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Vehicle Exhaust Remote Sensing Device Method to Screen Vehicles for Evaporative Running Loss Emissions

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Abstract

Vehicle hydrocarbon (HC) emissions can be emitted from either tailpipe or non-tailpipe locations and understanding their fleet apportionment is important for successful air pollution policy. Vehicles initially misidentified as having elevated tailpipe HC emissions first indicated that roadside exhaust sensors could detect the presence of evaporative HC emissions as increased noise in the HC/carbon dioxide (CO₂) correlation measurement. The 90th percentile of the largest residual of the HC/CO₂ correlation is defined as a running loss index (RLI) for each measurement. An RLI that is three standard deviations or greater above the instruments noise

indicates possible evaporative running loss emissions with the probability increasing with larger RLI values. Two databases of vehicle emission measurements previously collected in West Los Angeles in 2013 and 2015 were screened using this method. The screening estimated 0.09% (31/33,806) and 0.18% (49/27,413) of the attempted measurements indicated evaporative running loss emissions from a 9-year-old fleet. California LEV I certified vehicles (1994 – 2003 model years) accounted for the largest age group for both. Minimum detection limits for the instrument used were estimated at 2.8 and 1.6 g/mile on a propane basis for the 2013 and 2015 data respectively or 32 to 56 times the Federal Tier 2 and Tier 3 standards of 0.05 g/mile.

INTRODUCTION

With the continuing advances in vehicle combustion management, after-treatment technologies and improved reliability, on-road vehicle tailpipe emissions in the United States (U.S.) have experienced profound reductions.¹⁻³ Fuel based emission inventory estimates for Los Angeles, Houston and New York City have shown that carbon monoxide (CO) and non-methane hydrocarbon emissions, predominantly from vehicles, decreased by 80 to 90% between 1990 and 2010 though fuel use increased.⁴ Over this same time period gasoline engine oxides of nitrogen (NO_x) emissions are estimated to have decreased by 65% in Los Angeles.⁵ The California Vehicle Surveillance Program similarly found that tailpipe volatile organic compound (VOCs) emissions of California vehicles decreased by 80% between 1995 and 2003.⁶

These reductions have been driven by the need for many areas in the U.S. to meet National Ambient Air Quality Standards (NAAQS) as part of the requirements of the Federal Clean Air Act.^{7,8} Mobile source contributions to several NAAQS, such as CO, nitrogen dioxide (NO₂) and sulfur dioxide (SO₂), have been accomplished through controlling the direct emissions of these

species and removing sulfur from fuels. Ozone, however, is a pollutant not directly emitted by vehicles but is a non-linear product of the mixture of VOCs, nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) and sunlight that has proven to be difficult to reduce. In California's South Coast Air Basin, vehicles are major sources of VOCs and NO_x emissions, and while ozone concentrations have been reduced significantly over the last 50 years, unhealthy levels still persist in many parts of the basin.^{9, 10} The recent decrease of the eight-hour ozone NAAQS to 70ppb will likely mean that the South Coast Air Basin will remain out of compliance for several more decades requiring additional reductions in VOC and NO_x emissions to comply.¹¹

Mobile source VOC emissions from the tailpipe are due to incomplete combustion from cold starts or combustion management problems and from non-tailpipe sources that involve fuel evaporation, leaks or permeation. In the U.S., light-duty vehicles are certified for evaporative emissions under three modes, running losses (liquid or vapor leaks while in operation), hot soaks (liquid or vapor leaks from a fully warmed up vehicle that is subsequently parked with the engine off), and diurnal emissions (liquid or vapor losses from a parked vehicle over a 24hr ambient temperature cycle). In-use testing of vehicles has demonstrated that evaporative emissions, like tailpipe emissions, have also declined significantly. The California Vehicle Surveillance Program found that hot-soak emission rates for the majority of hydrocarbon compounds found in California gasoline decreased by more than 95% between the 1999 and 2003 California vehicle fleets.⁶ These improvements have continued with the introduction of U.S. Tier 2 vehicles as recent research has shown evaporative emission control efficiencies in excess of 98%.¹²

As emissions have decreased, the relative contributions to the atmosphere of evaporative and tailpipe emissions have been studied and debated. Pierson et al.'s review in the late 1990's concluded that 65 to 93% of ambient non-methane hydrocarbon emissions were attributable to

the tailpipe.¹³ Into the early 2000's, measurements still estimated that tailpipe emissions constituted the majority of the mobile source hydrocarbon (HC) emissions.^{14, 15} The gap between the fraction of the total HC's predicted by computer models to be contributed by evaporative emissions and tailpipe emissions has continued to shrink. The current California vehicle emissions computer model (EMFAC2017) predicts for the South Coast Air Basin for the gasoline fleet in the summer of 2015 that the two groups are approaching equal contributions. Tailpipe emissions (running, idle and starting) of total organic gases are estimated to be 49% of the total, and non-tailpipe emissions (running losses, resting losses, diurnal and hot soak) are 41% (diesel and natural gas vehicles make up the remainder, see supporting information).¹⁶ However, analysis of vehicle emissions and ambient measurements have disagreed over whether this is the case.^{17, 18} This is an important distinction to resolve as the two sources can contribute different classes of compounds which may affect the results from air quality models and forecasts differently, especially when control of ozone is desired.¹⁹

One piece of information that would be helpful in addressing this question would be to know the fraction of in-use vehicles suspected of having measurable running loss emissions. U.S. certification tests for evaporative emissions involve a temperature controlled SHED which can be sealed with the test vehicle inside and the necessary instruments to record the amount and type of hydrocarbons emitted during the test.²⁰ The accuracy and precision of this certification test is unmatched but the time and cost required preclude its use on large numbers of vehicles and recruiting a representative fleet for this type of testing is not realistic. In the U.S., surveys have been conducted at State Inspection and Maintenance (I/M) stations using a handheld combustible gas detector for recruitment purposes or as a fleet survey mechanism. While I/M stations are a logical sampling location and the handheld sniffers are inexpensive, most states

exempt large numbers of new and old vehicles from such testing, and the testing frequency is low (typically biennial) limiting the ability to survey a representative in-use fleet. On-road vehicle exhaust remote sensing devices (RSD) allow for a large number of vehicles to be unobtrusively tested, and measurements collected with this method have successfully demonstrated an ability to define the fleet-wide emission distributions.^{9, 21-23} Development of an evaporative emissions screening tool for RSDs could expand its usefulness allowing an in-use fleet evaporative emissions survey.

In the spring of 2013 and 2015 the University of Denver collected on-road emission measurements from an in-use fleet that uses the La Brea Ave. and I-10 interchange in West Los Angeles as part of a California Air Resources Board sponsored project (see the supporting information for a measurement summary of these campaigns).²⁴ Vehicle emission measurements have been collected at this site since 1999 and it was chosen because it collects vehicles from many areas across the Los Angeles Basin and is isolated from significant local traffic eliminating cold start vehicles. Using the Fuel Efficiency Automobile Test (FEAT) instrument the 2013 and 2015 measurements included the collection and storage of the high frequency data as part of each vehicle's emissions measurement. These individual measurement files were post-processed in this study to search the West Los Angeles fleet for vehicles suspected of running loss emissions using a new evaporative emission screening method enabling for the first time an estimate of the fleet fraction suspected of having measurable running loss emissions.

MATERIALS AND METHODS

FEAT is an RSD developed and used by the University of Denver that is composed of a dual beam infrared (IR) and ultraviolet (UV) light source located on one side of a single lane road and

a multi-wavelength detector unit opposite it with the beams approximately 36cm above the roadway. Four nondispersive lead selenide IR detectors provide a reference channel (3.9 μ m), CO (3.6 μ m), carbon dioxide (CO₂) (4.3 μ m), HC (3.3 μ m) and two dispersive ultraviolet spectrometers measure NO, SO₂ and ammonia (NH₃) between 198 to 227nm while the second records NO₂ spectra between 430 and 450nm. The 3.3 μ m region used for detecting HC emissions corresponds to the C–H stretching region and while it is most sensitive to alkane species more commonly found in evaporative emissions, the absorbance values reported by the detector are the sum of all the species found in vehicle emissions.²⁵ The HC detector is calibrated with known amounts of propane and all HC measurements are reported as propane equivalents. Absorbance measurements on all of the detectors, which are background corrected using the air in front of the vehicle, are collected at 100Hz and the system and methods have been fully described in the literature.²⁶⁻²⁸

In the mid-2000's, researchers at the Colorado Department of Public Health and Environment (CDPHE) found evidence that RSD's could possibly detect running loss emissions from vehicles. Several RSD-identified in-use high HC emitters brought to state I/M Technical Centers for confirmatory exhaust emissions tests were found to easily pass the tailpipe standards but were subsequently found to have gasoline vapor and/or liquid fuel leaks. CDPHE staff corroborated these previous observations by performing several simple experiments using test vehicles with deliberately defeated evaporative emissions control systems to challenge a commercial RSD instrument that resulted in the successful detection of the evaporative emissions.

RSD instruments were designed to exploit the observation that a vehicle's exhaust constituents are well mixed when they leave the tailpipe and that, despite any associated turbulence, will disperse proportionally over short time scales (< a few seconds). This allows the various exhaust

species to be correlated against CO₂ with the slope of this linear correlation corresponding to the fuel specific emissions for that species.²⁸ What the CDPHE researchers discovered was that when there is an additional source of HC that is not associated with combustion emissions from the tailpipe it can disrupt the linear correlation between HC and CO₂. When a vehicle exits the FEAT sensing beam it triggers the collection of 100 Hz data on all detector channels for 0.5s behind the vehicle. Figure 1A shows an example of this time series for CO, CO₂ and HC collected behind a 1967 sedan measured on-road in Los Angeles. The initial rapid increase in emissions for all three species is followed by their decline as they disperse behind the vehicle. Figures 1B and 1C are the resulting correlation plots for CO versus CO₂ and HC versus CO₂. Figure 1B shows the expected linear correlation between CO and CO₂ with the slope of the best-fit line adequately describing their relationship. Figure 1C shows the lack of agreement between the data and the best-fit line due to the likely introduction of an additional source of HC not correlated with exhaust CO₂ that overlaps with the tailpipe HC emissions.

