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Effects of Particle Filters and Selective Catalytic Reduction on Heavy-Duty Diesel Drayage Truck Emissions at the Port of Oakland

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Supporting Information

ABSTRACT: Effects of fleet modernization and use of diesel particle filters (DPF) and selective catalytic reduction (SCR) on heavy-duty diesel truck emissions were studied at the Port of Oakland in California. Nitrogen oxides (NO_x), black carbon (BC), particle number (PN), and size distributions were measured in the exhaust plumes of ~1400 drayage trucks. Average NO_x, BC, and PN emission factors for newer engines (2010−2013 model years) equipped with both DPF and SCR were 69 ± 15%, 92 ± 32%, and 66 ± 35% lower, respectively, than 2004−2006 engines without these technologies. Intentional oxidation of NO to NO_2 for DPF regeneration increased tailpipe NO_2 emissions, especially from older (1994−2006) engines with retrofit DPFs. Increased deployment of advanced controls has further skewed emission factor distributions; a small number of trucks emit a disproportionately large fraction of total BC and NO_x. The fraction of DPF-equipped drayage trucks increased from 2 to 99% and the median engine age decreased from 11 to 6 years between 2009 and 2013. Over this period, fleet-average BC and NO_x emission factors decreased by 76 ± 22% and 53 ± 8%, respectively. Emission changes occurred rapidly compared to what would have been observed due to natural (i.e., unforced) turnover of the Port truck fleet. These results provide a preview of more widespread emission changes expected statewide and nationally in the coming years.

INTRODUCTION

Heavy-duty diesel trucks are a major source of nitrogen oxides (NO_x) and diesel particulate matter (PM) emissions in the United States.[1,2] These emissions contribute to fine particulate matter ($PM_{2.5}$) and ozone air quality problems,[3,4] and may lead to adverse health effects for exposed individuals.[3,5−7] Black carbon (BC) is a potent absorber of solar radiation and comprises the majority of diesel PM mass emissions.[8] Recent studies have suggested control of diesel BC emissions as a strategy to help mitigate global warming.[9,10]

PM and NO_x emission standards have been established nationally to limit emissions from heavy-duty diesel trucks. Current standards are typically met using exhaust after-treatment control technologies. Trucks with 2007 and newer engines are equipped with a diesel particle filter (DPF), and trucks with 2010 and newer engines also include selective catalytic reduction (SCR) systems for NO_x control. DPFs can be installed as retrofits on older engines that are already in use.[11]

Previous studies have shown that DPFs can reduce PM mass emissions from heavy-duty diesel engines by >90%.[12−14] Trapped carbon particles are oxidized to regenerate the filter either passively, by continuous reaction with nitrogen dioxide (NO_2) that is formed by catalytic oxidation of exhaust nitric oxide (NO), or actively, for example by periodic injection of fuel. The intentional conversion of NO to NO_2 in passively regenerated systems leads to increased primary NO_2 emissions and higher NO_2/NO_x emission ratios.[13,15−17] These emissions changes are of concern because NO_2 is toxic and increased primary NO_2 emissions promote ozone formation. Another concern is that DPF-related reductions in overall particle mass emissions may favor increased homogeneous nucleation rather than condensation of gases onto existing particle surfaces, thereby increasing formation of ultrafine particles (UFP) and total particle number (PN) emissions.[12,18,19] UFP (diameters <0.1 μm) can induce inflammatory and oxidant stress responses that have been linked to cardiovascular disease and mortality.[20−22] The increases in NO_2 and UFP emissions are both associated with high catalytic loading within the DPF and high exhaust temperatures.[13,18]

In California, the phase-in and use of DPFs has been greatly accelerated for drayage trucks, which are commonly used for short-haul freight transport at ports and rail yards. Over a three-year period between January 2010 and December 2012, all drayage trucks were required to be equipped with DPFs, either via retrofit or engine replacement.[23] Table S1 of the Supporting Information (SI) presents the schedule for required changes to

