*, 2016) and also an obvious point of real-world comparison for this study. Previous conventional across-road remote sensing (RSD) versus PEMS and/or On-Board Diagnostic (OBD) (e.g. Lawson *et al.,* 1990; Ropkins *et al.*, 2008; Kraan *et al.*, 2012; Carslaw & Priestman, 2015) studies demonstrate both the value and limitations of this evaluation strategy. Although the PEMS/EDAR comparison is most likely the most direct and real-world representative of the comparisons reported within this study, the degree of absolute agreement is likely to be limited by both the technical challenge associated with the time alignment of the two datasets and the difference in the time resolution of the two measurement types, 10-100 milliseconds for EDAR and 1 second for PEMS.

Of the 41 paired EDAR/PEMS records collected during the Greenwich EDAR deployment, 25 were part of smooth PEMS vehicle trajectories (before-to-after speed linear fit R>0.8), indicating that these were most likely to be suitable for comparing the two techniques. The outcomes are shown in Figure 6 where EDAR and PEMS emission paired measurement comparisons are shown on the basis of CO_2 ratios, the most common format used elsewhere to report VERSS data. Note that all data exclusion is on the basis of smoothness of vehicle trajectories, not the agreement of emission measurements.

[Figure 6 about here]

The CO/CO_2 EDAR/PEMS comparison plot is dominated by two much higher CO/CO_2 measurements that most likely overinflate the degree of agreement. So, this plot, Figure 6 Top Right, includes an insert in the top left corner showing the fit with these two higher points excluded. At this level, again two CO/CO_2 measurement pairs dominate and excluding these would further reduce the fit R^2 to *ca.* 0.9. However, at this point, most measurements were at or near to the PEMS CO detection limit, and it is likely that measurement noise would be an issue. As a result, the fit for measurement CO/CO_2 ratios < 0.01 (R^2 0.924; EDAR 0.73×PEMS) was selected as a 'best compromise' estimate of *in situ* agreement.

Agreement between smooth trajectory paired EDAR and PEMS NO/CO_2 measurements was good, R^2 0.968, EDAR 0.71×PEMS, and NO/CO_2 measurements from both sources were well distributed across the observed range, *ca.* 0.001 to 0.012.

The correlations for paired EDAR and PEMS NO_2/CO_2 measurements was the lowest observed (R^2 0.797 for a linear fit but possible non-linearity, R^2 0.843 for polynomial regression), and measurement agreement was least affected by PEMS vehicle trajectory. This suggests less confidence associated with these measurements. However, here, it is important to acknowledge the analytical challenges associated with the measurement of this highly reactive species. This is a consideration for both PEMS measuring NO_2 in the exhaust, where samples are wet, dirty and concentrated, and EDAR measuring NO_2 in the in-air plume where NO_2 is subject to significant secondary chemistry.

Across the reported EDAR measurement range 5 to 80 nanomoles.mole^{-1} PM/CO_2, good agreement (R^2 0.937) was observed with paired smooth trajectory PEMS PM/CO_2 measurements (20 to 200 ng/g).

For CO/CO_2 and NO/CO_2, the observed bias in EDAR/PEMS comparisons (EDAR under-estimated emissions by comparison to PEMS) most likely reflected the different time resolutions of the two measurement types and measurement/sampling point (in-exhaust for PEMS, in-post-exhaust-plume for EDAR) rather than an issue with either measurement type. This was also similar to bias reported in previous RSD/PEMS comparisons (e.g. Ropkins *et al.*, 2008; Kraan *et al.*, 2012). The larger measurement biases for NO_2/CO_2 and PM/CO_2 (EDAR *ca*. 0.3×PEMS) probably reflect measurement confidence and NO_2 reactivity for NO_2/CO_2 and unit, calibration and PM measurement metric response differences for PM/CO_2, respectively.

3.3. Car chaser (SNIFFER) comparison:

In SNIFFER experiments, the chased vehicle exhaust plume was sampled several seconds after emission. During this time the emitted species have undergone some degree of dilution, dispersion and atmospheric chemistry. As a result, an in-exhaust event that was 10-100 milliseconds in duration may generate an in-air plume that is several seconds in duration when sampled by the SNIFFER. This plume could also overlap with other in-air plumes/events, further complicating event isolation.

