Title

On-Board Sensor-Based NO_x Emissions from Heavy-Duty Diesel Vehicles

Journal

Environmental Science & Technology, 53(9)

ISSN

0013-936X 1520-5851

Authors

Tan, Yi
Henderick, Paul
Yoon, Seungju
et al.

Publication Date

2019-04-17

DOI

10.1021/acs.est.8b07048

Peer reviewed

Environmental Science & Technology

Article

Cite This: *Environ. Sci. Technol.* 2019, 53, 5504−5511

pubs.acs.org/est

On-Board Sensor-Based NO_x Emissions from Heavy-Duty Diesel Vehicles

Yi Tan,*[,†] Paul Henderick,[‡] Seungju Yoon,[†] Jorn Herner,[†] Thomas Montes,[‡] Kanok Boriboonsomsin,[§] Kent Johnson,[§] George Scora,[§] Daniel Sandez,[§] and Thomas D. Durbin[§]

[†]California Air Resources Board, 1001 I Street, Sacramento, California 95814, United States

[‡]California Air Resources Board, 9500 Telstar Avenue, Ste. #2, El Monte, California 91731, United States

[§]College of Engineering - Center for Environmental Research and Technology, University of California at Riverside, 1084 Columbia Avenue, Riverside, California 92507, United States

Supporting Information

ABSTRACT: Real-world nitrogen oxides (NO_x) emissions were estimated using on-board sensor readings from 72 heavy-duty diesel vehicles (HDDVs) equipped with a Selective Catalytic Reduction (SCR) system in California. The results showed that there were large differences between in-use and certification NO_x emissions, with 12 HDDVs emitting more than three times the standard during hot-running and idling operations in the real world. The overall NO_x conversion efficiencies of the SCR system on many vehicles were well below the 90% threshold that is expected for an efficient SCR system, even when the SCR system was above the optimum operating temperature threshold of 250 °C. This could potentially be associated with SCR catalyst deterioration on some engines. The Not-to-Exceed (NTE) requirements currently used by the heavy-duty in-use compliance program were evaluated using on-board NO_x sensor data. Valid NTE events covered only 4.2−16.4% of the engine operation and 6.6−34.6% of the estimated NO_x emissions. This work shows that low cost on-board NO_x sensors are a convenient tool to monitor in-use NO_x emissions in real-time, evaluate the SCR system performance, and identify vehicle operating modes with high NO_x emissions. This information can inform certification and compliance programs to ensure low in-use NO_x emissions.

INTRODUCTION

Nitrogen oxides (NO_x) play an important role in the formation of atmospheric ozone and fine particulate matter ($PM_{2.5}$). Controlling NO_x emissions is thought to be critical for attaining the National Ambient Air Quality Standards (NAAQS) for these two pollutants in the next decade in California, especially in the South Coast and San Joaquin Valley air basins.[1] The estimated contribution of on-road heavy-duty diesel vehicles (HDDVs) to the total NO_x emission inventory in California was ∼33% in 2014, which is considerably higher than the US average (∼15%).[2,3] Recent studies have shown that on-road diesel emissions could be underestimated by emission inventory models.[4,5] In order to reduce NO_x emissions from these vehicles, 2010 and newer model year (MY) heavy-duty diesel engines must comply with a NO_x emissions standard of 0.20 g/bhp-hr or a Family Emission Limit (FEL) not exceeding 0.50 g/bhp-hr over engine-dynamometer test cycles, including the transient Federal Test Procedure (FTP) and the Supplemental Emissions Test (SET). These vehicles should also meet the not-to-exceed (NTE) emission standard to ensure that NO_x emissions are controlled under conditions thought to be experienced in-use.

To meet these NO_x standards, heavy-duty diesel engine manufacturers commonly employ Selective Catalytic Reduction (SCR) together with Exhaust Gas Recirculation (EGR) and other in-cylinder NO_x control strategies.[6] The EGR system

Received: December 13, 2018
Revised: March 28, 2019
Accepted: April 17, 2019
Published: April 17, 2019

DOI: 10.1021/acs.est.8b07048
Environ. Sci. Technol. 2019, 53, 5504−5511

cools and recirculates a portion of the exhaust back to the engine, which lowers the oxygen concentration, flame temperatures, and thus engine-out NO_x emissions. On the other hand, SCR uses diesel exhaust fluid (DEF) in conjunction with a catalyst to control NO_x emissions. DEF is an aqueous urea solution that when thermalized supplies gaseous ammonia for the SCR reaction. Platinum, vanadium, and zeolite are materials presently used for SCR catalysts.[7] Copper (Cu)- and iron (Fe)-based zeolite catalysts are widely used in heavy-duty applications because of their high NO_x conversion efficiency over a wide range of operating temperatures (200 °C−450 °C for Cu-zeolite and >250 °C for Fe-zeolite), relatively low cost, and improved thermal durability.[8] The application of platinum-based catalysts is very limited because of their narrow operating temperature range (<250 °C).[7] Although vanadium-based catalysts also have a wide operating temperature range (260 °C−450 °C), their instability against hydrothermal aging and environmental safety concerns have limited their applications.[7,8] To achieve high NO_x conversion efficiency, the SCR system requires relatively high exhaust temperatures and precise control of the DEF injection rate. The optimal DEF injection rate depends on the NO_x concentrations measurements at the inlet and outlet of the SCR system.

Laboratory and on-road measurements, however, show that in-use NO_x emissions from on-road HDDVs are often higher than the certification standard.[9−14] For example, Thiruvengadam et al. (2015) tested three diesel particulate filter (DPF)- and SCR-equipped HDDVs that were certified to the 2010 emission standard.[10] NO_x emissions over the near-dock and local drayage driving cycles were 5 to 7 times higher than the standard. Drayage cycles represent frequent stop-and-go operations, resulting in low SCR temperatures throughout much of the cycle. Misra et al. (2017) observed that in-use NO_x emissions of two HDDVs equipped with 2010 MY engines ranged from 0.50 to 1.24 g/bhp-hr, despite the exhaust temperature being high enough for proper SCR function.[13] Yoon et al. (2017) found that three HDDVs meeting the 2010 standard emitted 0.34−1.80 g/bhp-hr of NO_x over different chassis dynamometer cycles and 0.17−0.97 g/bhp-hr of NO_x over highway test routes in the real world.[14] These results show that in-use NO_x emissions exceeding emission standards may be a common problem among HDDVs. Based on the US Environmental Protection Agency's Motor Vehicle Emission Simulator (MOVES) model, Anenberg et al. (2017) estimated that excess in-use NO_x emissions (i.e., in excess of certification limits) from on-road HDDVs were associated with about 1,000 premature deaths in the United States in 2015.[15] In addition, many disadvantaged communities are located in close proximity to busy roadways and could suffer from higher than average NO_x exposures.[16,17]