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the truck fleet in more detail. Emissions from California drayage trucks have been evaluated as this fleet modernization program has been implemented.[17,24−26] At the Ports of Los Angeles and Long Beach in Southern California, the drayage fleet was almost entirely replaced with new trucks, so the mean age of trucks decreased from 12.7 years to 2.5 years between 2008 and 2010.[17] Associated emissions reductions observed for carbon monoxide, NO_x, and exhaust opacity were 30, 48, and 54%, respectively.[17] In contrast to the Southern California ports where truck replacement predominated, there was significant retrofitting of DPFs on older drayage trucks at the Port of Oakland. Dallmann et al.[24] reported a 41% reduction in NO_x and a 54% reduction in BC emissions between 2009 and 2010 after pre-1994 trucks were banned and trucks with 1994−2003 engines were either retrofit or replaced with newer equipment. The BC reductions resulted primarily from increased use of DPFs, whereas NO_x reductions were attributed to fleet modernization, as the newer engines met more stringent NO_x emission standards. The initial round of changes to the drayage truck fleet at the Port of Oakland led to an increased proportion of trucks with 2004 and newer engines, as well as a reduction in mean engine age from 11.0 to 8.3 years.[24]

The current study builds on previous work at the Port of Oakland and features new field measurements including additional pollutants not previously measured, namely NO_2, PN, and particle size distributions. Also, emission factors for individual trucks in the current study were linked to engine attributes through transcribed license plate data. This linkage makes it possible to compare emissions across different control technology groups, in addition to quantifying changes in fleet-average emission factors over time.

MATERIALS AND METHODS

Truck emissions were measured in November 2011 and March 2013 near the Port of Oakland. Following Dallmann et al.,[24] pollutant concentrations in the exhaust plumes of individual trucks were measured from an instrumented van that was positioned on an overpass. Westbound trucks heading toward the Port drove underneath on a major access road (SI Figure S1). The roadway at this location is level, and trucks were observed to be accelerating from a traffic light ∼50 m before the sampling point or cruising at a speed of ∼30 mph. Exhaust/ambient air mixtures sampled above the roadway were delivered to the van via a flexible aluminum duct, as shown in SI Figure S2. Concentrations of CO_2, NO_x, NO, BC, PN, and particle size distributions were measured at 1 Hz or faster, using instruments listed in SI Table S2. A video camera at roadway level recorded truck license plates, which were later transcribed and matched with data in California's Drayage Truck Registry, including engine model year and DPF retrofit status.

A sample pollutant concentration time series showing peaks associated with three trucks that drove by in succession is presented in SI Figure S3. Pollutant concentration peaks were integrated to calculate fuel-based emission factors, expressed in units of amount of pollutant emitted per kg of fuel burned, using a carbon balance method:[27]

$$E_P = \frac{\int_{t_1}^{t_2}([P]_t - [P]_{t_1})dt}{\int_{t_1}^{t_2}([CO_2]_t - [CO_2]_{t_1})dt}\frac{44}{12}w_c \quad (1)$$

The emission factor for pollutant P (E_p) is calculated over the time interval $t_1 \le t \le t_2$, with t_1 and t_2 determined independently by the inflection points of each peak to account for the fact that instruments operated with different response times. The numerator and denominator respectively represent the baseline-subtracted peak areas for pollutant P and CO_2. When [P] and [CO_2] have mass concentration units (e.g., $\mu g\ m^{-3}$), the ratio compares the relative abundances of pollutant P and CO_2 present in the exhaust. The weight fraction of carbon in diesel fuel (w_c = 0.87) is used to convert emission factors from per mass of carbon to mass of fuel burned,[27] and the factor of 44/12 converts CO_2 to carbon mass. This carbon balance assumes that all fuel carbon is converted to CO_2 during combustion, with negligible emissions of CO and VOC relative to emitted CO_2.[16] NO_2 emission factors for each truck were computed as the difference of NO_x and NO emission factors, which were measured simultaneously using two separate chemiluminescent analyzers. NO_x emission factors were calculated using the molecular weight of NO_2.

Emission factors were computed for trucks when the peak CO_2 concentration rose more than 7% above baseline roadway concentrations, following Dallmann et al.[24] The baseline was taken to be the concentration measured just prior to the passage of a truck, with the timing determined from the roadway level video. Emission factors were computed only when the CO_2 peak could be definitively attributed to a single truck. Thus, no plume analyses were attempted when multiple trucks drove by at the same time or in close succession. In cases where CO_2 plume capture was successful but without clearly detectable peaks for other pollutants, near-zero values of emission factors were still computed, as illustrated in SI Figure S3.