This combination of measurement contributions is illustrated by Figure 7 Left, which also demonstrates the analytical procedure used to estimate at-exhaust NO/CO_2 emissions from SNIFFER data collected during this study. For at-exhaust NO/CO_2 ratio calculation from SNIFFER data, average local background measurements were taken at time of EDAR/SNIFFER measurement and subtracted from plume and all O_3 depletion was attributed to NO conversion to NO_2. The different gas phase diffusion rates of NO and CO_2 were also taken into account to correct for the SNIFFER measured ratio to that of the EDAR which is measured just post exhaust. Diffusion of NO is faster than CO_2 and hence the SNIFFER measures a lower ratio NO/CO_2 ratio in the centre of the plume than the EDAR. The following literature values for the CO_2 and NO diffusion constants were used 0.160 and 0.230 cm^2s^{-1} (Marrero & Mason, 1972; Tang et al., 2014), respectively.

[Figure 7 about here]

Arguably this is the most analytically challenging of the comparisons employed within this study, and associated uncertainties are likely to be the largest.

Figure 7 Right shows NO/CO_2 emissions of eight paired EDAR and SNIFFER measurements.

Although the dataset is admittedly small, the degree of agreement for paired EDAR and SNIFFER data is, like the PEMS comparison, highly encouraging. The linear regression R^2 is 0.862, and although there is a fixed offset, indicated by the intercept and perhaps associated with analytical uncertainties, the relative agreement is near unity (gradient *ca.* 1).

The plot includes both same vehicle repeat measurements (CAR01, the second test vehicle which the SNIFFER vehicle repeat chased by the EDAR to benchmark reproducibility) and several other vehicles (one car, one small goods vehicle or LGV and two heavy goods vehicles or HGVs). The agreements seen across this sample strongly suggests that the one-vehicle agreement observed in the PEMS/EDAR real-world comparison could reasonably be expected for other vehicles in the larger fleet.

4. Conclusions and Outlook:

The CDPHE/ERG simulated exhaust gas test exercise used conventional VERSS auditing methods to investigate the instrumental accuracy of the EDAR. This study found that EDAR measured NO, CO, and CH_4 concentrations at levels representative of in-use vehicle emissions with high linearity, low bias, low speed dependence, and low drift over a wide range of concentrations and vehicle speeds. Similar findings were also observed for C_3H_8 once vehicle speed had been taken into account. It is, however, important to note here that EDAR provided discreet and independent measures of CH_4 and a non-methane hydrocarbon, and this alone is currently a novel output for a VERSS. Furthermore, the observed lack of drift makes it a viable candidate for unattended operation. The observed detection limits for CO, NO and CH_4 were 50 to 100ppm, 10 to 30 ppm and 15 to 35 ppmC, respectively. The potential to differentiate hydrocarbons, demonstrated here by discrete CH_4 and C_3H_8 measurement could also significantly extend diagnostic capabilities of VERSS. As advances in vehicle emissions control system performance and continued fleet turnover drive down vehicle emissions and we seek to more effectively manage emissions across our vehicle fleets, such sensitivity and selectivity are likely to become increasingly important considerations for the emissions measurement community.

That said, a simulated exhaust gas study is a highly standardized case, and the point of reference is a dry gas released at a fixed rate. To address this issue, we also present findings from the UoB/UoL/KCL study that used the comparison of EDAR, PEMS and car chaser/SNIFFER measurements collected under real-world conditions to provide a measure of *in situ* EDAR performance. Given the analytical challenges associated with aligning these very different measurements and acknowledging the limitations of sample size, the observed degrees of agreement (e.g. EDAR/PEMS R^2 0.92 for CO/CO_2; 0.97 for NO/CO_2; *ca.* 0.82 for NO_2/CO_2, 0.80 linear or 0.84 non-linear; and, 0.94 for PM/CO_2, and EDAR/SNIFFER R^2 0.862 for NO/CO_2) were all highly encouraging and suggest that EDAR provides a representative measure of vehicle emissions under real-world conditions. While we cannot rigorously attribute specific proportions of the measurement errors to EDAR, PEMS, SNIFFER or the alignment method used to compare them, uncertainties typically associated with the latter are comparable to those observed here. So, although we cannot say unequivocally that EDAR performs as well in the real-world as it does relative to a simulated exhaust gas, we have no evidence that it does not, and NO_2 and PM measurement capabilities, not as easily assessed using simulated exhaust gas study methods, also provide highly encouraging results.