Based on CDPHE's early findings, they conducted a large staged running loss testing study, over 3500 metered propane releases from test vehicles, to investigate the possibility of using RSD to detect and quantify in-use running loss emissions (see the supporting information). Eastern Research Group used the data to develop a running loss index (RLI) as a proxy that ranks any scatter caused by HC plumes not originating from the vehicle's tailpipe.²⁹ After the linear fit is determined for the HC versus CO₂ correlation plot, fitting residuals are calculated and are the measured HC value minus the fit predicted HC value (see equation 1). The residuals are then ordered from largest to smallest and the value of the 90th percentile is the RLI value expressed in parts per million-centimeter (see the residual plot for the data in Figure 1C in Figure S1 in the supporting information) for that vehicle. Using the 90th percentile as opposed to the

Figure 1. A) Percent emissions versus time for a 1967 sedan measured at the West LA site in 2015 with the University of Denver's FEAT instrument. Percentages are calculated assuming that all of the vehicle's exhaust has been compressed into an 8cm cell. Correlation graphs for %CO versus % CO_2 (B) and %HC versus % CO_2 (C) for the data in graph A and least squares best fit lines are shown. The lack of correlation between the observed HC and CO_2 emissions resulted in an invalid HC exhaust measurement as determined by the FEAT software.

largest residual increases the level of certainty in detection and reduces the occurrence of false positives.

$$HC(ppm \cdot cm)_{\text{residual}} = HC(ppm \cdot cm)_{\text{measured}} - HC(ppm \cdot cm)_{\text{predicted}} \quad (1a)$$

$$RLI(ppm \cdot cm) \equiv \{HC(ppm \cdot cm)_{\text{residual}}\}^{90\text{th Percentile}} \quad (1b)$$

Results from the running loss staged testing data indicated that the RLI value is only dependent on the vehicle's running loss emissions rate and the RSD instrument noise (see the supporting information). Because the running loss signal and the instrument noise are both random variables, we described their combined effects on the measured RLI using the square root of the sum of their squares:

$$RLI(ppm \cdot cm) = \sqrt{RLI_{\text{Signal}}^2 + RLI_{\text{Noise}}^2} \quad (2)$$

This relationship has good properties for the two asymptotic behaviors of RLI: 1) when $RLI_{\text{Signal}} \gg RLI_{\text{Noise}}$, $RLI = RLI_{\text{Signal}}$ and 2) when $RLI_{\text{Noise}} \gg RLI_{\text{Signal}}$, $RLI = RLI_{\text{Noise}}$. Regression modeling using the staged testing data was used to investigate the dependencies of RLI_{Signal} . Exhaust HC was not expected to influence the running loss signal, but a term was included in the model statement in the event that an effect was present. The resulting regression model statement for the running loss signal is given by:

$$RLI_{\text{Signal}} = \exp[\text{Location} + a \cdot \ln(\text{Propane}) + b \cdot \ln(\text{Speed}) + c \cdot \text{Exhaust_HC}] \quad (3)$$

where the location is a categorical variable for the propane release point, Propane is the flow rate in g/hr, Speed is the vehicle speed in miles/hr and Exhaust_HC is the tailpipe exhaust HC concentration in parts per thousand propane.

The RLI_{Noise} value is dominated by the instrument's electronic noise, which was found to change over time, but there can be other measurement or environmental factors that may impact this component. The staged testing found that the slope of the exhaust HC vs. CO_2 correlation also influences instrument noise and while speed was not expected to be a factor a term was included in the model statement in case an effect was present. The resulting regression model statement for the noise signal is:

$$RLI_{Noise} = \text{Instrument} + d * \text{abs}(\text{Exhaust_HC}) + e * \text{Speed} \quad (4)$$

where the Instrument coefficient is a categorical variable for the specific instrument being used, Exhaust_HC is the tailpipe exhaust HC concentration in parts per thousand propane and Speed is the vehicle speed in miles/hr. When exhaust HC concentrations are zero, the zero slope can be either positive or negative, because zero measurements are normally distributed around zero, necessitating using the absolute value (abs) of this term.

Regression modeling of equations 3 and 4 using the staged testing data found that all the emissions release locations displayed a major response but without a significant difference between the nine tested locations. In addition, coefficients a, b, c and e were found to not be significantly different from 1, -1, 0 and 0, respectively (see Tables S8 and S9 and the discussion in the supporting information). With these substitutions, equation 3 reduces to:

$$RLI_{Signal} = 22.8 \text{ ppm}\cdot\text{cm} / \text{g/mile} * \text{PropaneEmissionRate (g/mile)} \quad (5)$$

showing that RLI_{Signal} is simply proportional to the running loss emissions rate in g/mile, independent of vehicle speed, leak location and body shape. Equation 4 reduces to:

$$RLI_{Noise} = \text{Instrument} + 13.2 \text{ ppm}\cdot\text{cm} / \text{ppthC}_3 * \text{Exhaust_HC (ppthC}_3) \quad (6)$$

where the noise component is a function of the instrument coefficient and exhaust HC concentration. When the Exhaust_HC of the vehicle is zero then RLI_{Noise} is equal to the Instrument noise coefficient allowing RLI_{Noise} to be calculated by averaging RLI values of the newest model year vehicles and assuming their running loss emissions are close to zero ($RLI_{Noise} \gg RLI_{Signal}$).

Since for the majority of vehicles the instrument noise (RLI_{Noise}) will generally be larger than the measured RLI, it is not possible to solve equation 2 for the running loss emissions rate. However, the RLI values can be used to rank the probability that a vehicle's measured RLI is higher than what would be measured if the vehicle had a zero running loss emission rate. The dependence between RLI and a vehicle's tailpipe HC emissions arises in part because as the HC/CO₂ slope increases with tailpipe emissions, the magnitude of the fitting residuals will also increase even for well-correlated data. Instead of correcting the RLI values, the RLI values are binned using bin boundaries that are dependent on the observed HC tailpipe emissions. This compensates for any positive bias introduced with increasing HC emissions. Bin values for each measurement are the number of standard deviations that the transformed measured RLI is above the transformed RLI for a vehicle with a zero running loss emissions and a comparable exhaust tailpipe HC concentration (see supporting information). As the bin number increases so does the probability of the vehicle having running loss emissions.

We tested this hypothesis using hot soak emissions data collected at the two Colorado Inspection and Maintenance (I/M) stations in 2008.²⁹ 238 vehicles were recruited using a stratified sampling plan after being evaluated for running loss emissions by a commercial RSD instrument as they entered the stations. The vehicles were driven on the road for at least 15 minutes and returned to the test locations where two additional RLI measurements were

collected. The vehicle's engine was shut-off and it was rolled into a temporary (plastic and duct tape) portable hot soak shed (PSHED) and the circulation fans started. The PSHED was sealed and total HC concentration, temperature and barometric pressure measurements were recorded for the next 15 minutes.

The RLI values assigned each vehicle to Bins -2 to 6 and the resulting distribution is shown in Table 1 and is compared against the PSHED results. Vehicles assigned to RLI Bins -2, -1, and 0 were found to have a 9% chance (21 of 244 measurements) of having a PSHED hot soak emissions of greater than 1 gram VOCs in 15 minutes (the U.S. EPA estimates that Tier 2 vehicles should have less than 0.3g/15 minutes). 89% (51 of 57) of the vehicles with an RLI Bin of 4, 5, or 6 were found to exceed this level. As hypothesized, the probability of finding vehicle evaporative emissions increased with increasing RLI bin number. Since the I/M program exempted the newest seven model years from testing, the fleet represented in Table 1 is not representative of the on-road fleet. Applying this probability distribution to an on-road fleet should significantly reduce the 9% probability in Bins -2 to 0 due to the large number of newer vehicles that would be included while having less effect on the higher bins.

Generally, the FEAT remote sensing setup does not record the high frequency data (see Figure 1A) for each individual vehicle as it slows the response time of the instrument. For the 2013 and 2015 West Los Angeles measurements, which were collected on the on-ramp from southbound La Brea Ave to eastbound I-10, we were asked by the California Air Resources Board to collect and store the high frequency data.²⁴ This resulted in two data sets, the first collected between Saturday April 27 through Saturday May 4, 2013 with 33,806 attempted measurements and 27,247 with valid tailpipe emission measurements and vehicle registration data. The second was

Table 1. Comparison of RLI Measurements and PSHED Hot Soak Tests

RLI Bin	PSHED Tests		Total Tests	Percent of Tests ≥ 1 g/15 min
	< 1 g/15 min	≥ 1 g/15 min		
-2, -1, 0	223	21	244	9%
1	90	33	123	27%
2	25	36	61	59%
3	11	43	54	80%
4	4	27	31	87%
5	0	17	17	100%
6	2	7	9	78%

collected between Saturday March 28 through Friday April 3, 2015 with 27,413 attempted measurements and had 22,124 valid and registration matched records.

Each attempted measurement included a companion comma delimited file that contained the high frequency data. These files contain the date and time of the measurement, the IR detector normalization voltages (used for normalizing to the background concentrations found in front of the car), 50 voltages (every 10ms) for the IR reference, CO, CO₂ and HC detectors and the 50 molar concentrations for CO, CO₂, and HC derived from the voltages. Each ultraviolet spectrophotometer supplies 50 measured concentrations and uncertainty estimates for the NO, NH₃, SO₂ and NO₂ species. The file also contains the final emission results of the original analysis, validity flags and the measured speed and acceleration if valid. Digital images of the rear of each vehicle with a valid tailpipe emissions measurement were saved with the 2013 data set while the images for all of the measurements (valid or invalid) were recorded for the 2015 data. We developed a program to post process these files to recalculate the original fits and

results, calculate an RLI index value and assigned that value to its designated bin for each record. For this study, because the standard validity criteria for exhaust measurements could invalidate measurements for vehicles with running loss emissions, we expanded our screening to include all vehicles, not just those we were able to successfully measure the tailpipe emissions.

RESULTS AND DISCUSSION

Table 2 shows the measurement distribution and the RLI means values for each bin obtained from the RLI screening of all the 2013 and 2015 attempted measurements at the West Los Angeles location. All bins less than or equal to Bin 0 have been combined into Bin 0. Screening the high frequency files found that 96.9% and 92.9% of the attempted measurements had RLI measurements that resulted in Bin values of 1 or less and are not expected to have any running loss emissions for the 2013 and 2015 data sets respectively. Conversely where we believe there is a higher probability of running loss emissions, Bins 3 and above, the RLI screening identified 83 (~0.2%) and 656 (2.4%) of the measurements in 2013 and 2015 respectively. The increase between 2013 and 2015 is the result of the RLI_{Noise} coefficient increasing by 30% on a single day (March 30th) above the averaged noise value used in the bin calculations due to unexplained negative voltage deflections in the HC channel; increasing the number of vehicles in Bins 2, 3 and 4 that were not visually confirmed. Of the 31 visually confirmed vehicles in 2013 6 have no registration information and similarly 15 out of the 49 visually confirmed vehicles in 2015.

The RLI screening is a way to search for added noise in each vehicle's tailpipe correlation measurement between HC and CO₂ caused by the addition of non-tailpipe HC. However, there are other sources of measurement noise, electronic and partial physical blockages of the sampling beam for example, which can interfere with the correlation measurements that are not

Table 2. RLI Measurements and Visual Screening Results for the 2013 and 2015 Data Sets.