The PN emission factors reported below were derived from an ultrafine, water-based condensation particle counter (CPC), as noted in SI Table S2. Normalized particle size distributions were measured using a fast mobility particle sizer (FMPS) to estimate size-resolved PN emission factors:

$$\Delta E_{PN} = \frac{\Delta N}{N}E_{PN} \quad (2)$$

Particle number concentrations measured in each size bin at the leading side of the particle number concentration peak, ΔN, were baseline-subtracted and normalized to the total particle number concentration, N. The product of this normalized size distribution and the FMPS-derived PN emission factor, E_{PN}, gives the particle emission rate in each size bin in units of 10^{15} particles emitted per kg of fuel burned.

The configuration of the particle sampling instruments used in this study is described in the SI and shown in SI Figure S4. An in-line dilution system was used to avoid exceeding the concentration limits of the CPCs used to measure PN concentrations. The dilution rate was actively monitored during the study. An aerosol photoacoustic absorption spectrometer was used in conjunction with an aethalometer to measure BC concentrations. The former instrument aided in postprocessing the aethalometer data to minimize the influence of the aethalometer's filter loading artifact, as described in the SI. Prior to field measurements, the full suite of instruments was staged and tested in the laboratory. In this assessment, we verified the NO_2 conversion efficiency of the chemiluminescent NO_x analyzers, identified a time response issue with a CO_2 analyzer used in prior studies at the Port of Oakland, and verified the CO_2 analyzer used in the current study did not suffer from the same issue. Additionally, we evaluated a sampling artifact of the FMPS in which an artificial increase in

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Table 1. Average Emission Factors (± 95% Confidence Interval) for Heavy-Duty Diesel Drayage Trucks Characterized by Engine Control Technology and Engine Model Year[a]

fleet or truck category	range of engine model years	median engine model year	number of trucks[c]	NO_x (g kg^{-1})	NO_2 (g kg^{-1})	NO_2/NO_x emission ratio	BC (g kg^{-1})	PN (10^{15} particles kg^{-1})
2009 Fleet[b] (2% DPF, 0% SCR)	1970–2009	1997	169–172	32.6 ± 2.3	1.11 ± 0.58	0.034 ± 0.018	1.15 ± 0.19	N/A
2011 Fleet (54% DPF, 2% SCR)	1994–2011	2004	359–378	18.0 ± 1.2	2.10 ± 0.39	0.117 ± 0.023	0.67 ± 0.14	3.59 ± 0.81
2013 Fleet (99% DPF, 9% SCR)	1994–2013	2007	960–1005	15.4 ± 0.9	2.84 ± 0.22	0.184 ± 0.018	0.28 ± 0.05	2.47 ± 0.48
Retrofit DPF	1994–2006	1998	390–401	26.0 ± 1.3	3.91 ± 0.38	0.150 ± 0.017	0.32 ± 0.06	2.61 ± 0.76
No DPF	2004–2006	2005	178–188	16.5 ± 1.7	0.56 ± 0.28	0.034 ± 0.018	1.11 ± 0.26	4.72 ± 0.97
DPF	2007–2009	2008	657–695	11.9 ± 0.9	2.68 ± 0.27	0.225 ± 0.029	0.26 ± 0.06	2.52 ± 0.62
DPF + SCR	2010–2013	2011	93–99	5.1 ± 1.2	1.14 ± 0.27	0.221 ± 0.084	0.09 ± 0.04	1.59 ± 1.15

[a]Results shown in the last four rows are based on combined data from 2011 and 2013. [b]2009 emission factors are from Dallmann et al.[24] after adjustment to account for a CO_2 sampling artifact and consistent application of a time-dependent correction of the aethalometer, as described in the SI. The 2009 NO_2 emission factor was estimated based on the NO_2/NO_x emission ratio measured in this study in 2011 for trucks without DPFs, as indicated in the text. [c]The number of trucks used for each fleet or truck category analysis depended on the data available from each instrument; the maximum number in the given range corresponds to NO_x, NO_2, and NO_2/NO_x calculations and the minimum number typically refers to UFP. The sample size for BC analysis generally falls in the middle of the range.

UFP less than 10 nm was reported during periods of rapidly decreasing PN concentrations, which subsequently guided our use of the data from this instrument. These instrument evaluations and related issues are discussed in greater detail in the SI.