Laboratory and on-road emission measurements using regulation-compliant instrumentation are labor- and cost-intensive, which limits the number of vehicles that can be tested. Plume capture devices and other roadside instruments can measure a large number of vehicles in a relatively short time period.[18−22] However, a single plume capture setup does not cover emissions under different operating conditions. Publicly broadcast data from the Engine Control Unit (ECU), including NO_x concentrations from on-board NO_x sensors, can be used to estimate instantaneous NO_x emissions over a wide range of real-world operations.[23,24] Previous research has shown that on-board NO_x sensors can be effectively used to monitor NO_x concentration in diesel exhaust gas.[25,26] Using on-board NO_x sensor data from two diesel transit buses, Kotz et al. (2016) showed that these buses emitted NO_x at rates 3 to 9 times higher than the standard, primarily due to low load and low-speed operations. NO_x hotspots were identified at bus stops, during cold starts, on inclines, and during accelerations.[23]

In this work, ECU data collected from 72 HDDVs operating in various vocations were used to estimate in-use NO_x emissions (Table S1).[24] In-use NO_x emissions and SCR performance of these vehicles were evaluated under different operating conditions. Since NO_x sensors were active during engine operations that were subject to the NTE requirements, an NTE evaluation was also conducted on NO_x from 15 vehicles to assess the effectiveness of current NTE emission standards in monitoring in-use emissions. The results were used to explore the need of a better regulatory framework to meet emission reduction goals.

■ METHODS

Data Collection. The College of Engineering - Center for Environmental Research and Technology (CE-CERT) of the University of California at Riverside conducted a large-scale data collection program in which real-world vehicle and engine activity data were collected from 90 heavy-duty vehicles in California.[24] The Society of Automotive Engineers (SAE) has developed the J1939 standard that assigns Suspect Parameter Number (SPN) code terms for specific ECU parameters. The data were collected using a J1939 Mini Logger (HEM Data Corporation). This instrument recorded more than 170 SPNs from the ECU and GPS locations at 1 Hz. The data collection effort spanned from November 2014 to September 2016. Data were collected from each vehicle for 1−3 months, resulting in a total of more than 29,682 h of data. For HDDVs, engine-out and tailpipe NO_x concentrations were read from on-board NO_x sensors located at the SCR inlet and outlet, respectively. Tailpipe NO_x data were available from 72 HDDVs, but engine-out NO_x concentrations were not recorded from four of these vehicles. All the vehicles were of 2010 or newer MY and equipped with DPF and SCR equipment. These vehicles were classified into 16 vocational groups, including line-haul, drayage, construction, local distribution, refuse, public, and utility repair vehicles (Table S1).

NO_x Emission Estimation. NO_x concentrations from the on-board NO_x sensors at the SCR inlet and outlet (SPN 3216 and 3226, respectively), engine intake air mass flow rate (SPN 132), and engine fuel rate (SPN 183) were used to calculate the instantaneous NO_x emission rate (g/s). To be consistent with Code of Federal Regulations, Title 40, Chapter I, Section 1065.655 (40 CFR 1065.655), the molar mass of NO_2 was used as the effective molar mass of all NO_x species while calculating the NO_x mass emissions. The instantaneous exhaust flow rate was estimated from the intake air mass flow rate and engine fuel rate, and the NO_x mass flow rate was calculated following 40 CFR 89.418. Engine power and brake horsepower were calculated using engine speed (SPN 190), actual percentage torque (SPN 513), nominal friction torque (SPN 514), and reference torque (SPN 544). Brake-specific NO_x emissions were calculated by dividing the NO_x mass emission rate in grams per hour by the brake horsepower. The Infrequent Regeneration Adjustment Factors were not applied to the brake-specific emissions, as the NO_x emissions during

DOI: 10.1021/acs.est.8b07048
Environ. Sci. Technol. 2019, 53, 5504−5511

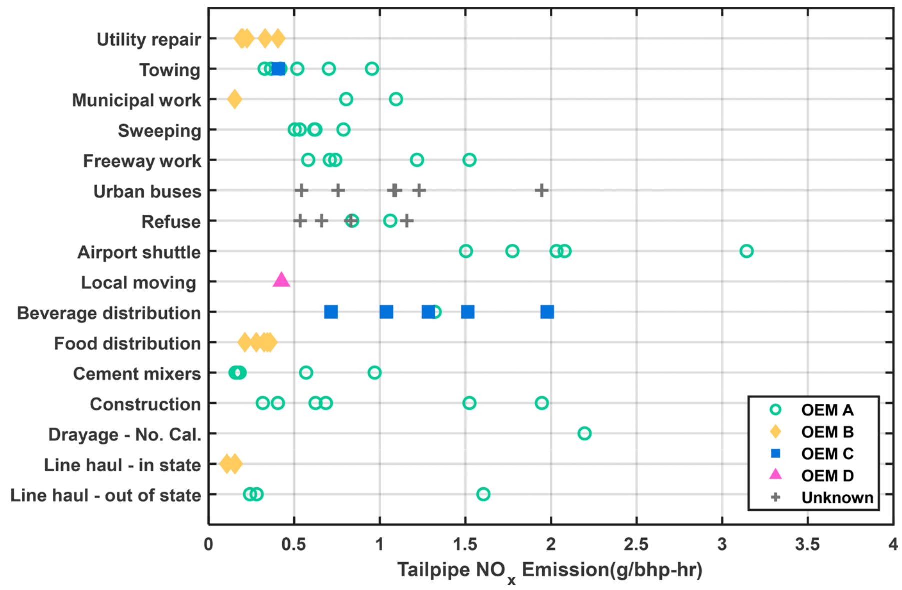

Figure 1. Brake-specific tailpipe NO_x emissions of 72 HDDVs. Markers distinguish Original Equipment Manufacturers (OEM).

DPF regeneration events were measured by the on-board NO_x sensors.

When the exhaust gas temperature is lower than about 150 °C, on-board NO_x sensors remain inactive to avoid malfunction caused by water condensation. Data when NO_x sensors were turned off or warming up were excluded to ensure erroneous data were not incorporated into the results. On average, the SCR outlet NO_x sensors were active for more than 70% of the engine operation time. Following 40 CFR 1065.650, negative NO_x emission rates and power values were set to zero. Setting negative values to zero for emission averages could slightly bias the results high, typically by less than 1% or 0.02 g/bhp-hr (Table S1). NO_x conversion efficiencies were calculated for the 68 vehicles with engine-out NO_x data, using the total mass of engine-out and tailpipe NO_x. Due to the exclusion of data when NO_x sensors were deactivated and the SCR was not efficient, the overall NO_x conversion efficiencies calculated in this study represent upper-bound estimations, and real-world NO_x conversion will be lower.