Recent events such as diesel-gate, the exposure of use of test-detection software to circumvent regulatory procedures by some vehicle manufacturers, and growing concerns more generally about attempts to game regulations have highlighted the discrepancy between vehicle test and on-road performance. This has also been identified as a major element in the under-performance of recent air pollution management strategies (see e.g. discussion in Anenberg *et al.*, 2017). Significant questions would need to addressed, both technical (e.g. regarding vehicle measurements under more extreme engine loads, weather conditions, etc.) and legislative/ethical (can and should we act on individual measurements), before any VERSS can be used in anything approaching a regulatory fashion. But, that accepted, if we want to actually target the worst polluters as part of e.g. the next generation of Low Emission or Clean Air Zone schemes, this is a challenge we urgently need to address, and EDAR is arguably one of the tools we should be considering as part of that process.

Our on-going challenges in work to benchmark EDAR are to extend the body of evidence on real-world performance, e.g. using different vehicles, fuels and reference methods, so we can better characterise measurement confounders and to identify unique applications of the technology. But we also need to look at the questions that are applicable to VERSS as an instrument class rather than the EDAR in isolation, e.g. how we validation emission measurements across broader ranges of driving activities and conditions and how the accuracy of these post-exhaust measurements is affected by different emission abatement strategies.

Acknowledgements:

The authors gratefully acknowledge the following support: Evaluation of EDAR by Measurement of Simulated Exhaust Emissions, partially funded as part of the US EPA PEMS contract with Eastern Research Group Inc, and in-kind support from the Colorado Department of Public Health and Environment and Eastern Research Group Inc. The Birmingham and London EDAR (Emissions Detection And Reporting) Demonstration and Evaluation project awards, funded as part of the UK Department of Transport Local Transport Air Quality Challenge Innovation Grant October 2015 funding scheme. Although this work was in part sponsored by various institutions, this paper reflects the views of the authors and does not necessarily reflect the official views or policies of any sponsor: it does not constitute a standard, specification, or regulation.

THD gratefully acknowledges the contributions of Carl Fulper and US Environmental Protection Agency, Colorado Department of Public Health and Environment, and colleagues at Eastern Research Group Inc. KR and FP gratefully acknowledge the contributions of Jim Mills and the Air Monitors team as part of the Birmingham and

London EDAR deployments, and the input, help and advice of multiple collaborators at UK Department for Transport, Transport Systems Catapult, UK Department for Environment, Food and Rural Affairs, Transport for London, Mark Wolstencroft and colleagues at Birmingham City Council, Rosa Colucci, Nick Marks and colleagues at Greenwich Council, University of Westminster and Westminster City Council involved of the logistics of the same work. FP also gratefully acknowledges the contributions of Dr Hao Huang (then University of Birmingham, now Air Monitors). KR also gratefully acknowledges the comments and suggestions of Kent Johnson (University of California Riverside) when refining the PEMS/EDAR data comparison method and the support of Katrina Hemingway in the preparation of this manuscript.

Finally, the authors also gratefully acknowledge the time and valuable input of the editor and reviewers, whose anonymous contributions are greatly appreciated.

References

Abshire, J.B., Riris, H., Allan, G.R., Weaver, C.J., Mao, J., Sun, X., Hasselbrack, W.E., Kawa, S.R. and Biraud, S., 2010. Pulsed airborne lidar measurements of atmospheric CO_2 column absorption. *Tellus B*, 62(5), pp.770-783. DOI: 10.1111/j.1600-0889.2010.00502.x.

Ambrico, P.F., Amodeo, A., Di Girolamo, P. and Spinelli, N., 2000. Sensitivity analysis of differential absorption lidar measurements in the mid-infrared region. Applied optics, 39(36), pp.6847-6865. DOI: 10.1364/AO.39.006847.

Anenberg, S.C., Miller, J., Minjares, R., Du, L., Henze, D.K., Lacey, F., Malley, C.S., Emberson, L., Franco, V., Klimont, Z., and Heyes, C., 2017. Impacts and mitigation of excess diesel-related NOx emissions in 11 major vehicle markets. *Nature,* 1476-4687. DOI: 10.1038/nature22086.

Bishop, G.A., Hottor-Raguindin, R., Stedman, D.H., McClintock, P., Theobald, E., Johnson, J.D., Lee, D.W., Zietsman, J. and Misra, C., 2015. On-road heavy-duty vehicle emissions monitoring system. *Environmental science & technology*, 49(3), pp.1639-1645. DOI: 10.1021/es505534e.