RESULTS AND DISCUSSION

Port Truck Age Distribution. The age distribution of trucks operating at the Port of Oakland changed dramatically over a short period of time, as shown in Figure 1. In 2008, the median engine age for Port trucks was 11 years, and only 2% of trucks had 2007 or newer engines equipped with DPFs.[28] By early 2013, 99% of trucks were equipped with DPFs, 9% had 2010 or newer engines with both DPF and SCR systems, and median engine age decreased to 6 years.

Average Emission Rates. Fleet-average emission factors for Port trucks are presented in Table 1. Emission factors measured previously in 2009[24] are compared to those measured in 2011 and 2013. Also, emission factors from the 2011 and 2013 measurement campaigns were together disaggregated into four truck categories based on engine model year and installed emission controls. Results are reported in Table 1 and shown in Figures 2 and 4, and SI Figures S9 and S10 for (1) 1994–2006

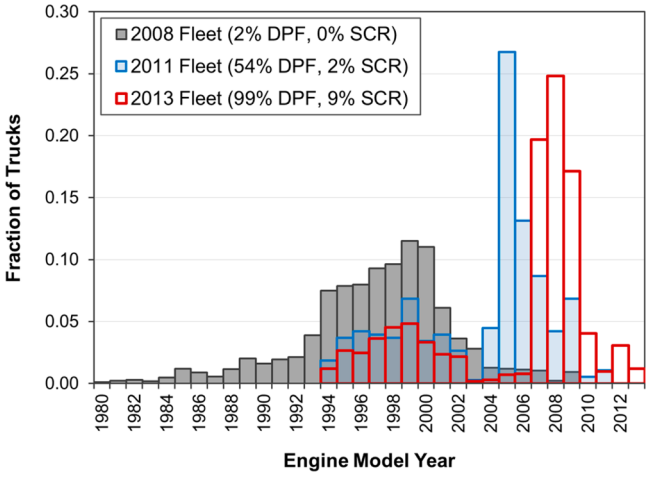

Figure 1. Distribution of heavy-duty diesel truck engine model years at the Port of Oakland prior to California's Drayage Truck Regulation in 2008[28] and after phased implementation of the regulation in 2011 and 2013. The model year distribution from 2008 is based on the truck chassis; some engines may be one year older than the chassis.

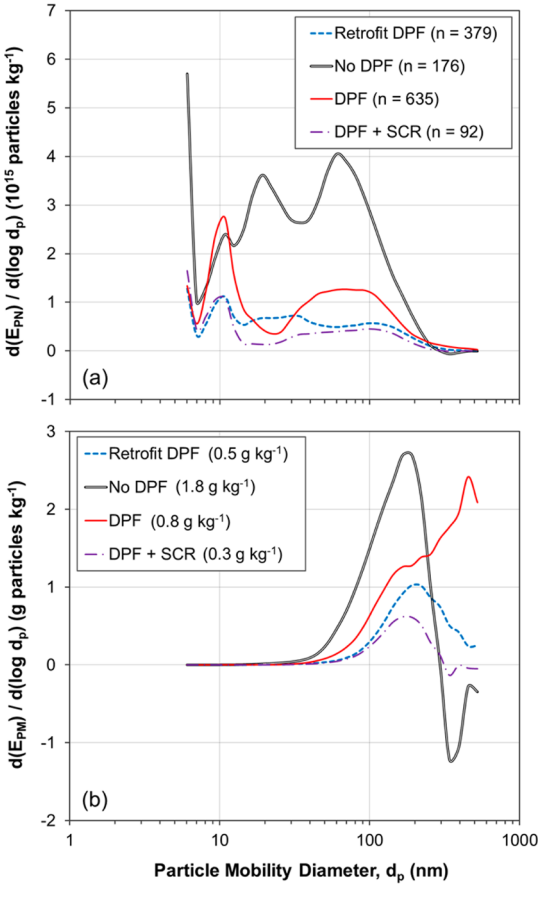

Figure 2. Characteristic particle (a) number and (b) mass emission rate distributions for each emission control technology, based on combined 2011 and 2013 data. The particle mass emission factor estimated from each size distribution is noted in the figure legend. Note that data above ~200 nm in (b) are not reliable, given low particle number concentrations in this size range as shown in (a).