The accuracy of on-board sensor based NO_x emission estimates was evaluated by comparing them with results from an AVL M.O.V.E. GAS Portable Emission Measurement System (PEMS). Seven HDDVs were tested on major freeways in Southern California with the PEMS and the J1939 Mini Logger instrumented simultaneously. Brake-specific NO_x emissions from the on-board sensor-based estimates showed good correlation with the results from PEMS measurements, with an average difference of 13% and a Pearson correlation coefficient of 0.98 (Figure S1).

In-Use NO_x Emissions Normalized to Certification. For purposes of emissions certification, heavy-duty diesel engines are grouped into engine families. Engines within the same engine family must have similar emission control systems and calibrations and are expected to have similar emission characteristics. For the 48 HDDVs with engine family information, the corresponding NO_x emission standard or FEL was obtained. Engine information for the remaining 24 HDDVs was either incomplete (i.e., engine MY was identified but engine family was missing) or not available.

For some early model year engines certified to the 2010 NO_x standard (FEL engines), manufacturers submitted FELs higher than the 0.20 g/bhp-hr standard, using banked emission credits from the averaging, banking, and trading program. The 40 CFR 86.007 required that the NO_x FELs may not exceed 0.50 g/bhp-hr. As these credits were exhausted, most later model year engines were certified to the 0.20 g/bhp-hr standard (STD engines). As a result, FEL engines were older, on average, than STD engines. This work analyzed 17 FEL engines, comprising 15 2010−2012 MY and two 2013+ MY engines, and 31 STD engines comprising seven 2010−2012 MY and 24 2013+ MY engines. This work used the "normalized emission" as a consistent metric to compare in-use emissions to certification requirements. The normalized emission for a given HDDV was defined as its average in-use NO_x emission value divided by the corresponding standard or FEL (i.e., in-use NO_x/FEL for FEL engines and in-use NO_x/0.20 for STD engines).

NTE Evaluation. The NTE standard was designed to evaluate the in-use emissions of heavy-duty engines. An NTE event occurs when the engine continuously operates within a control area (the "NTE zone") for at least 30 s. As described in 40 CFR 86.1370, the engine operates in the NTE zone when the engine speed is above a threshold determined by the power curve, where the engine load is higher than 30% of the maximum torque and the engine power is greater than 30% of the maximum power produced by the engine. All the engines in this study were equipped with EGR, thus the NTE events were also subject to the cold temperature operating exclusion. Cold temperature operation is defined as when the intake manifold temperature (IMT) or engine coolant temperature (ECT) broadcast by ECU is less than or equal to a calculated temperature defined by the relationship with absolute intake manifold pressure (IMP): IMT = (IMP + 7.75)/0.0875 and ECT = (IMP + 9.8889)/0.0778. An NTE event is excluded from the NTE evaluation when the IMT or the ECT broadcast by ECU is below the temperature defined by these equations or when the exhaust temperature within 30 cm downstream of the SCR system is lower than 250 °C. The SCR outlet NO_x sensor was always active during NTE events so data from the

DOI: 10.1021/acs.est.8b07048
Environ. Sci. Technol. 2019, 53, 5504−5511

sensor could be used to evaluate in-use NO_x emissions during these events.

An NTE event passes the NTE standard when the brake-specific NO_x is less than 1.5 times the FTP emission limit plus a 0.15 g/bhp-hr field measurement accuracy margin. The vehicle passes the in-use compliance requirements if the time-weighted pass ratio, defined as the sum of the duration time of all passing events divided by the sum of the duration time of all valid events, is above 90%. In this study, NTE evaluations were conducted on 15 vehicles for which the IMP was properly recorded.

RESULTS AND DISCUSSION

In-Use NO_x Emissions. The 2010 NO_x standard of 0.20 g/bhp-hr is more than 90% lower compared to the most recent pre-2010 standard of 2.4 g/bhp-hr. However, due to large differences between in-use and certification NO_x emissions, emission reductions from the pre-2010 NO_x standard were not as great as might be expected based on a direct comparison of the emissions standards.[5,9−14,18−22] In-use NO_x emissions of the 72 HDDVs ranged from 0.11 to 3.14 g/bhp-hr (Figure 1), and 46 HDDVs had NO_x emissions higher than the maximum allowable FEL of 0.50 g/bhp-hr. Among the 48 vehicles with detailed engine information, the normalized NO_x emissions ranged from 0.54 to 9.62 (Figure S2). The normalized emissions exceeded 1.0 for 38 vehicles and exceeded 3.0 for 12 of them. Note that high normalized emissions do not imply that these vehicles failed to meet in-use compliance requirements, as in-use compliance is determined based on emissions measured by PEMS during NTE events. Figure 1 also showed that in-use NO_x emissions varied substantially among different vehicle vocations and engine makes. Line-haul trucks represent the largest fraction of Vehicle Miles Traveled (VMT) in California (67% of the HDDV VMT statewide).[1] Although the NO_x emission of an out-of-state line-haul truck was 1.56 g/bhp-hr, the other five in-state and out-of-state line-haul trucks had NO_x emissions of below 0.30 g/bhp-hr. Twenty-two vocational vehicles had NO_x emissions higher than 1.00 g/bhp-hr, indicating that excessive in-use NO_x emissions could be prevalent in the vocational HDDV fleet. For example, airport shuttles in this study had an average NO_x emission of 2.10 ± 0.62 g/bhp-hr and emitted 328,739 g NO_x over 57,518 miles. If all the airport shuttles met the 0.20 g/bhp-hr standard in the real-world, their total estimated NO_x emission could be reduced by 298,341 g. Based on EMFAC2017, vocational vehicles contributed a significant amount of VMT and NO_x in densely populated urban areas (e.g., 61% and 63% of the HDDV VMT and NO_x in the south coast air basin, respectively).[1] Therefore, better emission control or adopting zero and near-zero emission technologies for vocational vehicles is required to achieve significant NO_x reductions in urban areas considering their high in-use NO_x emissions observed in this study.