RLI Bin	2013		2015	
	Measurements (%) Mean RLI (ppm•cm)	Visual Confirmation Mean RLI (ppm•cm)	Measurements (%) Mean RLI (ppm•cm)	Visual Confirmation Mean RLI (ppm•cm)
≤0	26816 (79.32%) 26		21390 (78.10%) 17	
1	5966 (17.64%) 43		4050 (14.79%) 28	
2	972 (2.88%) 62	13 (1.3%) 133	1286 (4.69%) 41	5 (0.4%) 251
3	72 (0.21%) 105	14 (19.4%) 114	483 (1.76%) 63	10 (2.1%) 218
4	12 (0.04%) 688	4 (33.3%) 1616	160 (0.58%) 140	17 (10.6%) 598
5	1 (0.01%) 232	0	14 (0.05%) 192	9 (64.3%) 224
6	0		6 (0.02%) 256	5 (83.3%) 267
7	0		3 (0.01%) 408	3 (100%) 408
Totals	33,806 (100%) 30	31 (0.1%) 315	27,392 (100%) 21	49 (0.2%) 371

caused by gas absorption. We therefore designed a graphical display program to view the high frequency data files and confirm the presence of an added HC signal. Due to the manageable number of suspected vehicles found, we chose to review all of the measurements that were categorized in Bin 2 or higher. This process is necessarily subjective; however, exclusions due to identifiable reasons other than running loss emissions are important for deriving the final numbers and establishing guidelines for any future studies. The results of the visual inspection of the measurements in RLI Bin 2 and higher are also included in Table 2 (see Tables S11 and S12 in supporting information for detailed list). As expected, very few Bin 2 values (1.3% in 2013 and 0.4% in 2015) were judged to have any possible running loss emissions. As found with the PSHED testing, the percentage of vehicles flagged by the RLI screening and visually confirmed increased with increasing bin number for both years' data sets.

For the visually confirmed vehicles identified by registration data, California LEV I vehicles (1994 – 2003 model years) account for the largest age group for both years (13 in 2013 and 14 unique vehicles in 2015 with 16 records as one vehicle was identified three times). This represents 52% (13/25) and 47% (16/34) of the visually confirmed and plate-matched vehicles in 2013 and 2015 respectively (see Tables S11 and S12 in the supporting information). For the 2013 and 2015 data sets there were 6 and 9 (this does not count the 1936 coupe visually identified) vehicles respectively in each of the 2004 & newer and 1993 & older age groups. The 15 vehicles without registration information identified in 2015 that we have plate images for are composed of 3 cars and 11 trucks.

Fewer vehicles were suspected of running loss emissions in 2013 than in 2015 and these differences are in part the result of higher observed instrument noise levels in 2013. The regression results (see Table S9 in the supporting information) can be used to estimate the

minimum detection limit (MDL) as defined by 40 CFR Part 136 Appendix B for each FEAT data set.³⁰ This provides an emissions rate floor for those vehicles visually confirmed and found in Bins 3 and higher. The estimated running loss MDL's for the FEAT 3002 instrument used for both campaigns are 2.8 and 1.6 g/mile on a propane basis for the 2013 and 2015 data respectively. For comparison, the Federal Tier 2 and 3 running loss standard is 0.05 g/mile.

All of the vehicles visually identified possessed emission patterns that suggest the presence of running loss emissions (see the supporting information for a complete listing by measurement year). There were five prominent patterns found in the 2013 and 2015 data that we labeled 1) anti-correlated (19% and 14%, see Figure S14A), 2) consistently decreasing HC signal (36% and 25%, see Figure S14B), 3) HC exponential decay (16% and 20%, see Figure 1A), 4) rising or late burst of HC signal (26% and 29%, see Figure 3) and 5) high background (3% and 12%). Using the high frequency time series data, Figure S14 in the supporting information gives examples of the anti-correlated and the consistently decreasing HC signal for two of the suspected running loss emission vehicles. Figure S14A graphs the %CO₂ (—, left axis) and %HC (□, right axis) emissions versus time for a 1998 pickup truck measured in 2015 (RLI 246, Bin 5) showing the HC emissions falling as the CO₂ emissions rise and then having that pattern reverse before both species start to decrease together as they dilute behind the truck. This is one of three visually confirmed measurements for this vehicle in 2015. Figure S14B is a similar graph for a 2005 van measured in 2015 (RLI 92, Bin 3) again showing little correlation between the two species with the HC concentrations consistently decreasing during the 0.5s measurement. A third common pattern is an exponential decay (see Figure 1A for an example) that is similar to the pattern shown in Figure S14B but exhibits a steeper decline early in the measurement.

Since it was not possible to confirm actual running loss emissions from any of the vehicles identified by the RLI screening, we are left to present three proofs of concept identifications that we believe supports this contention. Figure 2 presents the plate image (A) and the emissions time series plot (B) for a pickup truck measured in 2015 (RLI 392, Bin 3). Clearly visible are a large liquefied gas (LP) tank in the bed of the truck and some type of equipment for dispensing tar attached at the rear. This vehicle had an unreadable license plate and was not included in the emissions database but was discovered by the RLI screening. The time series plot for HC and CO₂ (Figure 2B) shows apparent correlation during the first half of the measurement but the HC concentration steadily increases throughout the remainder of the measurement. The FEAT software invalidated the HC results because of the overall poor correlation. The HC/CO₂ correlation before 0.25s is also suspect for two reasons: 1) if you look carefully at 0.05s in the time series you can see that the HC concentration starts to rise before the CO₂ does indicating separate plumes and 2) the magnitude of the early correlation indicates a gross emitting HC vehicle (gHC/kg of Fuel > 200, 2015 fleet mean was 1.3) yet this vehicle has no tailpipe CO emissions. HC emissions at this level are indicative of a misfire and the unburned fuel would be expected to have been partially converted to CO on the catalytic converter until the available oxygen is consumed increasing the CO emissions. These two pieces of information suggest that the early correlation is happen-stance and occurs due to the turbulence behind the truck diluting the tar or LP emissions along with the tailpipe CO₂.

Figure 3 includes the plate image (A) for a 1936 antique car (identified by a car buff for the authors) measured in 2015 (RLI 213, Bin 4). This vehicle has more CO in its exhaust than CO₂ as might be expected for its age. Figure 3B are the time series plots for HC and CO₂ and shows a late burst in the HC emissions during the second half of the measurement indicating two HC

Figure 2. A) Rear image of a pickup truck with a propane tank and some type of asphalt patching system. The image includes counter-clockwise from the lower left, tailpipe %CO, %HC (stars indicate software invalidated this result), %NO, %CO₂, time and date. B) %CO₂ (solid line, left axis) and %HC (dashed line, right axis) versus time for this truck's measurement.

Figure 3. A) Rear image of an antique car, identified as a 1936 Coupe. The image includes counter-clockwise from the lower left, tailpipe %CO, %HC, %NO, %CO₂, time and date. B) %CO₂ (solid line, left axis) and %HC (dashed line, right axis) versus time for this car's measurement.

emission sources. With no catalytic converter and running very rich, there are undoubtedly HC emissions in the tailpipe exhaust as evidenced by the correlation in the first half of the measurement. This vehicle will also have a gas tank vented to the atmosphere that we suspect is behind the large increase in the HC concentrations during the second half of the measurement.

The final vehicle is a natural gas powered (identified by the visibility of the cylinder and its blue fill cap in the plate image) garbage truck measured in 2015 (RLI 103, Bin 4). The time series plots of HC and CO₂ in Figure S15B in the supporting information shows a burst of methane behind the vehicle well before the exhaust emissions show up around 0.3s indicating some type of leak in the vehicle's fuel system or storage container. An elevated exhaust may be behind the delay requiring additional time for the exhaust emissions to reach ground level.

These three vehicles were unknown prior to the RLI screening and not in the West Los Angeles 2015 emissions database because they lacked a readable license plate. While the pickup truck's tar pot may not technically qualify as a running loss emission and the antique car is not typical of today's modern vehicles, they both undoubtedly have a source of hydrocarbon emissions other than from the tailpipe that the screening process found while the third vehicle shows an HC emission in the absence of any exhaust CO₂. For comparison see Figure S16 in the supporting information for examples of two vehicles with low RLIs, one with high tailpipe HC and one with low.

As previously discussed, additional sources of noise can be introduced by the instrument or the vehicles that can elevate the RLI values and were eliminated as false positives during the visual inspection process. Vehicles can introduce noise through the presence of partial beam blockages during the 0.5s emission measurement. The FEAT instrument's reference detector (3.9 μm) is a critical signal used by the instrument to detect the difference between physical blockages of the

light beam and gas absorbance since there are no vehicle emissions that absorb in this wavelength region. As a quality check, if the software detects voltage reductions in this channel that exceed $\pm 2.5\%$ of the before-car values it eliminates these measurements from use in the correlation graphs. However, the multi-detector system is not perfectly twinned and beam blockages that result in a voltage drop larger than 2.5% but that are not large enough to trigger a measurement restart can produce outlying points that can inflate the RLI value. Figures S17 and S18, in the supporting information, show data from a medium-duty truck (RLI 43, Bin 2) that illustrates how a physical blockage of the beam during the measurement can inflate the RLI value and produce a false positive.

Assuming that all of the vehicles visually identified from each year's data sets are running loss emitters we found that in 2013 only 0.09% (31) of the 33,806 attempted measurements were suspected. This doubled in 2015 to 0.18% (49) of the 27,413 attempted measurements. Assuming the unidentified attempts have the same repeat measurement frequency as the plate matched vehicles, the running loss fleet percentages for unique vehicles increase to 0.13% (31/24583) and 0.22% (47/21574) for 2013 and 2015 respectively. Therefore, from this screening, between 0.1 and 0.2% of the in-use fleet at the West Los Angeles site in 2013 and 2015 are expected to have running loss emissions that are at least a factor of 32 to 56 times larger than the U.S. Federal Tier 2 and 3 standards.

Unfortunately, there has not been an attempt to directly apportion tailpipe and non-tailpipe VOC emissions since Gentner et al.'s work in 2005.¹⁵ Their measurements supported the idea that tailpipe HC emissions remain the dominant source in ambient air in the Los Angeles Basin. Warneke et al. analyzed over five decades of ambient measurements (1960 – 2010) and showed that the VOC/CO ratio has remained constant over this time period again emphasizing the link

with tailpipe emissions.³ Borbon et al. used ground-based and airborne VOC measurements collected in Los Angeles in 2010 to show that the butane/CO ratios were similar during the daytime and nighttime measurements.³¹ Since the heating during the day should elevate evaporative emissions this observation minimizes the importance of non-tailpipe evaporative emissions in the Los Angeles Basin. Our finding of only a small number of vehicles (0.1 – 0.2%) in a 9 year-old fleet in West Los Angeles with measurable running loss emissions is consistent with these ambient measurements.

This work has been a first attempt to produce a non-intrusive screening method applicable to large numbers of vehicles making it possible to estimate the size of an in-use fleet with elevated running loss emissions. Additional research is needed to expand on the confirmation testing previously conducted in Colorado to include vehicles identified on-road and further improve the method and to establish false positive and false negative detection rates.

ASSOCIATED CONTENT

Supporting Information.

The Supporting Information is available free of charge on the ACS Publications website at DOI:.