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engines with retrofit DPFs, (2) 2004−2006 engines without DPFs, (3) 2007−2009 engines with DPFs, and (4) 2010−2013 engines with DPFs and SCR. Unless explicitly labeled as a retrofit, DPF and SCR controls were installed as original equipment at the time of manufacture. In Table 1 and elsewhere, uncertainty ranges represent 95% confidence intervals and model years refer to the engine, not the truck chassis.

Nitrogen Oxides. Due to fleet modernization and associated decreases in median engine age, the fleet-average NO_x emission factor decreased by 53 ± 8% between 2009 and 2013 (Table 1). The NO_2/NO_x emission ratio in 2009 was not measured, but was assumed to be similar to that for 187 trucks without DPFs measured in 2011 (Table 1). A baseline NO_2 emission factor for the 2009 fleet was estimated by applying this NO_2/NO_x ratio of 0.03 ± 0.02 to the measured fleet-average NO_x emission factor. The NO_2/NO_x emission ratio increased from 0.03 to 0.18 between 2009 and 2013 as use of DPFs on Port trucks became universal.

NO_2/NO_x emission ratios were highest for newer trucks with DPFs, with or without SCR systems, but engines with SCR had lower absolute NO_2 and NO_x emission rates (Table 1, SI Figure S9). The NO_2 emission rate for trucks with SCR systems was comparable to the low value determined for the pre-DPF truck fleet measured in 2009. This highlights the important role of SCR in mitigating the undesired increase in primary NO_2 emissions associated with the use of DPFs to control exhaust PM emissions.

Black Carbon. Between 2009 and 2013, fleet-average BC emission factors decreased by 76 ± 22% (Table 1). BC emission factors for older trucks with retrofit DPFs and 2007−2009 trucks originally equipped with DPFs were similar (Table 1, SI Figure S10). The newest trucks with 2010+ engines equipped with both DPFs and SCR had the lowest BC emission factors. Relative to modern trucks (2004−2006 engines) without these emission controls, trucks with pre-2010 engines equipped with DPFs emitted 74 ± 30% less BC. The newest trucks (2010+ engines) emitted 92 ± 32% less BC than the 2004−2006 trucks, which is consistent with prior laboratory studies of DPF effectiveness.[12−14] As the newest trucks enter into Port service, BC emissions could decrease to ∼10% of 2009 levels, with little if any increase in primary NO_2 emissions.

Particle Number. The PN emission rate measured for 178 trucks without DPFs is used here as a reference value to approximate baseline conditions in 2009 (Table 1). Relative to this baseline of $(4.7 \pm 1.0) \times 10^{15}$ particles kg^{-1}, there was a 47 ± 25% reduction in the fleet-average PN emission rate by 2013, when DPFs were required on all Port trucks. Trucks with 2010+ engines had the lowest PN emission rates, emitting one-third the number of particles per kg of fuel burned compared to trucks without DPFs (Table 1, SI Figure S10).

PN emission factors presented above are based on total particle number concentrations measured with an ultrafine water-based CPC. As presented in SI Figure S11 and Table S3, results were highly correlated (R^2 = 0.86) but 30% higher, on average, than PN emission factors for the same trucks calculated from measurements made using an ultrafine butanol-based CPC. This difference could be due to CPC sensitivity to particle composition that depends on the condensing fluid (i.e., water versus butanol), or due to other differences in CPC design.[29] PN emission factors derived from ultrafine water-based CPC and FMPS measurements were not as well correlated (R^2 = 0.49). On average, emission factors based on the ultrafine water-based CPC were 80% higher than those based on the FMPS. The lower values derived from FMPS measurements may in part be because the ultrafine CPC measures particles as small as 2.5 nm, whereas the low cutoff of the FMPS is 5.6 nm. This finding agrees with Jeong and Evans,[30] who noted that ultrafine water-based CPC measurements of PN concentration exceed FMPS measurements. In contrast, Zimmerman et al.[31] found that the FMPS can overstate PN concentrations when measuring emissions from high-emitting vehicles at high time resolution.