To further characterize in-use NO_x emissions, instantaneous data were populated into a matrix of 15 bins based on vehicle speed and engine brake output power (Table 1). Bins 1−5 represent low load operations that are common in urban areas, bins 6−10 represent medium load operations, and bins 11−15 represent high load conditions. Vehicle speed increases with bin number under each load condition, ranging from idling to speeds higher than 40 mph. Low load idling (bin 1) was the most common vehicle operation mode. As for unweighted fleet averages, idling accounted for 34% of the time when the SCR outlet NO_x sensor was active and contributed 14% of the total estimated NO_x (Figure 2a). Other low-load operations (bins 2−5) comprised 43% of the operating time and contributed 36% of the total estimated NO_x. Furthermore, if the data when the SCR outlet NO_x sensor was inactive were included, 81% of

Table 1. Characterizing NO_x Emissions with 15 Bins Based on Vehicle Speed and the Percentage of the Maximum Power Produced by the Engine (Rated Power)

	vehicle speed (mph)				
% of rated power	idle	1−10	10−25	25−40	40+
<25	Bin 1	Bin 2	Bin 3	Bin 4	Bin 5
25−50	Bin 6	Bin 7	Bin 8	Bin 9	Bin 10
50+	Bin 11	Bin 12	Bin 13	Bin 14	Bin 15

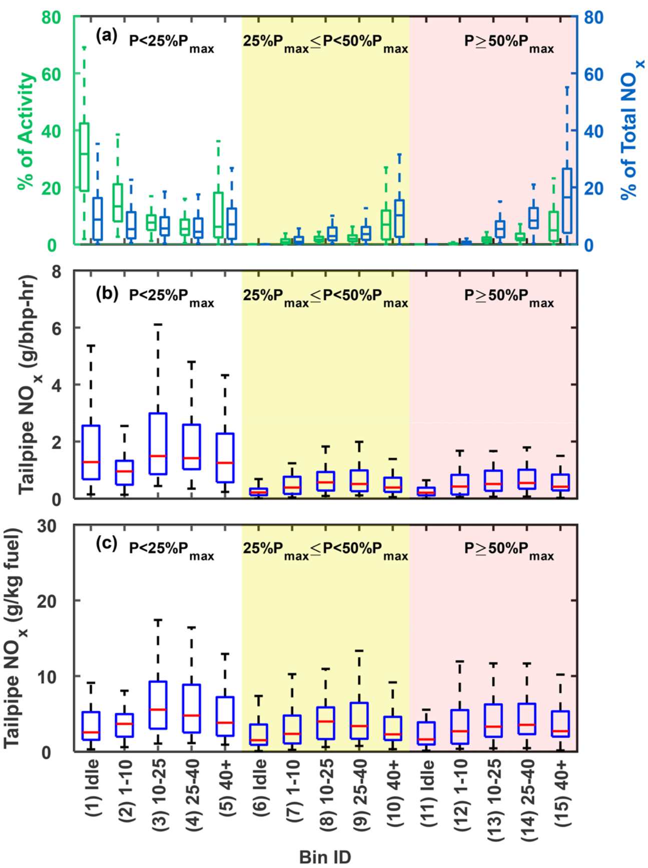

Figure 2. (a) Percentages of activity when the SCR outlet NO_x sensor was active and NO_x emissions of 72 HDDVs in different engine load and vehicle speed bins; (b) brake-specific NO_x emissions (g/bhp-hr) in different engine load and vehicle speed bins; and (c) fuel-specific NO_x emissions (g/kg fuel) in different engine load and vehicle speed bins. The *x*-axis shows the bin number (in parentheses) and the vehicle speed range (mph) of each bin. The white zone represents low engine-load conditions, the yellow zone represents medium engine-load conditions, and the red zone represents high engine-load conditions. In panel (a), green box whiskers show the statistical distribution of the percentage of vehicle activity in each bin, and the blue box whiskers show the statistical distribution of the percentage of total tailpipe NO_x emission in each bin. On each box, the central line indicates the median value, and the bottom and top edges of the box indicate the 25th and 75th percentiles, respectively. The whiskers extend to the extreme data points, excluding outliers. P indicates the brake output power, and P_{max} indicates the maximum power produced by the engine.

DOI: 10.1021/acs.est.8b07048
Environ. Sci. Technol. 2019, 53, 5504−5511

the total engine operating time was in low load operations (bins 1−5) (Figure S3), so NO_x emitted during low load operations could be more significant than the current estimation as well. As current certification cycles do not account for sustained low load operations, it is not necessary for manufacturers to optimize engine and aftertreatment calibrations for these operations. However, to meet future emission reduction goals, it will be important to better control NO_x emitted during low load operations with advanced engine and aftertreatment control strategies, such as implementing cylinder deactivation or using a mini-burner to keep the aftertreatment components at effective operating temperatures.[27,28]

Brake-specific NO_x emissions were the highest and showed the largest variations under low load conditions (Figure 2b). Meanwhile, instantaneous and distance-specific NO_x emission rates were the lowest under low load conditions and showed comparable or even smaller interquartile ranges compared to medium and high load conditions (Figure S4). Unlike time and distance that are accurately measured, brake output power is calculated from engine speed and torque values broadcast by ECU. Friction torque is usually very close to actual torque at low load, so the brake output torque that is the difference between the two is even smaller and can have very large uncertainties. Therefore, the high brake-specific emissions at low load may partly be explained by the low brake horsepower relative to the NO_x emissions.

Since brake-specific emissions are very sensitive to the accuracy of broadcast torque at low load, alternative metrics such as fuel-specific emissions may be more suitable to evaluate emissions at low load. The engine fuel rate broadcast by the ECU is typically estimated from models using various sensor data such as the throttle valve position and can be more accurate than the calculated brake output power at low load. Fuel-specific emissions for low load bins were 21−67% higher than those for medium and high load bins with the same speed (Figure 2c). Preble et al. (2015) observed 5.1 ± 1.2 g NO_x/kg fuel for SCR equipped HDDVs in a plume capture study conducted at the Port of Oakland in California, where trucks were observed to be accelerating from a traffic light ∼50 m before the sampling point or cruising at a speed of ∼30 mph.[20] The results were consistent with fuel-specific emissions of Bin 9 (4.5 ± 3.5 g NO_x/kg fuel) and Bin 14 (4.4 ± 3.1 g NO_x/kg fuel) in this study. In another plume capture study by Haugen and Bishop (2018), the fleet average emissions observed for chassis model years 2011 and newer were 20.1 ± 0.9 g NO_x/kg fuel at the Port of Los Angeles and 10.6 ± 1.2 g NO_x/kg fuel at Cottonwood in California.[22] The vehicles were traveling at 8−12 mph, while decelerating at the Port of Los Angeles site and slightly accelerating at the Cottonwood site. The average exhaust temperature was ∼86 °C at the Port of LA and ∼108 °C at Cottonwood. The fuel-specific emission factors of Haugen and Bishop (2018) were more than two times higher than the relevant bins in this study likely due to the much colder exhaust temperatures at which NO_x sensors would be inactive.