Bishop, G.A., Schuchmann, B.G., Stedman, D.H. and Lawson, D.R., 2012. Multispecies remote sensing measurements of vehicle emissions on Sherman Way in Van Nuys, California. *Journal of the Air & Waste Management Association*, 62(10), pp.1127-1133. DOI: 10.1080/10962247.2012.699015.

Bishop, G.A., Starkey, J.R., Ihlenfeldt, A., Williams, W.J. and Stedman, D.H., 1989. IR long-path photometry: a remote sensing tool for automobile emissions. *Analytical Chemistry*, 61(10), pp.671A-677A. DOI: 10.1021/ac00185a746.

Bishop, G.A., Stedman, D.H., 2014. On-Road Remote Sensing of Automobile Emissions in the Denver Area: Winter 2013. Online at: http://www.feat.biochem.du.edu/assets/databases/Colorado/6th_I25/Denver_Year_7_DU13.pdf (as of 12/01/2017).

Burgard, D.A., Bishop, G.A., Stadtmuller, R.S., Dalton, T.R. and Stedman, D.H., 2006a. Spectroscopy applied to on-road mobile source emissions. *Applied Spectroscopy*, 60(5), pp.135A-148A.

Burgard, D.A., Dalton, T.R., Bishop, G.A., Starkey, J.R. and Stedman, D.H., 2006b. Nitrogen dioxide, sulfur dioxide, and ammonia detector for remote sensing of vehicle emissions. *Review of Scientific Instruments*, 77(1), p.014101. DOI: 10.1063/1.2162432.

Carslaw, D.C. and Priestman, M., 2015. Analysis of the 2013 vehicle emission remote sensing campaigns data. King's College London Report to UK Defra. Online at: https://uk-air.defra.gov.uk/library/reports?report_id=831 (as of 12/01/2017).

Carslaw, D.C., Williams, M.L., Tate, J.E. and Beevers, S.D., 2013. The importance of high vehicle power for passenger car emissions. *Atmospheric Environment*, 68, pp.8-16. DOI: 10.1016/j.atmosenv.2012.11.033.

Chen, Y. and Borken-Kleefeld, J., 2014. Real-driving emissions from cars and light commercial vehicles–Results from 13 years remote sensing at Zurich/CH. *Atmospheric Environment*, 88, pp.157-164. DOI: 10.1016/j.atmosenv.2014.01.040.

DeFries, T.H., 2016. Evaluation of EDAR for Measurement of Simulated Exhaust Emissions. Eastern Research Group, Inc., January 2, 2016.

DeFries, T.H., Kishan, S., Sidebottom, J.A., Kemper, J., and Fulper, C., 2017. Preliminary Evaluation: HEAT EDAR. 2017 PEMS Conference at CE-CERT, Riverside, California, March 30, 2017.

ESP (Environmental Systems Products) Holdings, 2005. RSD4600 Remote Sensing Device. Operator's Manual. Edition 3, Tucson, AZ, 2005.

ESP (Environmental Systems Products) Holdings, 2010. Smoke Factor Measurements with Remote Sensing Device Technology. Recommended Practice. ESP report and guidance document, Tucson, AZ, 2010.

Franco, V., Kousoulidou, M., Muntean, M., Ntziachristos, L., Hausberger, S. and Dilara, P., 2013. Road vehicle emission factors development: A review. *Atmospheric Environment*, 70, pp.84-97. DOI: 10.1016/j.atmosenv.2013.01.006.

Frey, H.C. and Eichenberger, D.A., 1997. Remote Sensing of Mobile Source Air Pollutant Emissions: Variability and Uncertainty in On-Road Emissions Estimates of Carbon Monoxide and Hydrocarbons for School and Transit Buses (No. FHWA/NC/97-005). Center for Transportation Engineering Studies, Department of Civil Engineering, North Carolina State University.

Full, G. and ESP (Environmental Systems Products) Holdings, 2009. System and method for remote emissions sensing including calculation and calibration techniques compensating for temperature and pressure effects. U.S. Patent 7,485,861.

Giechaskiel, B., Vlachos, T., Riccobono, F., Forni, F., Colombo, R., Montigny, F., Le-Lijour, P., Carriero, M., Bonnel, P. and Weiss, M., 2016. Implementation of Portable Emissions Measurement Systems (PEMS) for the Real-driving Emissions (RDE) Regulation in Europe. JoVE (Journal of Visualized Experiments), (118), pp.e54753-e54753.