EMFAC2017 total organic gas for the South Coast Air Basin in 2015; Measurement summary statistics; %HC residuals plot; Residual listing; Emissions versus time with no running loss emissions; Correlation graphs for previous measurements; Emissions versus time with running loss emissions; Correlation graphs for the previous measurements; Test vehicle listing; Summary of propane running loss tests; Summary of liquid gasoline running loss tests; Summary of induced running loss tests; Running loss emission rate

effects; Speed impacts on running loss emissions; Leak location effects; RLI values at different running loss emission rates and speeds; Regression coefficients for first model; Regression coefficients for simplified model; Parity plot for simplified model; Frequency Histogram of transformed RLI values; Mean RLI values by model year; FEAT 2015 speed versus exhaust HC; RLI Bin values versus propane release rate; Examples of anti-correlated and decreasing HC patterns; Tag image and time series for a CNG garbage truck; Examples of a high and low exhaust HC and no running loss emissions; Measurement data for a truck visually excluded; Tag image of truck; Listing of 2013 vehicles visually confirmed; Listing of 2015 vehicles visually confirmed; (PDF)

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Supporting Information For:

A Vehicle Exhaust Remote Sensing Device Method to Screen Vehicles for Evaporative Running Loss Emissions

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Table S1. Total Organic Gases Results using EMFAC2017 for the South Coast Air Basin in 2015

<https://www.arb.ca.gov/emfac/2017/>

EMFAC2017 (v1.0.2) Emissions Inventory

Region Type: Air Basin

Region: SOUTH COAST

Calendar Year: 2015

Season: Summer

Vehicle Classification: EMFAC2011 Categories

Units: tons/day for Emissions

Fuel	RUNEX	IDLEX	STREX	TOTEX	DIURN	HTSK	RUNLS	RESTL	TOG_TOTAL
DSL	8.16	0.58	0.00	8.74	0.00	0.00	0.00	0.00	8.74
ELEC	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
GAS	40.54	0.23	28.40	69.17	10.41	11.25	28.47	7.92	127.22
NG	6.30	0.00	0.00	6.30	0.00	0.00	0.00	0.00	6.30
									142.26

Gasoline Running Exhaust 69.17 tons/day (49% of TOG_TOTAL of 142.26)

Gasoline Evaporative Emissions 58.05 tons/day (41% of TOG_TOTAL of 142.26)

Table S2. Summary Statistics for 2013 and 2015 West Los Angeles Measurements.

Dates	Attempts / Plates / Matched Mean Model Year Mean Speed (mph) Mean Acceleration (mph/s)	Mean (g/kg) of Fuel Emissions and SEMs ^a			
		CO	HC	NO ^b NO ₂ NO _x ^c	NH ₃
4/27 - 5/4 2013	33806 / 27808 / 27247 2004.7 21.9 -0.2	16.4 ± 0.6	2.2 ± 0.2	2.1 ± 0.1 0.15 ± 0.02 3.4 ± 0.1	0.58 ± 0.02
3/28 - 4/3 2015	27413 / 25908 / 22124 2006.9 18.8 1.2	13.2 ± 0.4	1.3 ± 0.1	1.8 ± 0.1 -0.06 ± 0.01 2.6 ± 0.2	0.72 ± 0.02

^aCalculated using a carbon mass fraction of 0.86. Standard error of the means (SEMs) are calculated using the daily measurements.

^bmoles of NO.

^cmoles of NO₂.

Figure S1. %HC residuals plotted versus %CO₂ for a 1967 sedan measured in Los Angeles in 2015 with the 90th percentile point marked by the arrow. The residuals are calculated using the least squares best fit line shown in Figure 1C in the manuscript text. The FEAT HC concentration data is recorded as percent but the RLI index by definition is expressed in ppm·cm. The 90th percentile and RLI index for this vehicle is 5,731, Bin 4.

Table S3. Measurements and Residuals for Figure S1 Rank Ordered by %HC Residuals with the 90th Percentile Highlighted.

Measurement Seconds	%CO ₂ (Measured)	%HC (Measured)	%HC (Best Fit)	%HC (Residual)
0.02	1.837	2.6555	0.4851	2.1703
0.01	2.044	2.5773	0.5160	2.0613
0.03	1.921	2.0522	0.4976	1.5546
0.04	2.289	1.3510	0.5528	0.7982
0.05	2.690	1.1859	0.6129	0.5731
0.06	2.926	1.1060	0.6483	0.4577
0.07	3.266	1.0261	0.6992	0.3268
0.08	3.624	1.0419	0.7529	0.2890
0.09	3.809	0.9157	0.7806	0.1351
0.34	1.776	0.4344	0.4760	-0.0416
0.1	3.851	0.7413	0.7869	-0.0456
0.26	2.079	0.4439	0.5213	-0.0774
0.33	1.896	0.4155	0.4938	-0.0784
0.35	1.744	0.3916	0.4712	-0.0796
0.14	2.971	0.5545	0.6550	-0.1006
0.11	4.175	0.7344	0.8355	-0.1011
0.25	2.095	0.4048	0.5237	-0.1189
0.27	2.116	0.3963	0.5268	-0.1305
0.28	2.142	0.3845	0.5307	-0.1462
0.32	1.980	0.3511	0.5064	-0.1553
0.36	1.844	0.3219	0.4861	-0.1642
0.13	3.575	0.5752	0.7455	-0.1703
0.31	2.000	0.3059	0.5096	-0.2036
0.5	1.407	0.2119	0.4206	-0.2087
0.18	2.394	0.3586	0.5686	-0.2099
0.29	2.063	0.3071	0.5190	-0.2118
0.3	2.085	0.3056	0.5222	-0.2166
0.37	1.980	0.2893	0.5064	-0.2171
0.2	2.264	0.3297	0.5490	-0.2193
0.19	2.200	0.3165	0.5395	-0.2230
0.15	3.029	0.4376	0.6637	-0.2261
0.12	4.249	0.6086	0.8465	-0.2379
0.49	1.524	0.2000	0.4381	-0.2381
0.48	1.632	0.2143	0.4544	-0.2401
0.21	2.445	0.3311	0.5762	-0.2451
0.47	1.728	0.2221	0.4687	-0.2466
0.44	1.789	0.2268	0.4778	-0.2510
0.45	1.846	0.2340	0.4864	-0.2524
0.41	1.693	0.2087	0.4635	-0.2547
0.38	2.044	0.2605	0.5161	-0.2556
0.42	1.698	0.2057	0.4642	-0.2585
0.46	1.791	0.2181	0.4782	-0.2601
0.17	2.694	0.3500	0.6134	-0.2634
0.43	1.682	0.1966	0.4618	-0.2651
0.24	2.182	0.2648	0.5368	-0.2720
0.39	1.999	0.2363	0.5094	-0.2730
0.4	1.818	0.2069	0.4822	-0.2753
0.16	3.042	0.3893	0.6656	-0.2764
0.22	2.605	0.2964	0.6002	-0.3038
0.23	2.455	0.2267	0.5777	-0.3509

Development of the Running Loss Index

In spite of turbulence, the optical absorbance (concentration*path length, ppm·cm) for all exhaust pollutants will be proportional to each other during a single on-road RSD measurement because the exhaust gases are well mixed, constant with time, and no reactions occur in the plume during the 0.5s measurement. Many RSD instruments take measurements every 10ms for 500ms or more after the light beam is unblocked by the vehicle. While different calculation algorithms can be used, the basic idea is that plots of exhaust HC (ppm·cm), CO (%·cm), or NO (ppm·cm) vs. CO₂ (%·cm) have a strong tendency to be straight lines with a slope proportional to the tailpipe exhaust HC, CO, or NO concentration (refer to Figures S2 and S3).

Running losses from a vehicle are hydrocarbons emitted from a non-tailpipe source on the vehicle. These include permeation from fuel system connections, breakthrough from the evaporative emissions control canister, a cracked rubber vapor line, or a leaking fuel-injector. Therefore, the running loss HC is likely not well-mixed with the tailpipe exhaust gas emissions plume. If a portion of the running loss emission plume wafts into the RSD light beam, some of the fifty HC absorbance measurements will have higher absorption values than if absent the running loss emissions. Figures S2 - S5 demonstrate the effect of running loss emissions on the RSD data for two runs of a test vehicle driven at 12 mph without and with 15 scfh (standard cubic feet per hour, 70 °F, 760 torr) propane running loss emissions added by simulating a leak at the fuel fill door. Figure S2 shows the time series of the first measurement pass with 1100 ppm of propane from the tailpipe exhaust and Figure S3 shows the resulting correlation graphs. All of the species measured show excellent correlation with CO₂.

Figures S4 and S5 show the results of the second pass of the test vehicle this time with an added simulated leak of 15 scfh of propane released from the fuel fill door. Figure S4 is the time series of species absorbance versus time. Notice that the HC and CO₂ concentrations at the beginning of the measurement appear well correlated but from 150 to 350ms the running losses produce a large increase in the HC absorbance that is not correlated with CO₂. Figure S5 is the resulting correlation graph for each species versus CO₂ and shows how the correlation of HC versus CO₂ has broken down.

Data Collection. The Colorado Department of Public Health and Environment (CDPHE) and Eastern Research Group (ERG) constructed an emissions test program to collect in-use drive-by emissions data on seven test vehicles (see Table S4) using a commercial ESP Series 4600 remote sensing instrument and standard data collection procedures. Vehicle 0 was the State of Colorado's on-road audit truck that has its exhaust pipe re-routed to release exhaust high above the truck cab so that the combustion gases have a low probability of being detected by the ground-level RSD instrument. The truck is equipped with a compressed gas cylinder rack that allows different known mixtures of dry simulated vehicle exhaust to be released during testing. For this testing three gas mixtures were used 1) 15.07% carbon dioxide (CO₂) in nitrogen, 2) 1100 ppm propane, 3% carbon monoxide (CO), 500 ppm nitric oxide (NO) and 12.92% CO₂ in

Figure S2. Emissions versus time for a tailpipe exhaust measurement of the test vehicle with no additional running loss emissions added.

Figure S3. Correlation graphs for the tailpipe exhaust measurement of the test vehicle without adding simulated running loss emissions.

Figure S4. Emissions versus time for a tailpipe exhaust measurement of the test vehicle with 15 scfh of propane released from the fuel fill door of the vehicle.

Figure S5. Correlation graphs for the tailpipe exhaust measurement of the test vehicle with added simulated running loss emissions.

Table S4. Test Vehicles used for Staged Running Loss Emissions Measurements.

Test Vehicle	Model Year	Make	Model
0	N/A	Audit Truck	N/A
1	2008	Ford	Escape
2	1992	Oldsmobile	Eighty-Eight
3	1982	Chevrolet	Caprice
4	1989	Dodge	Caravan
5	1992	Chevrolet	Corsica
6	1990	Chevrolet	Lumina

nitrogen and 3) 6015 ppm propane, 5% CO, 250 ppm NO and 11.55% CO₂ in nitrogen. The truck released these simulated dry exhaust gases at approximately 30 scfm (standard cubic feet per minute, 70 °F, 760 torr) to an artificial tailpipe located at the traditional tailpipe location for each measurement pass.

Equipment to produce metered, simulated running loss emissions was added to each of the test vehicles. Gaseous running loss emissions were produced by metering propane from a small bottle through a rotameter. Tubing from the rotameter routed the propane to a desired release location on the vehicle. Liquid gasoline running loss emissions were produced by releasing gasoline using a metering valve on a peristaltic pump via thin tubing to a release location on the vehicle. In general, the tubing was situated to drop the gasoline directly onto the pavement where it would not be run over by the tires of the vehicle. In addition to producing simulated gaseous running loss emissions, running loss emissions were attempted by defeating the evaporative emissions control systems on several of the test vehicles. Note that the presence of running loss emissions was not confirmed by independent measurements for these tests.