SCR System Performance. Insufficient SCR NO_x conversion was the main reason for the high in-use NO_x emissions observed in this study. As shown in Figure 3, the NO_x conversion efficiencies of 21 vehicles were lower than 70%, resulting in NO_x emissions of 1.60 ± 0.52 g/bhp-hr. These vehicles emitted significantly more NO_x than the 19 vehicles with higher than 90% NO_x conversion efficiencies, whose NO_x emissions were only 0.25 ± 0.09 g/bhp-hr. Based on an analysis of variance, SCR performance varied significantly depending on vocation (p = 0.006) and engine make (p = 0.014) but not model year (p = 0.149). On average, the in-state line-haul trucks had the highest NO_x conversion efficiencies (96.1 ± 1.0%), and the airport shuttles had the lowest NO_x conversion efficiencies (60.4 ± 4.0%). All the engines from Manufacturer B had high NO_x conversion efficiencies, ranging from 89.8% to 97.3%. In comparison, the NO_x conversion efficiencies of engines from Manufacturer A ranged from 54.6 to 94.3%.

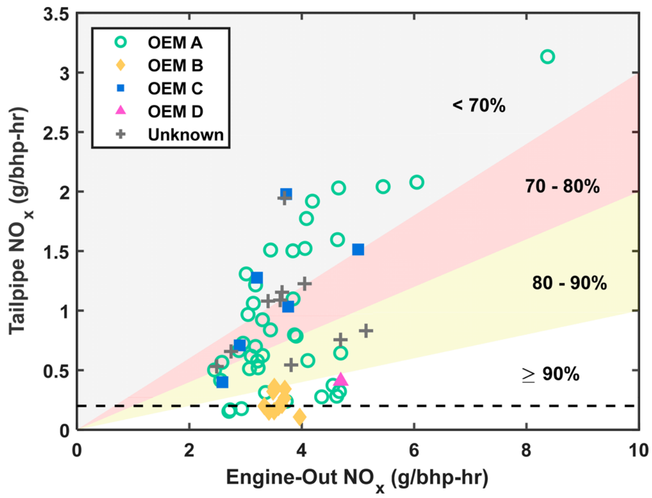

Figure 3. Engine-out and tailpipe NO_x emissions of 68 HDDVs. Points represent the average engine-out and tailpipe emissions of individual vehicles when both SCR inlet and outlet NO_x sensors were active. Different symbols represent vehicles with different engine makes, and shaded areas represent different NO_x conversion efficiency ranges. The dotted line represents the 0.20 g/bhp-hr NO_x standard.

After the introduction of SCR technology, diesel engines were able to reduce tailpipe NO_x emissions while simultaneously achieving fuel-consumption savings by allowing more fuel-efficient engine calibrations. Meanwhile, more fuel-efficient engine calibrations can result in higher combustion temperature and thus, higher engine-out NO_x emissions.[6] Figure 3 shows that engine-out NO_x emissions of all the vehicles in this study were higher than 2.4 g/bhp-hr. As a result, although the NO_x conversion efficiencies of 19 trucks were higher than 90%, only 7 of them achieved tailpipe NO_x emissions lower than 0.20 g/bhp-hr (all of them emitted less than 0.45 g/bhp-hr NO_x). Therefore, further optimization of aftertreatment system technologies and engine control strategies is needed to control NO_x emissions better without fuel consumption penalties.

Major design and operational factors that affect SCR performance include residence time, degree of mixing between the DEF and the exhaust, DEF dosing quantity, engine-out NO_x concentration, and catalyst reactivity. The catalyst reactivity strongly depends on its inlet temperature. Figure 4 shows average engine operation time and NO_x conversion efficiencies of the 68 HDDVs in three different SCR inlet temperature zones. The low SCR inlet temperature (<200 °C) zone had the lowest NO_x conversion efficiencies (59.0 ± 18.6%), accounting for 11.7 ± 9.5% of the engine operation time. The medium SCR inlet temperature (200−250 °C) zone had more engine operation time (37.5 ± 15.1%) and higher NO_x conversion efficiencies (73.7 ± 14.3%), and the high SCR

DOI: 10.1021/acs.est.8b07048
Environ. Sci. Technol. 2019, 53, 5504−5511

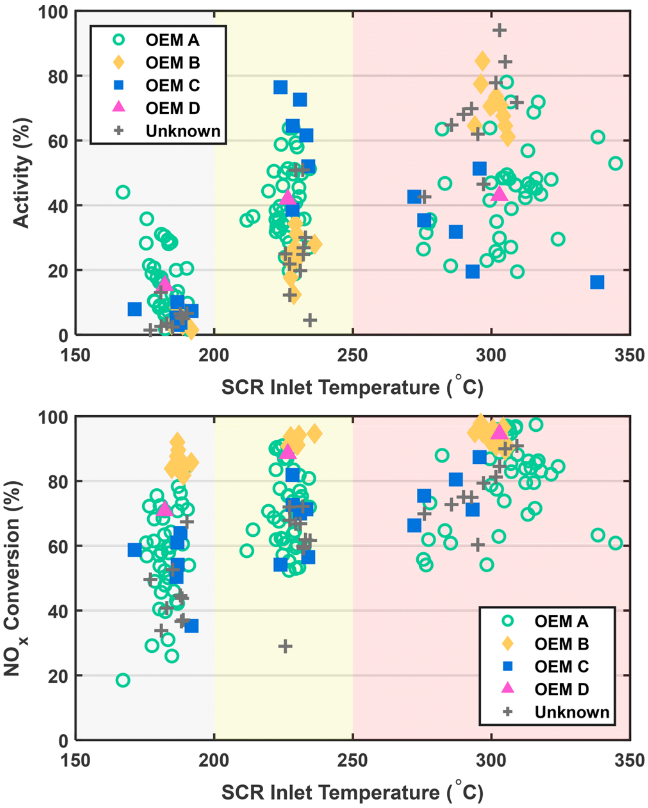

Figure 4. Vehicle activities and NO_x conversion efficiencies in three SCR inlet temperature zones. The gray area represents when the SCR inlet temperature was lower than 200 °C, the yellow area represents when the SCR inlet temperature was between 200 and 250 °C, and the red area represents when the SCR inlet temperature was above 250 °C. The percentage of vehicle activity (top panel) and the average NO_x conversion efficiency (bottom panel) of each vehicle are presented in each SCR inlet temperature zone, and the *x*-axis represents the average SCR inlet temperature when the vehicle was operating in the corresponding SCR inlet temperature zone. Different symbols represent vehicles with engines from different manufacturers.