Guenther, P.L., Stedman, D.H., Bishop, G.A., Beaton, S.P., Bean, J.H. and Quine, R.W., 1995. A hydrocarbon detector for the remote sensing of vehicle exhaust emissions. *Review of Scientific Instruments*, 66(4), pp.3024-3029. DOI: 10.1063/1.1146498.

Hager, J.S. 2015. Vehicle Remote Sensing - Next Generation – Results. PEMS Conference & Workshop no. 5. 25th – 26th March, 2015, University of California Riverside.

Hart, C., Hawkins, D., Fulper, C., Stanard, A., DeFries, T.H., Kishan. S., Glinsky, G., Reek, A., Hager, J.S. 2015. Canister Degradation Study. 25th CRC On-Road Vehicle Emission Workshop. Long Beach, California, 22nd-25th March, 2015.

Hoshizaki, H., Wood, A.D. and Kemp, D.D., 1973. Final Report Vehicle Inspection Instrumentation. Lockheed Palo Alto Research Laboratory.

Jack, M.D., Ahlgren, W., Alves, J.F. and Palen, E.J. (1995) Remote and On-Board Instrumentation for Automotive Emissions Monitoring. 75th Annual Transportation Research Board Meeting, Washington, DC.

Ježek, I., Drinovec, L., Oprešnik Rodman, S., Ferrero, L., Carriero, M., Westerdahl, D., Katrašnik, T., Močnik, G. 2016. Number Concentration and Nitrogen Oxide Vehicle Emission Factors: On-Road Chasing Campaign Results. PEMS Conference & Workshop no. 6. 17th-18th March, 2016, University of California Riverside.

Jimenez-Palacios, J.L., 1998. Understanding and quantifying motor vehicle emissions with vehicle specific power and TILDAS remote sensing (Doctoral dissertation, Massachusetts Institute of Technology).

Jiménez, J.L., Koplow, M.D., Nelson, D.D., Zahniser, M.S. and Schmidt, S.E., 1999. Characterization of on-road vehicle NO emissions by a TILDAS remote sensor. *Journal of the Air & Waste Management Association*, 49(4), pp.463-470. DOI: 10.1080/10473289.1999.10463814.

Kraan, T.C., Baarbe, H.L., Eijk, A.R.A, Stelwagen, U. and Vonk, W.A., 2012. Consistency tests of remote sensing for vehicle exhaust emissions, 19th Transport and Air Pollution Conference, Thessaloniki, Greece, 26 - 27 November 2012.

Lawson, D.R., Groblicki, P.J., Stedman, D.H., Bishop, G.A., and Guenther, P.L., 1990. Emissions from in-use motor vehicles in Los Angeles: a pilot study of remote sensing and the inspection and maintenance programs, *Journal of the Air & Waste Management Association*, 40 (8), 1096-1105.

Marrero, T.R., and Mason, E.A, 1973. Gaseous diffusion coefficients. *Journal of Physical and Chemical Reference Data*, 1.1 (1972): 3-118.

Mazzoleni, C., Kuhns, H.D. and Moosmüller, H., 2010. Monitoring automotive particulate matter emissions with LiDAR: A review. *Remote Sensing*, 2(4), pp.1077-1119.

McClintock, P., 2011. Enhanced remote sensing performance based pilot program. Environmental Systems Products Inc, Tiburon, CA, USA.

Measures, R.M. and Pilon, G., 1972. A study of tunable laser techniques for remote mapping of specific gaseous constituents of the atmosphere. *Optical and Quantum Electronics*, 4(2), pp.141-153.

Menzies, R.T. and Tratt, D.M., 2003. Differential laser absorption spectrometry for global profiling of tropospheric carbon dioxide: selection of optimum sounding frequencies for high-precision measurements. Applied optics, 42(33), pp.6569-6577.

Moosmüller, H., Mazzoleni, C., Barber, P.W., Kuhns, H.D., Keislar, R.E. and Watson, J.G., 2003. On-road measurement of automotive particle emissions by ultraviolet lidar and transmissometer: Instrument. *Environmental science & technology*, *37*(21), pp.4971-4978. DOI: 10.1021/es034443p.