RSD measurement data were collected by driving the test vehicles past the instrument at approximately 12, 34, or 55 mph, with steady accelerator position, and with light acceleration to keep the vehicle specific power in the 5 to 20 kW/tonne range. The simulated running loss emissions, either propane gas or liquid gasoline, were released at different locations on the vehicles to evaluate the ability of RSD to detect running losses emitted from different points at the different speeds. For some tests, running loss emissions were induced by either removing the gas cap or disconnecting the vapor lines to the evaporative emissions control canister.

Each test condition was made up of twenty-eight to forty vehicle passes – twenty with the added running loss state (propane flowing, gasoline dripping, or evaporative emission control system defeated) and eight to twenty in the no-running-loss state (propane off, gasoline off, and evaporative emission control system operating). Within each test condition, the twenty measurements with running losses were conducted in groups of five runs with two to five runs without running losses interspersed between the groups. Table S5 shows the summary of the 138 test conditions and 3,501 RSD measurements using propane to simulate vapor running loss emissions. The flow of propane was started about 10 seconds before the measurement and was

Table S5. Summary of Propane Vapor Running Loss Tests Performed.

Test Vehicle	Measured Exhaust HC (ppmC ₃) Range ^a	Speed (mph)	Artificial Running Loss	
			Propane Emission Rate (scfh)	Release Location
0	-98 to 42	12, 34	0, 0.15, 0.45, 1.5, 4.5, 15	Fuel Fill Door
	1033 to 1158	12, 34	0, 0.15, 0.45, 1.5, 4.5, 15	Fuel Fill Door
	5594 to 6468	12, 34	0, 0.15, 0.45, 1.5, 4.5, 15	Fuel Fill Door
1	-30 to 14	12, 34, 55	0, 0.15, 0.45, 1.5, 4.5, 15	Under Hood
	-44 to 59	12, 34, 55	0, 4.5	Top of Gas Tank
	-31 to 6	12, 34, 55	0, 4.5	Fuel Fill Door
2	1183 to 5604	12, 34, 55	0, 0.15, 0.45, 1.5, 4.5, 15	Under Hood
	1468 to 4765	12, 34, 55	0, 4.5	Top of Gas Tank
	1974 to 4684	12, 34, 55	0, 4.5	Fuel Fill Door
3	9387 to 12039	12, 34, 55	0, 0.15, 0.45, 1.5, 4.5, 15	Under Hood
	8924 to 13356	12, 34, 55	0, 4.5	Top of Gas Tank
	8509 to 12617	12, 34, 55	0, 4.5	Fuel Fill Door
6	14906 to 19373	12	0, 0.15, 0.45, 1.5, 4.5, 15	Under Hood
7	-137 to 219	12	0, 0.15, 0.45, 1.5, 4.5, 15	Under Hood

^a The exhaust ranges are from measurements with no artificial running loss emissions.

not turned off until well after the vehicle passed the instrument. Table S6 shows the summary for the 38 test conditions and 1,040 RSD measurements using liquid gasoline releases. The flow of gasoline was left on for each set of five with-running-loss RSD measurements to minimize the possibility of vapor bubbles forming in the line to the release point. Table S7 details the 22 defeats of the evaporative emissions control systems on five of the test vehicles to attempt to induce vapor running loss emissions. These tests produced 44 test conditions and 682 RSD measurements.

Table S6. Summary of Liquid Gasoline Running Loss Tests Performed.

Test Vehicle	Measured Exhaust HC (ppmC ₃) Range ^a	Speed (mph)	Artificial Running Loss	
			Gasoline Emission Rate (ml/sec)	Release Location
2	1738 to 5404	12, 34	0, 0.1, 0.3, 1.0	Front Bumper
	921 to 3974	12, 34	0, 0.1, 0.3, 1.0	Rear Bumper
5	931 to 1299	12	0, 0.8	Passenger Door
6	17228 to 20130	12	0, 0.05, 0.1, 0.5, 1.0	Left Front Wheel
	16471 to 21072	12	0, 0.05, 0.1, 0.5, 1.0	Rear Bumper
7	-70 to 85	12	0, 0.05, 0.1, 0.5, 1.0	Front Bumper
	-82 to 61	12	0, 0.05, 0.1, 0.5, 1.0	Rear Bumper

^a The exhaust ranges are from measurements with no artificial running loss emissions.

Table S7. Summary of Induced Running Loss Tests Performed.

Test Vehicle	Measured Exhaust HC (ppmC ₃) Range ^a	Speed (mph)	Method of Disabling Evaporative Emission Control System
0	-100 to 31 -57 to 47	12 12	Canister Disconnected Gas Cap Removed
1	-52 to 11 -41 to 23	12, 34, 55 12, 34, 55	Canister Disconnected Gas Cap Removed
2	2610 to 4950 1817 to 4734	12, 34, 55 12, 34, 55	Canister Disconnected Gas Cap Removed
3	7687 to 11925 8593 to 11989	12, 34, 55 12, 34, 55	Canister Disconnected Gas Cap Removed
4	93 to 370 11 to 220	12 12	Canister Disconnected Gas Cap Removed

^a The exhaust ranges are from measurements collected with canister connected and the gas cap properly installed.

Data Trends. The amount of the scatter in the HC vs. CO₂ 10-ms plots induced by the addition of a running loss plume is affected by a combination of several different factors. The first is the running loss emissions rate. Figure S6 shows HC vs. CO₂ 10-ms data plots for 0, 0.15, 0.45, 1.5, 4.5, and 15 scfh propane releases at 12 mph from Vehicle 0 with the 1100 ppmC₃ simulated dry exhaust mixture. Deviations from linearity are clear for the highest three propane release rates, but as the running loss emissions rate drops below 1.5 scfh, the correlations return to being reasonably linear and are comparable to the scatter for the 0 scfh case (the black symbols).

The speed of the vehicle can also influence the RSD observed absorption signal of the running loss emissions. Depending on the vehicle and the local conditions as speed increases the running loss emission plumes generally experience higher dilution rates. This lowers the HC concentrations behind the vehicle and consequently lowers the observed absorption values. Additionally, higher speeds can lower turbulence behind the vehicle (laminar flow) and prevent the running loss plume from intersecting the sampling beam. Both of these issues work to lower the running loss signals in the RSD data stream.

The plots in Figure S7 demonstrate the effect speed can have on the RSD absorption measurements. For this figure, 4.5 scfh of propane was released from the top of the fuel tank of Vehicle 2, which had a measured exhaust concentration of about 3000 ppmC₃, and the vehicle was driven at 12, 34 and 55 mph. The plots show that the correlation is relatively constant for the three speeds, but the data scatter, which reflects the running loss emissions, decreases as speed increases. For reference, the plot also shows a black trace for a test when no propane was released from the test vehicle.

A third factor that can affect plume detectability of the running loss emission plume is the

Figure S6. HC versus CO₂ for Vehicle 0 at 12mph and 1100 ppmC₃ exhaust hydrocarbon for different running loss release rates from the fuel fill door.

Figure S7. HC versus CO₂ for Vehicle 2 with 3000 ppmC₃ exhaust hydrocarbon for exhaust only and a 4.5 scfh running loss release rate from the top of the fuel tank at 12, 34, and 55 mph.

location of the release point on the vehicle. Figure S8 shows Vehicle 2 operated at 12 mph as propane was released at 4.5 scfh under the hood, at the top of the gas tank, and at the fuel fill door. The plot shows similar and substantially larger scatter for the traces corresponding to running loss releases from the three test locations in comparison with the no propane release (in black) for the same speed.

Figure S8. HC versus CO₂ for Vehicle 2 with 3000 ppmC₃ exhaust hydrocarbon for exhaust only and a 4.5 scfh running loss release rate from three release locations at 12 mph.

The development of the RSD running loss index (RLI) was aimed at quantifying the HC versus CO₂ point scatter induced by the running loss plume. Because running losses are not emitted from the tailpipe, the running loss plume may waft across the light beam many times or one time (or never) during the 0.5s RSD data acquisition period. In addition, the plume may waft across the light beam early or late in the data acquisition period. Because of these characteristics, an index based on all of the residuals, such as the standard error or the average residual of the regression, would not give equal weight to measurements that consisted of a single pass versus multiple passes of the running loss plume through the light beam. The data collected in the CDPHE study indicated that when running loss plumes are convincingly picked up by the RSD instrument, the plume usually elevates at least three or four of the HC absorbance values. A single elevated 10-ms HC value is rarely observed, and if one does appear to be observed, it is difficult to distinguish it as a true running loss indicator versus a single outlier point.

Consequently, designation of a vehicle as an elevated running loss emitter using an index based on the largest residual would be uncertain and unreasonably noisy. The choice of the ninetieth percentile residual was a compromise between using all and one regression residual.

Figure S9 shows the results for Vehicle 1 (0 ppmC₃ tailpipe) and how the RLI values change with propane release rate (scfh) and vehicle speed. The RLI values were measured at 12, 34, and 55 mph and with under-hood propane release rates of 0, 0.15, 0.45, 1.5, 4.5, and 15 scfh. Using a log scale, each plot shows the cumulative distribution of 20 replicate RLI values measured at each test condition. The eighteen distributions shown in the figure all have similar widths, although there may be a tendency for wider relative widths for larger RLI values. RLI distribution widths are influenced by the variability in the measured optical absorption values and the location of the running loss plumes with respect to the RSD light beam location.

As a reference, the black curve on the left of each panel defines the RLI distribution when no propane is released (0 running loss emissions) and establishes the lower bound for the RLI values. The propane release rates (0.15, 0.45, 1.5, 4.5, 15 scfh) were deliberately chosen for testing to be evenly spaced by factors of 3. Figure S9 shows that when propane release rates are substantially above the zero-running-loss (black) distribution, the RLI distributions are close to being evenly spaced on the log scale. This implies that RLI values tend to be proportional to the propane release rate (scfh). A comparison of the location of the curves as speed increases from Figures S9a to S9b to S9c shows that all constant-release-rate curves move to lower RLI values until they begin “piling up” at the black zero-running-loss-emissions line. This behavior implies that RLI values for non-zero propane release rates are inversely related to vehicle speed.

Any of the RLI distribution plots can be used to judge if a given running loss rate is capable of being detected by a single RLI measurement. If an RLI value, when running loss emissions are present, and the mean RLI value when running loss emissions are not present are separated by more than three standard deviations (3σ) of the distribution of RLIs, then the running loss is detected. For the purposes of visually judging detection from the plots, the definition can be approximated for this evaluation: If the median of the RLI when running losses are present is larger than the largest RLI value of the 20 values in the RLI distribution when running losses are not present, then the running loss is detected. For example, the blue curve in Figure S9a indicates that for 12 mph and 4.5 scfh propane the RLI values ranged from about 250 to 1000 with a median of about 500. The repeatability (the relative distribution width) for RLIs at this condition is therefore about +200% and -50%. Thus, the difference in location of a colored curve and the black curve shows the effect on the RLI values of the propane being released.