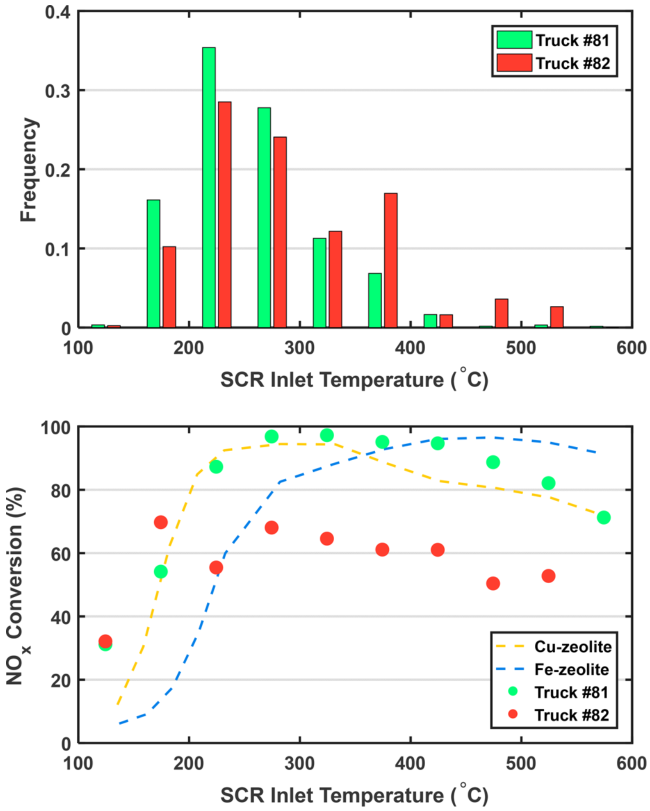

Figure 5. SCR inlet temperature profiles of two construction trucks and their average NO_x conversion efficiencies at different temperatures. In the upper panel, green and red bars represent the SCR inlet temperature profile of trucks #81 and #82, respectively. In the lower panel, dashed lines show the experimental NO_x conversion efficiencies of Cu-zeolite and Fe-zeolite catalysts from Cavataio et al. (2007),[8] and green and red dots show the estimated NO_x conversion efficiencies of trucks #81 and #82 at different temperatures, respectively.

inlet temperature zone (≥250 °C) had the majority of engine operation time (50.8 ± 18.4%) and the highest NO_x conversion efficiencies (81.1 ± 14.0%). However, because this analysis excluded data when NO_x sensors were turned off due to low exhaust temperature, operations in the low SCR inlet temperature zone with low NO_x conversion efficiencies could be more prevalent in the real world. Figure 4 also shows that engines from Manufacturer B consistently showed high SCR performance in different SCR inlet temperature zones. In particular, these engines were capable of maintaining NO_x conversion efficiencies above 80% when the SCR inlet was relatively cold (<200 °C). The fact that Manufacturer B engines had a wide range of mileage and vocational applications demonstrated that well calibrated and maintained SCR systems are capable of effectively reducing NO_x emissions in real-world operations.

Among the factors that could cause poor SCR performance, catalyst deterioration could be important in the real world and was associated with the recall of more than 500,000 Cummins engines.[29] Although further investigations are needed, catalyst deterioration was a potential explanation for observations in this sample set. For example, Figure 5 shows the SCR inlet temperature profiles and NO_x conversion efficiencies as functions of the SCR inlet temperature of two construction trucks (truck #81 and #82) from the same engine make, engine family, and model year. It was reasonable to assume that their SCR systems used the same catalyst. Despite similar engine-out NO_x emissions (4.58 g/bhp-hr for truck #81 and 4.53 g/bhp-hr for truck #82) and SCR inlet temperature profiles, tailpipe NO_x emissions of truck #81 and #82 were 0.40 and 1.52 g/bhp-hr, respectively. Previous laboratory testing showed that a working Cu-zeolite SCR system exceeded 80% NO_x conversion efficiency around 200 °C and exceeded 90% efficiency when the SCR inlet temperature was between 220 and 360 °C, while a working Fe-zeolite SCR system exceeded 80% efficiency around 280 °C and exceeded 90% efficiency when the SCR inlet was hotter than 350 °C.[8] When compared to the experimental NO_x conversion efficiencies as a function of the SCR inlet temperature, the in-use NO_x conversion efficiency of truck #81 closely followed the experimental curve of the Cu-zeolite catalyst. However, the NO_x conversion efficiency of truck #82 was below 70% even when the SCR inlet temperature was above 250 °C, significantly lower than the experimental efficiency curves of Cu-zeolite and Fe-zeolite catalysts. Therefore, if other malfunctions such as urea crystallization could be excluded, the SCR catalyst of truck #82 likely deteriorated faster than expected. The On-Board Diagnostics (OBD) system should detect the high NO_x emission problem and alert the operator to fix the SCR

DOI: 10.1021/acs.est.8b07048
Environ. Sci. Technol. 2019, 53, 5504−5511

system. However, the Malfunction Indicator Lamp (MIL) was not illuminated on truck #82.

NTE Evaluation. Among the 15 trucks with IMP data that had STD engines, 8 trucks failed the NTE evaluation because their time-weighted pass ratios were below 90%. However, in-use NO_x emissions of all 15 trucks were lower than 0.45 g/bhp-hr. Therefore, failing the NTE evaluation in this study might not necessarily indicate that the engine had very high in-use emissions. This is partly because operations meeting the NTE criteria only accounted for 6.6−34.6% of the estimated NO_x emission and 4.2−16.4% of the engine operation when NO_x sensors were active (Figure 6). Since high NO_x emissions can occur when NO_x sensors were inactive, such as during the engine-starting phase, the coverages of engine operation and NO_x emissions under the NTE requirements were even more limited for the full extent of the real-world conditions that include the periods when the NO_x sensors are inactive. Modifications to the current NTE requirements, such as removing the cold temperature operation exclusion, lowering the exhaust temperature requirement from 250 °C, or decreasing the event duration requirement of a consecutive 30 s, could improve the coverage of in-use NO_x emissions. For example, if the cold temperature operation exclusion was removed from the NTE criteria while keeping other requirements the same, the modified NTE criteria would cover 5.7−18.7% of the engine operation and 14.2−42.4% of the NO_x emission. Additional analyses, including exploring alternative paradigms such as the work or CO_2 based Moving Average Window method, are needed to identify the best method to monitor in-use emissions more effectively.

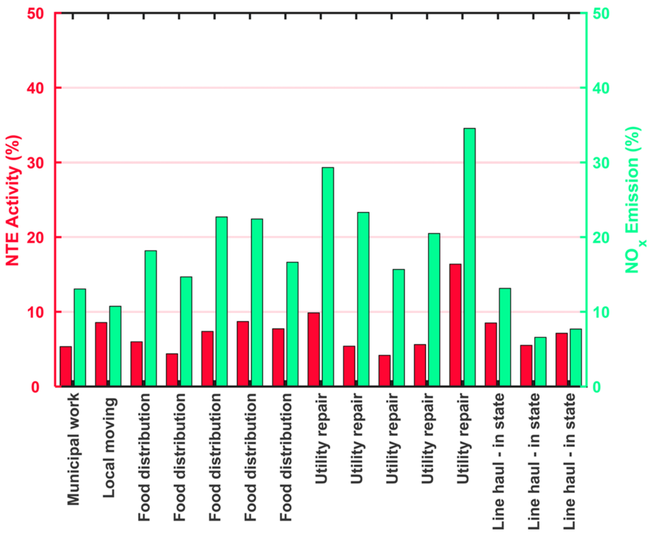

Figure 6. Vehicle activities and in-use NO_x emissions covered by the current NTE requirements. Red and green bars represent activities and NO_x emissions covered by the current NTE requirements, respectively. The x-axis shows the vocation of each truck.