Emission factor-weighted particle size distributions were used to derive a characteristic particle number emission profile for each truck category. Measured size distributions for each truck were weighted by corresponding FMPS-derived PN emission factors. As shown in Figure 2, particle emissions ranging in size between 5.6 and ∼300 nm were measured, with a majority occurring in the ultrafine mode below 100 nm. Particle size distributions for all four groupings of trucks include an apparent sharp increase in the smallest size bin of the FMPS. We present these data as measured, though it is unclear if they are truly indicative of a peak in the number concentration of particles smaller than the lower sizing limit of the FMPS.

Figure 2 shows that the average size distribution for all trucks without DPFs was trimodal, including broad peaks around 10, 20, and 80 nm. Overall, DPFs appear to be most effective in reducing the emission rate of particles larger than ∼15 nm. On average, the emission rates of such particles from trucks without filters were approximately 3.5 times those from DPF-equipped trucks. These results indicate that use of DPFs on drayage trucks for controlling particle mass does not increase the emission factor of nucleation mode particles, which was a potential concern raised in other studies.[12,18] DPFs on trucks with 2007 and newer engines typically include active filter regeneration systems (e.g., periodic injection of unburned fuel to oxidize trapped particles). Nucleation is likely to occur during such active regeneration events,[18] with increased emissions of <30 nm particles.[32] In this study, the emission factor of ∼10 nm particles from 2007 to 2009 DPF-equipped engines was 2.5 times the levels measured from both older truck engines with retrofit DPFs and 2010+ engines equipped with DPFs and SCR.

Particle Mass. Since direct measurements of PM mass were not made during this study, the size-resolved FMPS particle count data was used to estimate mass emissions. Assuming spherical particles with a density of 1 g cm^{-3} across the entire particle size range, PN distributions were converted into mass emission distributions. The PM emission factor for each truck category could ideally be estimated from the integrated area under each respective mass emission rate distribution. However, low number concentrations in the larger size ranges result in noise in the upper size bins that is magnified when number concentrations are converted to mass concentrations, as seen in Figure 2b. Therefore, PM emission factors corresponding to each truck category were determined assuming log-normal distributions and doubling the area to the left of the apparent peak value of each mass emission rate distribution. This peak in the mass emission distribution was typically around 200 nm. Recent studies indicate that additional particle mass exists beyond the upper size limit of the FMPS for diesel exhaust.[33,34] Therefore, the PM emission factors derived from FMPS measurements and reported in Figure 2b may understate the true PM emission rates. The estimated PM

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emission factor for 2010+ engines equipped with both DPFs and SCR was ~86% lower than that found for 2004–2006 engines without these emission controls. This decrease in PM emissions is a little smaller than the 92% decrease found for BC (Table 1). Similarly, the average reduction in estimated PM emission factor for all DPF-equipped trucks compared to trucks without filters (~72%) was slightly lower than that found for BC (80%).

Pollutant Relationships. As shown in SI Figure S12, the highest emitters of BC tended to have low emissions of UFP and NO_2—and vice versa—regardless of the type or vintage of emission control equipment. Such a relationship between BC and PN has been observed previously.[27] This result is consistent with the hypothesis that UFP formation is suppressed when large amounts of particle surface area are available, such that condensation onto existing particle surfaces is favored over nucleation to form UFP.[19] Likewise, the relationship between BC and NO_2 has been reported previously,[16] and is the expected result of well-functioning DPF systems, which reduce BC mass emissions while deliberately oxidizing NO to NO_2 to aid in filter regeneration.

Emission Factor Distributions. As fleet-average emission factors have decreased over time, emission factor distributions have become increasingly skewed, such that a small fraction of the fleet is responsible for an increasing fraction of total emissions. As shown in Figure 3 and SI Figure S13, particle-related emission factor distributions are more skewed than those for nitrogen oxides. In 2013, the highest emitting 10% of trucks were responsible for 65% of total BC and 80% of total PN, compared to only 32% of total NO_x emissions (Figure 3b). The skewness of NO_x emission factor distributions is increasing, though, and this trend is likely to continue as the number of engines equipped with SCR increases in future years (SI Figure S13a).