Using the 3σ definition of detection, Figure S9a, for 12 mph, indicates that the upper three propane release rates (1.5, 4.5, and 15 scfh) are successfully detected by RLI. Note that in Figure S9a the median (y-axis = 0.5) of the green curve for 1.5 scfh is just larger than the largest value of the 0 scfh curve (black dots). However, the medians of the red (0.15 scfh) and orange (0.45 scfh) curves stay within the range of the 0 scfh RLI values. While the release rates lower than

Figure S9. Cumulative distributions of RLI values for replicate tests of Vehicle 1 (0 ppmC₃ tailpipe) driving at three speeds while releasing propane from under the hood at six flow rates. Top graph is 12mph, middle is 34mph, and bottom is 55mph.

1.5 scfh do not meet the 3σ definition for a positive identification they do suggest that the RLI values may be used as a screening test to increase the probability of selecting a vehicle that has elevated running loss emissions. At 34 mph, Figure S9b (middle graph) shows that 0.15 and 0.45 scfh propane releases are not distinguishable from 0 scfh. Again, the medians for the 1.5, 4.5, and 15 scfh curves are all above the largest 0 scfh RLI values indicating a positive detection.

Similarly, Figure S9c (bottom graph) shows that at 55 mph the RLI distributions for 0.15, 0.45, and 1.5 scfh are close to the same location as for 0 scfh, and therefore those running loss rates are not detected. At 55 mph only the propane released at 15 scfh (purple line) is unequivocally detected though the 4.5 scfh release (blue curve) is substantially shifted with respect to the 0 scfh curve indicating detection of that running loss emission. These plots show that as speed increases, the RLI distributions for all non-zero propane release rates shift toward the no release distribution.

For in-use measurements we need a method to establish the baseline zero running loss emission RLI distribution since we generally will not use an audit vehicle for this purpose. We know that the majority of vehicles in the on-road fleet have negligible tailpipe and running loss HC emissions. Thus, we will use the RLI measurements collected from the late model vehicles to approximate the RLI zero running loss characteristics of the RSD instrument to define this baseline distribution.

Regression Modeling. The plots such as those in Figure S9 indicated that the relationship between RLI, running loss emission rate (g/mile) and vehicle operating conditions might be able to be quantified using the 3,501-observation staged test dataset. Early, project-specific functions of RLI were used by RSDs in the driveways of inspection/maintenance facilities at Ken Caryl and Broomfield stations in the Denver, Colorado area to screen for vehicles with elevated hot-soak evaporative emissions.^{1,2} At the time, RLI was called EI23. For the analysis of this West Los Angeles data, we developed a new, more widely applicable function of RLI to better screen for running loss emissions.

The trends of RLI vs. exhaust HC, speed, and propane release rate indicate that the RLI values can be thought of as being made up of two parts: 1) a running loss emissions signal (RLI_{Signal}) influenced by propane release location on the vehicle, propane release rate, vehicle speed, and exhaust HC concentration, and 2) a variability and noise floor (RLI_{Noise}) influenced by RSD instrument noise and measurement variability, exhaust HC, and vehicle speed. Their effects on the measured RLI can be described using the square root of the sum of their squares:

$$RLI = \text{SQRT} (RLI_{Signal}^2 + RLI_{Noise}^2) \quad (S1)$$

Raw data plotting, like that above, and early models using categorical variables for the propane release rate and the nominal vehicle speed indicated that the running loss emissions signal was close to proportional to the propane release rate and inversely proportional to the vehicle speed. Exhaust HC was not expected to influence the running loss signal, but a term was included in the model statement in the event that an effect was present. The resulting regression model statement for the running loss signal is given by:

$$RLI_{Signal} = \exp[\text{Location} + a*\ln(\text{Propane}) + b*\ln(\text{Speed}) + c*\text{Exhaust_HC}] \quad (S2)$$

Where:

Location = a categorical variable for the vehicle and location of the propane release,
Propane = propane flow rate in g/hr (1 scfh C₃H₈ = 51.8 g/hr C₃H₈),
Speed = vehicle speed in miles/hr at the time of the RSD measurement,
Exhaust_HC = the RSD exhaust HC concentration in parts per thousand propane.

The noise floor signal is defined to be the RLI value when the vehicle has zero running losses. Therefore, the noise floor does not depend on propane release rate. In addition, when exhaust HC is present, more noise is introduced into RLI arising from the detection of the exhaust HC. More specifically, RLI_{Noise} is influenced by the slope of the plot of HC absorption vs. CO₂ absorption. When exhaust HC concentrations are zero, the zero slope can be either positive or negative because zero measurements are normally distributed around zero. Examination of the data indicated that negative slopes affect RLI_{Noise} just as positive slopes do. Therefore, we quantify the influence of the slope on RLI_{Noise} using the absolute value (abs) of the RSD-reported exhaust HC concentration. The one exception is for negative slopes that have been invalidated by the RSD software. In these few cases the Exhaust_HC is set to zero. Speed was not expected to influence the noise floor signal, but a term was included in the model statement in case an effect was present. The resulting regression model statement for the noise floor signal is given by:

$$RLI_{Noise} = Instrument + d * abs(Exhaust_HC) + e * Speed \quad (S3)$$

Where:

Instrument = a categorical variable for the RSD instrument collecting the data,
Exhaust_HC = the RSD exhaust HC concentration in parts per thousand propane,
Speed = vehicle speed in miles/hr at the time of the RSD measurement.

Examination of the replicate measured RLI values at each test condition in the dataset revealed that the variance of the RLI values was inhomogeneous across test conditions. We therefore chose to make the variance of the modeled response variable more homogeneous while retaining the functional form of Equation S1 by transforming both sides of the equation using a power and then using non-linear regression (SAS NLIN procedure) to estimate the coefficients in Equations S2 and S3. The power of the transformation was adjusted until the optimal Box-Cox lambda (SAS TRANSREG procedure) for the observed transformed RLI values against the predicted transformed RLI values, was found to be 1. The optimum power for the transformation of RLI was found to be -0.281. The resulting regression coefficients for Equations S2 and S3 are given in Table S8. In the transformed space, the model had an r-square of 0.702, an F-value of 9357, and the model's residuals for the transformed RLI values (RLI ^ -0.281) had a standard deviation of 0.0344.

In the top part of Table S8 for the RLI_{Signal} component of RLI, examination of the 95% confidence intervals of the Location coefficients indicates that nine of the twelve coefficients are not significantly different from each other. The other three Location coefficients differ from the first nine but not substantially so. We conclude, while there are probably differences in the

Table S8. Regression coefficients for first regression model.

	Variable	Coefficient	Estimate of Coefficient	Approximate Standard Error	Approximate 95% Confidence Limits	
					Lower	Upper
In(RLI _{Signal})	Leak Location	Veh. 0 Fuel Fill Door	3.4505	0.1823	3.0930	3.8080
		Veh. 1 Fuel Fill Door	3.2877	0.1993	2.8968	3.6785
		Veh. 1 Top of Gas Tank	4.2171	0.2029	3.8192	4.6149
		Veh. 1 Under Hood	3.9754	0.1819	3.6187	4.3321
		Veh. 2 Fuel Fill Door	3.7328	0.2091	3.3229	4.1427
		Veh. 2 Top of Gas Tank	3.8500	0.2083	3.4416	4.2584
		Veh. 2 Under Hood	3.8737	0.1942	3.4929	4.2545
		Veh. 3 Fuel Fill Door	4.2763	0.2471	3.7917	4.7608
		Veh. 3 Top of Gas Tank	3.9499	0.2795	3.4019	4.4979
		Veh. 3 Under Hood	3.9583	0.2390	3.4897	4.4270
		Veh. 6 Under Hood	3.8220	0.3272	3.1805	4.4636
		Veh. 7 Under Hood	2.9728	0.1995	2.5818	3.3639
	ln(Propane_gph)	a	0.9727	0.0313	0.9114	1.0340
	ln(Speed_mph)	b	-1.1210	0.0464	-1.2121	-1.0300
	ExhaustHC_ppthC ₃	c	-0.0205	0.0128	-0.0456	0.0047
RLI _{Noise}	Instrument	RSD Unit 4627	92.6701	2.2596	88.2399	97.1003
		RSD Unit 4628	50.6895	2.5090	45.7702	55.6089
		RSD Unit 4630	43.2663	2.4780	38.4078	48.1247
	abs(ExhaustHC_ppthC ₃)	d	13.1632	0.3541	12.4690	13.8574
	Speed_mph	e	0.1227	0.0646	-0.0039	0.2493

response of RLI to running loss emissions release locations and vehicle body shapes, all of the vehicles and locations tested displayed some major response.

We also note that coefficients a, b, and c have values not significantly different from or quite near to 1, -1, and 0, respectively. These coefficients lead us to conclude that RLI_{Signal} has no significant dependence on Exhaust_HC concentration. Since the modeled quantity was the log of RLI_{Signal}, a is the coefficient for ln(Propane_gph), and b is the coefficient for ln(Speed_mph), the regression indicates that RLI_{Signal} is close to directly proportional to the ratio of grams per hour divided by miles per hour, which is grams per mile.

The model for the RLI_{Noise} component in the bottom part of Table S8 indicates that the Instrument coefficient dominates this RLI component. Three different commercial RSD instruments were used to collect the staged data. While the statistical significance of the d coefficient for the absolute value of Exhaust_HC concentration is strong, the effect on RLI_{Noise} is relatively small unless the exhaust HC concentration is quite large.

Overall, the regression indicates that RLI_{Signal} is approximately proportional to release rate (g/hr) and inversely proportional to speed (miles/hr), but when those two predictors are combined

(g/hr / miles/hr = g/mile), it becomes apparent that RLI_{Signal} is proportional to emission rate (g/mile) and is independent of speed. Speed has no significant effect on RLI_{Noise} .

Based on the Table S8 regression results of the first regression model, we performed a second regression on the same data but used some simplifying assumptions. We forced a, b, c, and e to be exactly 1, -1, 0, and 0, respectively, and assumed that there were no differences in the response of RLI_{Signal} to Location. The coefficients for the new regression are given in Table S9.

In the transformed space ($RLI^{-0.281}$), the Table S9 model, which was built on 3,501 data points, had an r-square of 0.687, an F-value of 35724, the residuals for the transformed RLI values had a standard deviation of 0.0353, and the distribution of the residuals was not significantly different from a normal distribution. The parity plot of measured vs. predicted values in the transformed space in Figure S10 shows that regression residuals are near homogeneous.

Table S9. Regression coefficients for second regression model.