Engine dynamometer cycles used in the engine certification process are known to be different from real-world operations, and current NTE testing procedures only monitor a small fraction of in-use emissions. In comparison, on-board NO_x sensors are inexpensive and a convenient tool to monitor in-use NO_x emissions, SCR functionality, and deterioration and to identify possible high emitters. With standardization regulations that ensure all manufacturers broadcast the same type of data, they will likely become an important part of possible future mobile source emissions control programs. For example, 2022 or newer MY HDDVs will be required to add software to store aggregated data from NO_x sensors.[30] This could be more efficient than laboratory or PEMS testing in providing comprehensive feedback about in-use NO_x emissions, ensure that the benefits of the emission standards programs are achieved in-use throughout the entire life of the vehicle, and help understand the difference between in-use emissions of HDDVs and their certification values.

However, NO_x sensor-based monitoring cannot yet replace the current in-use compliance testing that uses PEMS to measure emissions over the entire vehicle operation. For example, heavy-duty natural gas engines that are also subject to in-use compliance typically do not have on-board NO_x sensors. The broadcast NO_x sensor data are processed by engine manufacturers, and the data quality should be verified. In-use PEMS testing can provide independent evidence on emission problems and help identify intentional use of "defeat device". In addition, the sensitivity and accuracy of current NO_x sensors has not been examined at NO_x emission levels well below 0.20 g/bhp-hr. Further investigations are needed to determine if on-board sensors are adequate to measure ultralow NO_x concentrations and assess the in-use compliance of such engines. A major limitation of current on-board NO_x sensors is that they cannot capture cold-start and cold operation emissions, so in-use NO_x emissions could be substantially underestimated. Nevertheless, NO_x sensors with integrated heaters could avoid water condensation once the sensor has reached warm-up temperatures and the exhaust is hotter than 150 °C. It is expected that such sensors could remain active even under low load urban driving conditions and monitor NO_x emissions over the entire vehicle operation unless there is another technical reason that requires them to turn off.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b07048.

Table S1, ECU data collected from 72 HDDVs operating in various vocations used to estimate in-use NO_x emissions; Figure S1, correlation between PEMS and OBD based NO_x emission estimates from on-road testing; Figure S2, normalized NO_x emissions of 48 trucks with engine family information; Figure S3, percentages of activity of 72 HDVs in different engine load and vehicle speed bins; Figure S4, instantaneous and distance-specific NO_x emission rates under low load conditions and comparable or even smaller interquartile ranges compared to medium and high load conditions (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: yi.tan@arb.ca.gov.

ORCID

Yi Tan: 0000-0002-5130-6056

Notes

The authors declare no competing financial interest.

DOI: 10.1021/acs.est.8b07048
Environ. Sci. Technol. 2019, 53, 5504−5511

■ ACKNOWLEDGMENTS

We thank John Collins, Walter Ham, Cody Howard, and Shaohua Hu from CARB, Carl Fulper from EPA, and Matthew Spears from the Truck and Engine Manufacturers Association (EMA) for useful discussions. The data collection campaign was funded by CARB through contract 13-301.