Popp, P.J., Bishop, G.A. and Stedman, D.H., 1999. Development of a high-speed ultraviolet spectrometer for remote sensing of mobile source nitric oxide emissions. *Journal of the Air & Waste Management Association*, 49(12), pp.1463-1468. DOI: 10.1080/10473289.1999.10463978.

REVEAL (Remote Measurement of Vehicle Emissions at Low Cost) 2002. Recommendations for use in Europe of Remote Sensing Devices (RSDs) for the Measurement of Motor Vehicle Exhaust Emissions. EU Project 1999-RD.10657 Key Deliverable 11. Sira Limited and Project Partners.

Ropkins, K. 2016. pems.ultils: Portable Emissions (and Other Mobile) Measurement System Utilities. R software package version 0.2.17.8. Current release version available under General Public Licence from CRAN (the Comprehensive R Archive Network) at: https://cran.r-project.org/web/packages/pems.utils/. For early release developer’s version (mailto: k.ropkins@its.leeds.ac.uk).

Ropkins, K., Beebe, J., Li, H., Daham, B., Tate, J.E., Bell, M.C. and Andrews, G.E. 2009. Real-World Vehicle Exhaust Emissions Monitoring: Review and Critical Discussion. *Critical Review in Environmental Science and Technology*. 39(2), pp.79-152. DOI: 10.1080/10643380701413377.

Ropkins, K., Oates, C., Tate, J.E. 2008. Evaluation of a Remote Sensing System ‘Dirty Emitter’ Measurement. 18th CRC On-Road Vehicle Emission Workshop. San Diego, US, 31st March – 2nd April, 2008.

Sjödin, Å. and Andréasson, K., 2000. Multi-year remote-sensing measurements of gasoline light-duty vehicle emissions on a freeway ramp. *Atmospheric environment*, 34(27), pp.4657-4665. DOI: 10.1016/S1352-2310(00)00158-8.

Stanard, A.P. and DeFries, T.H., 2014. Canister Degradation Study: Methods Development Shootout Results. Final Report prepared for U.S. EPA by Eastern Research Group Inc., July 7, 2014.

Stedman, D.H. and Bishop, G.A., 2002. Opacity enhancement of the on-road remote sensor for HC, CO and NO. Final Report prepared for CRC-E56-2.

Stedman, D.H. and Bishop, G.A., Colorado Seminary, 1994. Apparatus for remote analysis of vehicle emissions. U.S. Patent 5,319,199.

Stedman, D.H., Bishop, G., McLaren, S., Colorado Seminary, 1995. Apparatus for remote analysis of vehicle emissions. U.S. Patent 5,401,967.

Stephens, R.D. and Cadle, S.H., 1991. Remote sensing measurements of carbon monoxide emissions from on-road vehicles. *Journal of the Air & Waste Management Association*, 41(1), pp.39-46. DOI: 10.1080/10473289.1991.10466823.

Tang, M.J., Cox, R.A., and Kalberer, M. 2014. Compilation and evaluation of gas phase diffusion coefficients of reactive trace gases in the atmosphere: volume 1. Inorganic compounds. *Atmospheric Chemistry and Physics*, 14.17 (2014): 9233-9247.

US EPA (Environmental Protection Agency), 2015. Guidelines establishing test procedures for the analysis of pollutants (Part 136, Appendix B. Definition and procedure for the determination of the method detection limit-Revision 1.11, June 30, 1986): U.S. Code of Federal Regulations, Title 40, July 1, 2015 edition, p. 344–347.

Wang, J., Maiorov, M., Jeffries, J.B., Garbuzov, D.Z., Connolly, J.C. and Hanson, R.K., 2000. A potential remote sensor of CO in vehicle exhausts using 2.3 µm diode lasers. Measurement Science and Technology, 11(11), p.1576. DOI: 10.1088/0957-0233/11/11/306.

Zhang, Y., Bishop, G.A., Beaton, S.P., Guenther, P.L. and McVey, L.F., 1996. Enhancement of remote sensing for mobile source nitric oxide. *Journal of the Air & Waste Management Association*, 46(1), pp.25-29. DOI: 10.1080/10473289.1996.10467437.

Zhang, Y., Stedman, D.H., Bishop, G.A., Guenther, P.L. and Beaton, S.P., 1995. Worldwide on-road vehicle exhaust emissions study by remote sensing. *Environmental science & technology*, 29(9), pp.2286-2294. DOI: 10.1021/es00009a020.