	Variable	Coefficient	Estimate of Coefficient	Approximate Standard Error	Approximate 95% Confidence Limits	
					Lower	Upper
$\ln(RLI_{Signal})$	Leak Location	Any vehicle, any release location	3.1275	0.0231	3.0822	3.1728
	$\ln(\text{Propane_gph})$	a (forced to 1)	1			
	$\ln(\text{Speed_mph})$	b (forced to -1)	-1			
	ExhaustHC_ppthC ₃	c (forced to 0)	0			
RLI_{Noise}	Instrument	RSD Unit 4627	92.8842	2.1476	88.6734	97.0949
		RSD Unit 4628	56.1785	1.1100	54.0023	58.3548
		RSD Unit 4630	46.8141	1.2154	44.4310	49.1971
	$\text{abs}(\text{ExhaustHC_ppthC}_3)$	d	13.1897	0.3576	12.4887	13.8908
	Speed_mph	e (forced to 0)	0			

Using the regression coefficients from Table S9 and substituting, Equation S2 in steps can be reduced to:

$$\ln(RLI_{Signal}) = 3.1275 + 1 * \ln(\text{Propane}) + (-1) * \ln(\text{Speed}) + 0 * \text{Exhaust_HC} \quad (S4)$$

taking the exponential of both sides:

$$RLI_{Signal} = \exp(3.1275) * (\text{Propane})^1 * (\text{Speed})^{-1} \quad (S5)$$

$$RLI_{Signal} = 22.8 * (\text{Propane}) / (\text{Speed}) \quad (S6)$$

Figure S10. Parity plot of the 3,501 measurements as modeled by the second regression summarized by the coefficients in Table S9.

Propane is in g/hour and speed is in miles/hr which condenses to g/mile:

$$RLI_{\text{Signal}} = 22.8 * \text{Propane (g/mile)} \quad (\text{S7})$$

Similarly, using the regression coefficients from Table S9 and substituting into Equation S3 results in the following:

$$RLI_{\text{Noise}} = \text{Instrument} + 13.1897 * \text{abs(Exhaust_HC)} + 0 * \text{Speed} \quad (\text{S8})$$

$$RLI_{\text{Noise}} = \text{Instrument} + 13.2 * \text{abs(Exhaust_HC)} \quad (\text{S9})$$

Overall, Equations S1, S7 and S9 quantify the influences of running loss emission rate (g/mile), exhaust HC concentration, and RSD instrument noise on RLI. However, these relationships were developed using commercial RSD instrument data collected during 3,501 staged running loss emissions tests. Therefore, it is important to ask about the general applicability of these equations and specifically their usefulness for describing the running loss emission measurements collected with the FEAT instrument in West Los Angeles. The commercial RSD instruments used in the equations' development are based on the FEAT instrument and are therefore technically similar and have similar operating and measurement protocols. In addition, the $RLI^{-0.281}$ power transformation produced normally distributed residuals for the regression analysis that produced

Equations S1, S7 and S9. Figure S11 shows the distribution of $RLI^{0.281}$ power transformed values for the 2015 FEAT West Los Angeles dataset. The distribution is close to normally distributed and has a standard deviation of 0.0442, which is only 25% higher than the regression standard deviation. The similarities of the FEAT running loss measurements and the commercial instruments staged running loss measurements for vehicles, that are expected to generally have low running loss emissions and exhaust HC emissions, supports the use of Equations S1, S7 and S9 with the FEAT measurements after the proper instrument coefficient is calculated.

Figure S11. Distribution of 1999 - 2015 model year RLI values for the FEAT 2015 West Los Angeles dataset.

As mentioned earlier, the Equation S9 instrument coefficient is equal to the RLI for vehicles that have running loss emissions that are near 0 g/mile and have exhaust HC concentrations that are near 0 ppm. The newest model year light-duty gasoline vehicles will be dominated by vehicles with these characteristics. Thus, the instrument coefficient can be estimated for the FEAT instrument for each dataset by determining the average RLI for late model year vehicles. Accordingly, Table S10 shows the average RLI by model year for the 2013 and 2015 West Los Angeles datasets. The average RLI for the 2013 measurements is 29.6 and 20.4 for the 2015 dataset. The increase in the instrument coefficient value for the 2013 measurements mirrors the reported differences in the measurement noise for the FEAT instruments HC channel with the 2013 HC measurements experiencing 28% higher noise levels than the 2015 measurements.³ Also, the RLI noise stability is demonstrated by the day-by-day average RLIs of the 1999-2015

model year vehicles for the seven testing days from 3/28/2015 to 4/3/2015: 16.4, 18.4, 23.1, 18.1, 18.0, 18.9, and 17.0.

RLI Minimum Detection Limit. The Table S9 regression results can be used to estimate the FEAT running loss emission rate (g/mile) minimum detection limit (MDL) as defined by 40 CFR Part 136 Appendix B.⁴ This defines the MDL as the lowest emission rate at which there is a 99% chance that the emission rate is greater than zero. For a zero-emission vehicle the RLI value is determined from Equation S1 and is just the instrument coefficient previously determined. We calculate the 99% confidence interval from these values in the transformed space where residuals are near normal and are separated by 2.326 standard deviations (0.0353). For the 2013 and 2015 measurement campaigns, the FEAT instrument had average zero running loss RLIs of 29.6 and 20.4 as shown in Table S10. To get up to the corresponding MDL 99% probability RLIs of 69 and 44, using Equation S1 the RLI_{Signal} would need to increase by 63 and 38, respectively. According to Equation S7, these increases would be provided by running loss emissions of 2.8 and 1.6 g/mile, respectively.

Table S10. RLI Statistics for FEAT 2015 and 2013 West Los Angeles Data.

Model Year	2015 Counts	2015 Mean RLI	2013 Counts	2013 Mean RLI
2015	929	19.7		
2014	2208	20.0		
2013	2035	20.1	1401	29.1
2012	1523	19.7	2268	29.3
2011	1126	20.1	1717	29.9
2010	1059	19.6	1417	28.9
2009	932	20.0	1187	29.6
2008	1153	19.7	1684	29.2
2007	1395	20.3	1901	29.3
2006	1276	20.4	1858	29.8
2005	1242	21.0	1804	29.8
2004	1035	21.2	1592	29.7
2003	1047	21.7	1542	30.0
2002	937	20.7	1369	30.4
2001	812	20.7	1301	30.1
2000	734	21.9	1129	29.7
1999	602	21.7	1059	30.1
Total / Average	20,045	20.4	23,229	29.6

Accordingly, the running loss emission rate MDL for the FEAT 2013 measurement campaign is approximately 2.8 g/mile and the MDL for FEAT 2015 measurements is 1.6 g/mile. These MDLs are independent of speed but are a weak function of exhaust HC concentration. The MDL's speed independence might seem contrary to the speed trends seen in Figure S9. The reason is that the contours in Figure S9 are for release rate (g/hr). For a constant emission rate

(g/mile), release rates are proportionally higher at higher speeds. For example, for an emission rate at the 1.6 g/mile MDL, the release rate at 12 mph would be 19 g/hr, and at 55 mph the release rate would be 88 g/hr. The MDLs are a weak function of exhaust HC concentration. For example, for the FEAT instrument's 2015 West Los Angeles dataset when the exhaust HC increases from 0 to 1000 ppmC₃, the MDL increases from 1.6 to 3.3 g/mile.

RLI Probabilistic Method. The above analysis and discussion show that, in the absence of noise and RLI variability, RLI is directly proportional to running loss emission rate (g/mile). However, because of the functional form of Equation S1 and because most vehicles in the fleet have small values of RLI_{Signal} compared to values of RLI_{Noise}, it is not possible to explicitly solve Equations S1, S7 and S9 for the running loss emission rate Propane (g/mile) given measured RLI values.

We have not found a reliable way to correct the dependences of RLI on tailpipe HC concentration, but these effects need to be accounted for so that the assignment of vehicles using the RLI values is not systematically biased. This dependence arises due to the simple fact that for large HC/CO₂ values the 90th percentile, even for a well correlated relationship, can produce large RLI values. Instead of correcting the RLI values, the values of RLI are binned using bin boundaries that are dependent on the observed tailpipe HC emissions. In this way, the positive bias introduced by increasing levels of tailpipe HC emissions can be compensated for. Bins are calculated for each RLI measurement, taking into account the RSD measured exhaust HC concentration, and the RLI value is placed in its appropriate bin. As the bin number increases so does the expected certainty of the vehicle having running loss emissions.

Relative rankings are based on the number of standard deviations (RLI Bin) that the transformed measured RLI ($RLI_{meas}^{-0.281}$) is above the transformed expected RLI ($RLI_{clean}^{-0.281}$) for a reference vehicle with a zero running loss emission rate but with an exhaust HC concentration at the concentration measured by the RSD. We use the RSD-measured value of exhaust HC concentration since it is the best measure of exhaust HC concentration available. We also use the staged standard deviation value of 0.0353 since it has been determined using the 3,501 staged tests over a wide range of test conditions. Finally, we round the number of standard deviations to integers for convenience. The relationship is given by Equation S10:

$$RLI\ Bin = \text{round}[(RLI_{meas}^{-0.281} - RLI_{clean}^{-0.281}) / (-0.0353)] \quad (S10)$$

Where RLI_{clean} = the RLI value determined by Equations S1, S7 and S9 using:

Propane = 0 g/mile,
Exhaust_HC (ppthC₃) as determined for the vehicle by the RSD measurement, and
Instrument coefficient determined by the average measured RLI of late model vehicles.

As an internal check, the probabilistic method described above was tested against the staged data that was used to develop Equations S1, S7 and S9. We cannot do this check with the FEAT

measurement data because we do not have any propane release rate data to compare against. However, we can select conditions from the FEAT measurement campaign as the test conditions. Figure S12 shows the distribution of speeds and exhaust HC concentrations measured by the FEAT instrument in the 2015 West Los Angeles dataset. These ranges were used to select the staged test conditions (Exhaust HC ≤ 3000 ppmC₃, Speed = 12 and 34 mph) to display the relationship between RLI Bin and propane release rate (g/mile) measurements in the staged tests. Figure S13 shows that the calculated RLI Bins from the commercial RSD instrument increase with increasing propane release rate. The horizontal reference lines demarcate the integer RLI Bins. The plot clearly shows substantial scatter in the decimal and integer RLI Bin values. This is a consequence of noise and variability (RLI_{Noise} component) in the detection of running loss emissions by the instrument. Nevertheless, Figure S13, in addition to Equations S1, S7 and S9, provides a clear indication of the ability to sense running loss emissions.

Figure S12. Distribution of Speed and Exhaust HC for the FEAT 2015 West Los Angeles dataset. The FEAT speed system does not report speeds below 6 mph.

Figure S13. Relationship between RLI Bin and propane release rate for staged tests selected using 2015 FEAT measurement campaign conditions.

Literature Cited

1. DeFries, T.H., Palacios, C.F., Weatherby, M.F., Stanard, A.P., Kishan, S., *Estimated Summer Hot-Soak Distributions for Denver's Ken Caryl I/M Station Fleet*, Final Report, Version 8, EPA-420-R-14-008, March 2014, prepared by Eastern Research Group, February 8, 2012, revised December 20, 2013.
2. DeFries, T.H., Palacios, C.F., Weatherby, M.F., Kishan, S., *Estimated Summer Hot-Soak Distributions for the Denver Fleet*, prepared by Eastern Research Group, submitted to Colorado Department of Public Health and Environment, November 19, 2012.
3. Bishop, G. A.; Haugen, M. J., *Measuring Real-World Emissions from the On-road Passenger Fleet*; California Air Resources Board: Sacramento, CA, 2016;
http://www.feabiochemdu.edu/assets/databases/Cal/Univ_Denver_ARB_12_303_Final_report_2016.pdf.
4. Definition and Procedure for the Determination of the Method Detection Limit - Revision 2. In *Code of Federal Regulations*; Title 40, Part 136, Appendix B, 2017.