■ REFERENCES

(1) California Air Resources Board, EMFAC2017 Web Database. https://www.arb.ca.gov/emfac/2017/ (accessed August 15, 2018).
(2) United States Environmental Protection Agency, 2014 National Emissions Inventory (NEI) Data. https://www.epa.gov/air-emissions-inventories/2014-national-emissions-inventory-nei-data (accessed August 15, 2018).
(3) California Air Resources Board, CEPAM: 2016 SIP - Standard Emission Tool. https://www.arb.ca.gov/app/emsinv/fcemssumcat/fcemssumcat2016.php (accessed August 15, 2018).
(4) Hassler, B.; McDonald, B. C.; Frost, G. J.; Borbon, A.; Carslaw, D. C.; Civerolo, K.; Granier, C.; Monks, P. S.; Monks, S.; Parrish, D. D.; Pollack, I. B.; Rosenlof, K. H.; Ryerson, T. B.; von Schneidemesser, E.; Trainer, M. Analysis of long-term observations of NOx and CO in megacities and application to constraining emissions inventories. *Geophys. Res. Lett.* **2016**, *43* (18), 9920−9930.
(5) Jiang, Z.; McDonald, B. C.; Worden, H.; Worden, J. R.; Miyazaki, K.; Qu, Z.; Henze, D. K.; Jones, D. B. A.; Arellano, A. F.; Fischer, E. V.; Zhu, L.; Boersma, K. F. Unexpected slowdown of US pollutant emission reduction in the past decade. *Proc. Natl. Acad. Sci. U. S. A.* **2018**, *115* (20), 5099−5104.
(6) Johnson, T.; Joshi, A. Review of Vehicle Engine Efficiency and Emissions, WCX 17: SAE World Congress Experience, SAE Technical Paper 2017-01-0907; 2017; DOI: 10.4271/2017-01-0907.
(7) Piumetti, M.; Bensaid, S.; Fino, D.; Russo, N. Catalysis in Diesel engine NOx aftertreatment: a review. *Catalysis, Structure & Reactivity* **2015**, *1* (4), 155−173.
(8) Cavataio, G.; Girard, J.; Patterson, J. E.; Montreuil, C.; Cheng, Y.; Lambert, C. K. Laboratory testing of urea-SCR formulations to meet tier 2 bin 5 emissions, 2007 World Congress, SAE Technical Paper 2007-01-1575, 2007; DOI: 10.4271/2007-01-1575.
(9) Misra, C.; Collins, J. F.; Herner, J. D.; Sax, T.; Krishnamurthy, M.; Sobieralski, W.; Burntizki, M.; Chernich, D. In-Use NOx Emissions from Model Year 2010 and 2011 Heavy-Duty Diesel Engines Equipped with Aftertreatment Devices. *Environ. Sci. Technol.* **2013**, *47* (14), 7892−7898.
(10) Thiruvengadam, A.; Besch, M. C.; Thiruvengadam, P.; Pradhan, S.; Carder, D.; Kappanna, H.; Gautam, M.; Oshinuga, A.; Hogo, H.; Miyasato, M. Emission Rates of Regulated Pollutants from Current Technology Heavy-Duty Diesel and Natural Gas Goods Movement Vehicles. *Environ. Sci. Technol.* **2015**, *49* (8), 5236−5244.
(11) Quiros, D. C.; Thiruvengadam, A.; Pradhan, S.; Besch, M.; Thiruvengadam, P.; Demirgok, B.; Carder, D.; Oshinuga, A.; Huai, T.; Hu, S. Real-World Emissions from Modern Heavy-Duty Diesel, Natural Gas, and Hybrid Diesel Trucks Operating Along Major California Freight Corridors. *Emission Control Science and Technology* **2016**, *2* (3), 156−172.
(12) Dixit, P.; Miller, J. W.; Cocker, D. R.; Oshinuga, A.; Jiang, Y.; Durbin, T. D.; Johnson, K. C. Differences between emissions measured in urban driving and certification testing of heavy-duty diesel engines. *Atmos. Environ.* **2017**, *166*, 276−285.
(13) Misra, C.; Ruehl, C.; Collins, J.; Chernich, D.; Herner, J. In-Use NOx Emissions from Diesel and Liquefied Natural Gas Refuse Trucks Equipped with SCR and TWC, Respectively. *Environ. Sci. Technol.* **2017**, *51* (12), 6981−6989.
(14) Yoon, S.; Collins, J. F.; Misra, C.; Herner, J. D.; Carter, M. W.; Sax, T. P. In-Use Emissions from 2010-Technology Heavy-Duty Trucks. Transportation Research Record. *Transp. Res. Rec.* **2017**, *2627*, 1−8.
(15) Anenberg, S. C.; Miller, J.; Minjares, R.; Du, L.; Henze, D. K.; Lacey, F.; Malley, C. S.; Emberson, L.; Franco, V.; Klimont, Z.; Heyes, C. Impacts and mitigation of excess diesel-related NOx emissions in 11 major vehicle markets. *Nature* **2017**, *545* (7655), 467−471.
(16) Rowangould, G. M. A census of the US near-roadway population: Public health and environmental justice considerations. Transportation Research Part D. *Transport and Environment* **2013**, *25*, 59−67.
(17) Pratt, G. C.; Vadali, M. L.; Kvale, D. L.; Ellickson, K. M. Traffic, air pollution, minority and socio-economic status: Addressing inequities in exposure and risk. *Int. J. Environ. Res. Public Health* **2015**, *12* (5), 5355−5372.
(18) Dallmann, T. R.; DeMartini, S. J.; Kirchstetter, T. W.; Herndon, S. C.; Onasch, T. B.; Wood, E. C.; Harley, R. A. On-Road Measurement of Gas and Particle Phase Pollutant Emission Factors for Individual Heavy-Duty Diesel Trucks. *Environ. Sci. Technol.* **2012**, *46* (15), 8511−8518.
(19) McDonald, B. C.; Dallmann, T. R.; Martin, E. W.; Harley, R. A. Long-term trends in nitrogen oxide emissions from motor vehicles at national, state, and air basin scales. *J. Geophys. Res.: Atmos.* **2012**, DOI: 10.1029/2012JD018304.
(20) Preble, C. V.; Dallmann, T. R.; Kreisberg, N. M.; Hering, S. V.; Harley, R. A.; Kirchstetter, T. W. Effects of Particle Filters and Selective Catalytic Reduction on Heavy-Duty Diesel Drayage Truck Emissions at the Port of Oakland. *Environ. Sci. Technol.* **2015**, *49* (14), 8864−8871.
(21) Haugen, M. J.; Bishop, G. A. Repeat Fuel Specific Emission Measurements on Two California Heavy-Duty Truck Fleets. *Environ. Sci. Technol.* **2017**, *51* (7), 4100−4107.
(22) Haugen, M. J.; Bishop, G. A. Long-Term Fuel-Specific NOx and Particle Emission Trends for In-Use Heavy-Duty Vehicles in California. *Environ. Sci. Technol.* **2018**, *52* (10), 6070−6076.
(23) Kotz, A. J.; Kittelson, D. B.; Northrop, W. F. Lagrangian Hotspots of In-Use NOX Emissions from Transit Buses. *Environ. Sci. Technol.* **2016**, *50* (11), 5750−5756.
(24) Boriboonsomsin, K.; Durbin, T.; Scora, G.; Johnson, K.; Sandez, D.; Vu, A.; Jiang, Y.; Burnette, A.; Yoon, S.; Collins, J.; Dai, Z.; Fulper, C.; Kishan, S.; Sabisch, M.; Jackson, D. Real-world exhaust temperature profiles of on-road heavy-duty diesel vehicles equipped with selective catalytic reduction. *Sci. Total Environ.* **2018**, *634*, 909−921.
(25) Hofmann, L.; Rusch, K.; Fischer, S.; Lemire, B. Onboard emissions monitoring on a HD truck with an SCR system using NOx sensors, SAE 2004 World Congress & Exhibition, SAE Technical Paper 2004-01-1290, 2004; DOI: 10.4271/2004-01-1290.
(26) Pohle, R.; Magori, E.; Tawil, A.; Davydovskaya, P.; Fleischer, M. Detection of NOx in Combustion Engine Exhaust Gas by Applying the Pulsed Polarization Technique on YSZ Based Sensors. *Proceedings* **2017**, *1* (4), 490.
(27) Gosala, D. B.; Allen, C. M.; Ramesh, A. K.; Shaver, G. M.; McCarthy, J.; Stretch, D.; Koeberlein, E.; Farrell, L. Cylinder deactivation during dynamic diesel engine operation. *Int. J. Engine Res.* **2017**, *18* (10), 991−1004.
(28) Sharp, C.; Webb, C. C.; Neely, G.; Sarlashkar, J. V.; Rengarajan, S. B.; Yoon, S.; Henry, C.; Zavala, B. Achieving Ultra Low NOX Emissions Levels with a 2017 Heavy-Duty On-Highway TC Diesel Engine and an Advanced Technology Emissions System - NOX Management Strategies. *SAE Int. J. Engines* **2017**, *10* (4), 1736−1748.
(29) EPA Announces Largest Voluntary Recall of Medium- and Heavy-Duty Trucks. https://www.epa.gov/newsreleases/epa-announces-largest-voluntary-recall-medium-and-heavy-duty-trucks (accessed November 15, 2018).
(30) A Vision for Minimizing Real-World Emissions in the On-Road Sectors. https://www.arb.ca.gov/board/books/2018/111518/18-9-3pres.pdf (accessed November 15, 2018).

DOI: 10.1021/acs.est.8b07048
Environ. Sci. Technol. 2019, 53, 5504−5511