Figure S14. A) Percent CO₂ (—, left axis) and %HC (□, right axis) emissions versus time for a 1998 pickup (RLI 246, Bin 5) measured in 2015 demonstrating an anti-correlated pattern. B) %CO₂ (—, left axis) and %HC (□, right axis) emissions versus time for a 2005 van (RLI 92, Bin 3) measured in 2015 demonstrating a decreasing HC pattern. Percentages are calculated assuming that all of the exhaust has been compressed into an 8cm cell and there is no excess air involved in combustion.

Figure S15. A) Rear image of a CNG (see tank and cylinder cap along left side of image) powered garbage truck (RLI 103, Bin 4). The image includes counter-clockwise from the lower left, tailpipe %CO, %HC, %NO, %CO₂, time and date. B) %CO₂ (solid line, left axis) and %HC (dashed line, right axis) versus time for this truck's measurement.

Figure S16. The top panel shows a 1997 Honda (A) with excessively high CO and HC tailpipe emissions (two-stroke snowmobile levels) and the well correlated CO₂ and HC time series (B, %CO₂ (solid line, left axis) and %HC (dashed line, right axis) versus time). This vehicle had a reported RLI of 232 but when corrected for tailpipe HC it was placed in bin 0. The bottom panel shows a 2013 Nissan (C) emitting only CO₂ (D, %CO₂ (solid line, left axis) and %HC (dashed line, right axis) versus time). This vehicle had a reported RLI of 24 and was placed in bin 1. The images includes counter-clockwise from the lower left, tailpipe %CO, %HC, %NO, %CO₂, time and date.

Figure S17. Example measurement data for a vehicle excluded from consideration of non-tailpipe HC emissions. The top panel is the time series of the IR reference voltage. The middle panel is the time series of the calculated %CO₂ (left axis) and %HC (right axis) values for the half second measurement. The bottom left graph is %HC versus %CO₂ for the quality controlled data points and the bottom right graph is a similar plot using all of the points. The lines are the least squares best-fit lines.

Figure S18. Tag image of the vehicle measured and graphed in Figure S17 (RLI 43, Bin 2).

The reference voltage is reduced at the beginning and in the middle of the sampling time series (see Figure S17). From the license plate image, it is speculated that the first partial blockage is caused by the vehicle mud flaps and the second partial blockage may be due to the long rope tied to the back of the cargo door. The quality controlled data results plotted in the bottom left graph of Figure S17 results in a reasonable HC/CO₂ slope determination. However, the physical blockages still increase the scatter in a number of points that led to this vehicle's RLI of 43 and Bin 2 designation and subsequent elimination from consideration during the visual inspection.

Table S11. Listing of 2013 Vehicles Visually Confirmed.

Date	Time	Year	Make / Type	RLI Value	RLI Bin (Shape)	gCO/kg of Fuel	gHC/kg of Fuel
4/27/13	11:18:35	2010	HOND / 4D	52	2 (F)	4.65	
4/29/13	9:19:27			54	2 (F)		
5/4/13	14:41:38	2006	MITA / UT	59	2 (R)	-19.96	
4/27/13	10:51:38	1987	MAZD / PK	60	2 (R)	151.73	-1.44
5/3/13	14:09:56	2001	FORD / SN	61	2 (R)	-0.40	-6.44
5/1/13	16:05:31	2004	TOYT / 2L	61	2 (F)	4.21	16.65
5/4/13	15:24:12	2005	VOLV / UT	81	2 (E)	3.27	18.44
5/3/13	9:07:49	1968	FORD / 2D	103	2 (A)	34.10	64.23
4/28/13	18:14:13	2002	PORS / CV	136	2 (E)	-0.25	63.10
5/3/13	17:43:00	2003	MERC / 4D	147	2 (E)	39.55	
5/2/13	16:06:23	1995	FORD / PK	148	2 (F)	204.73	
4/29/13	10:27:38			163	2 (F)		
4/30/13	13:20:25	2001	JAGU / 4D	598	2 (E)	1.45	357.44
4/27/13	12:19:11	1996	LNDR / SW	82	3 (A)	225.82	1.70
5/1/13	17:09:54	2000	ACUR / 4D	93	3 (R)	0.67	1.51
5/4/13	9:45:39			97	3 (R)		
5/2/13	12:57:58	2001	GMC / VN	99	3 (F)	36.09	12.46
5/2/13	17:19:19	1995	FORD / VN	100	3 (E)	5.54	
4/28/13	14:35:41	1990	CHEV / CP	102	3 (A)	22.42	8.58
4/30/13	8:19:59	2007	FORD / SW	110	3 (F)	38.02	11.95
5/2/13	16:08:24			110	3 (A)	14.08	-0.54
5/3/13	9:33:31	2002	TOYT / UT	113	3 (F)	2.48	21.68
4/29/13	14:10:09	1999	GMC / LL	114	3 (A)	24.39	-8.30
5/3/13	10:26:22	2011	VOLK / 4D	118	3 (R)	6.50	7.59
4/29/13	7:36:43			118	3 (F)		
5/4/13	8:33:36	1994	INFI / 4D	149	3 (F)	14.99	31.15
5/1/13	14:25:57	1978	FORD / PK	186	3 (A)	64.43	-37.83
5/1/13	9:45:09			134	4 (H)	66.43	
5/1/13	11:41:41	1989	FORD / PK	170	4 (R)	733.66	-17.57
4/28/13	18:06:20	1998	FORD / CP	260	4 (F)	-1.87	
5/3/13	10:26:15	1991	FORD / PK	5899	4 (R)	250.78	

Leak shape definitions:

A – Anti-correlation between the HC and CO₂ emissions

D – Consistently decreasing HC signal that does not look exponential

E – Exponential decay of HC emissions

H – Consistently high HC emissions

R – Rising HC emissions or Late Burst

Table S12. Listing of 2015 Vehicles Visually Confirmed.

Date	Time	Year	Make / Type	RLI Value	RLI Bin (Shape)	gCO/kg of Fuel	gHC/kg of Fuel
3/28/15	10:50:21			50	2 (R)	5.05	5.05
4/1/15	13:26:44		CHEV / VN	64	2 (R)	25.96	21.52
3/28/15	15:28:50	1999	GMC / SV	80	2 (D)	87.91	39.55
3/31/15	8:22:55	2008	TOYT / 2D	112	2 (H)	-25.90	66.58
4/1/15	6:55:13	1974	VOLK / 2D	952	2 (E)	784.51	
4/1/15	12:29:30	2013	TOYT / 4D	51	3 (E)	26.02	
4/3/15	7:52:52	1998	DODG / PK	54	3 (A)	22.30	0.88
4/3/15	12:52:37		MD Truck	72	3 (D)	0.00	0.00
3/31/15	9:57:42	1997	CHEV / UT	73	3 (D)	89.65	13.39
4/2/15	10:50:19		Garbage Truck	75	3 (A)	0.00	0.00
4/3/15	9:14:54	2005	DODG / SV	92	3 (D)	10.01	-18.36
4/3/15	14:45:49	1997	FORD / VN	128	3 (D)	54.50	61.26
4/3/15	12:51:35	2000	CHEV / PK	135	3 (R)	88.85	74.14
3/31/15	9:54:35		Tar PK	392	3 (R)	-1.34	
4/3/15	12:00:43		CHEV / 2D	1111	3 (D)	-1.08	
3/30/15	15:47:33		MD Truck	78	4 (R)		
3/28/15	13:40:34	1997	DODG / SV	86	4 (D)	3.89	-6.70
4/3/15	8:58:15	2015	AUDI / UT	87	4 (R)	-2.96	-8.53
3/28/15	11:57:01	2002	MAZD / 4H	87	4 (D)	-3.56	11.49
4/3/15	14:42:10	2015	HYUN / 4D	97	4 (R)	2.26	1.92
4/1/15	10:12:06		MD Truck	98	4 (A)	24.82	8.51
3/30/15	14:01:42		Garbage Truck	103	4 (H)	9.04	19.18
3/28/15	13:06:53			113	4 (R)	1.58	10.78
3/31/15	10:52:09	1979	DODG / VN	146	4 (D)	479.68	35.68
4/2/15	8:04:50	2000	MERZ / UT	152	4 (A)	3.13	24.08
4/2/15	16:12:38	1992	HOND / CP	163	4 (A)	681.63	41.10
3/29/15	13:11:13	1936*	FORD / CP	213	4 (R)	0.00	0.00
4/2/15	8:20:10	2003	NISS / 4D	224	4 (E)	67.18	53.52
3/30/15	11:46:35	2004	HOND / 4D	528	4 (E)	-1.44	111.22
3/28/15	12:48:47	1997	HYUN / SW	645	4 (E)	50.35	
3/28/15	12:25:59	1998	DODG / PK	1619	4 (R)	5.11	
4/1/15	13:26:34	1967	CHRY	5731	4 (E)	396.08	
4/3/15	10:49:10	1992	TOYT / 4D	119	5 (A)	99.13	-3.60
3/30/15	14:02:54	1999	CHEV / SV	145	5 (H)	11.51	6.69
4/3/15	12:00:51	2014	MERZ / 4D	169	5 (H)	-2.25	6.79
3/28/15	12:47:23	2004	ACUR / 2H	186	5 (E)	20.77	12.02
4/2/15	14:18:34	1993	TOYT / CB	204	5 (R)	145.90	26.66
4/1/15	9:01:37	1998	DODG / PK	246	5 (H)	10.19	34.80
4/3/15	12:12:15	2003	CHEV / VN	257	5 (D)	13.15	23.00

Highlighted rows indicate same vehicle.

Table S12 Continued. Listing of 2015 Vehicles Visually Confirmed.

Date	Time	Year	Make / Type	RLI Value	RLI Bin (Shape)	gCO/kg of Fuel	gHC/kg of Fuel
3/30/15	11:39:34	1997	FORD/ UT	293	5 (E)	2.84	39.11
4/3/15	14:46:52	1999	CHEV / 3C	400	5 (E)	8.75	52.51
3/28/15	16:43:27	1986	CHEV/ CP	174	6 (H)	28.15	
4/1/15	14:46:53	1972	MERC /CP	198	6 (D)	143.34	
4/3/15	12:28:24	2005	CHRY /SV	273	6 (E)	3.22	-7.08
3/31/15	13:59:51		PK	292	6 (R)		
3/30/15	11:56:30		NISS / PK	400	6 (R)	664.28	27.92
4/3/15	12:38:25	1983	FORD / VN	339	7 (A)	7.15	
4/1/15	13:41:26		PK	345	7 (D)		
3/30/15	9:19:08		Car	541	7 (R)		

*This vehicle was visually identified but not plate matched and is not counted with the nine plate matched vehicles discussed in the text.