Emission factor distributions are shown separately for each engine model year, as measured in 2011 and 2013, in Figure 4. Trucks equipped with DPF and SCR systems not only had the lowest BC and NO_x emission factors, but also showed the least amount of variability in measured emission rates. The upper range of measured emission rates for many individual engine model years increased in 2013 relative to 2011, suggesting possible degradation or failure of some installed emission control systems over time. Our analysis identified two DPF-equipped trucks as the highest BC emitters in 2013 (Figure 4a). These trucks had emission factors of ~10 g BC kg^{-1}, significantly higher than the 2013 fleet-average of 0.28 ± 0.05 g kg^{-1} and approximately double the emission rate of next highest-emitting truck. These two high-emitting trucks represented 0.2% of the total number of trucks measured, but were responsible for 7% of total BC emissions (Figure 3b). This analysis also shows that trucks with 2007–2009 engines frequently emit NO_x at levels that are similar to what is observed from older trucks, even though the average emission rate for older trucks is approximately twice as high (Table 1 and Figure 4b). In summary, even though average emissions of BC and NO_x have decreased, some newer trucks were observed to emit BC and NO_x at high levels.

Identification and follow-up actions leading to repair of high-emitting trucks may be needed to help ensure that emission control systems remain in good working order, and to extend the early success in emission control efforts reported here. For instance, if the top 10% of BC emitters from the 2013 fleet were removed (Figure 3b), then fleet-average BC emissions

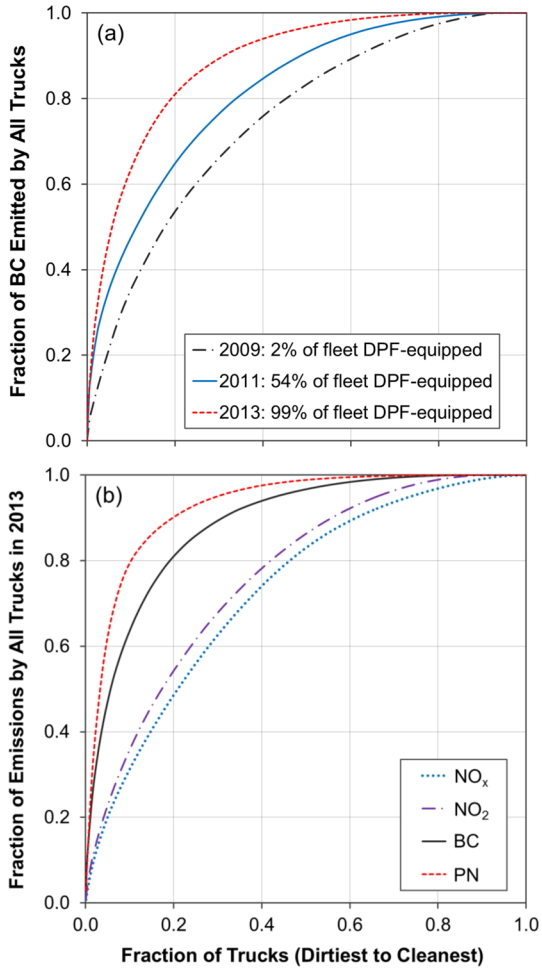

Figure 3. Cumulative emission factor distributions for (a) BC over time and (b) multiple pollutants in 2013, with trucks ranked from highest to lowest in terms of emission factors.

would decrease from 0.28 ± 0.05 to 0.11 ± 0.01 g kg^{-1}. The trucks that comprise this high emitting subfleet had an average BC emission factor of 1.80 ± 0.10 g kg^{-1}, which was approximately six times the 2013 fleet-average value. A number of the trucks included in the top 10% were measured multiple times in this study, and 64% of the repeat measurements were also included in the top 10% of BC emissions. This suggests that the high emitter problem for BC is more often chronic rather than intermittent in nature. If all repeat measurements of BC by trucks classified as in the dirtiest 10% of the 2013 fleet are included, the average BC emission factor of 1.36 ± 0.10 g kg^{-1} remains much higher than the overall fleet average. Elimination of these high-emitting trucks through repair or replacement would further increase fleet-average BC reductions measured in this study from 76 ± 21% (Table 1) to 90 ± 22%.

Emissions Variability. Differences in operating conditions can influence pollutant emission rates. This variability can be studied in the laboratory using different driving cycles, but laboratory testing is costly and generally limited to a small sample size. In-use emission evaluations as in the current study, on the other hand, can assess the emission performance of thousands of trucks under real-world conditions. These measurements provide emissions snapshots, though, and may not capture relevant variability in emissions. In the current study, we used repeat measurements of trucks that drove by

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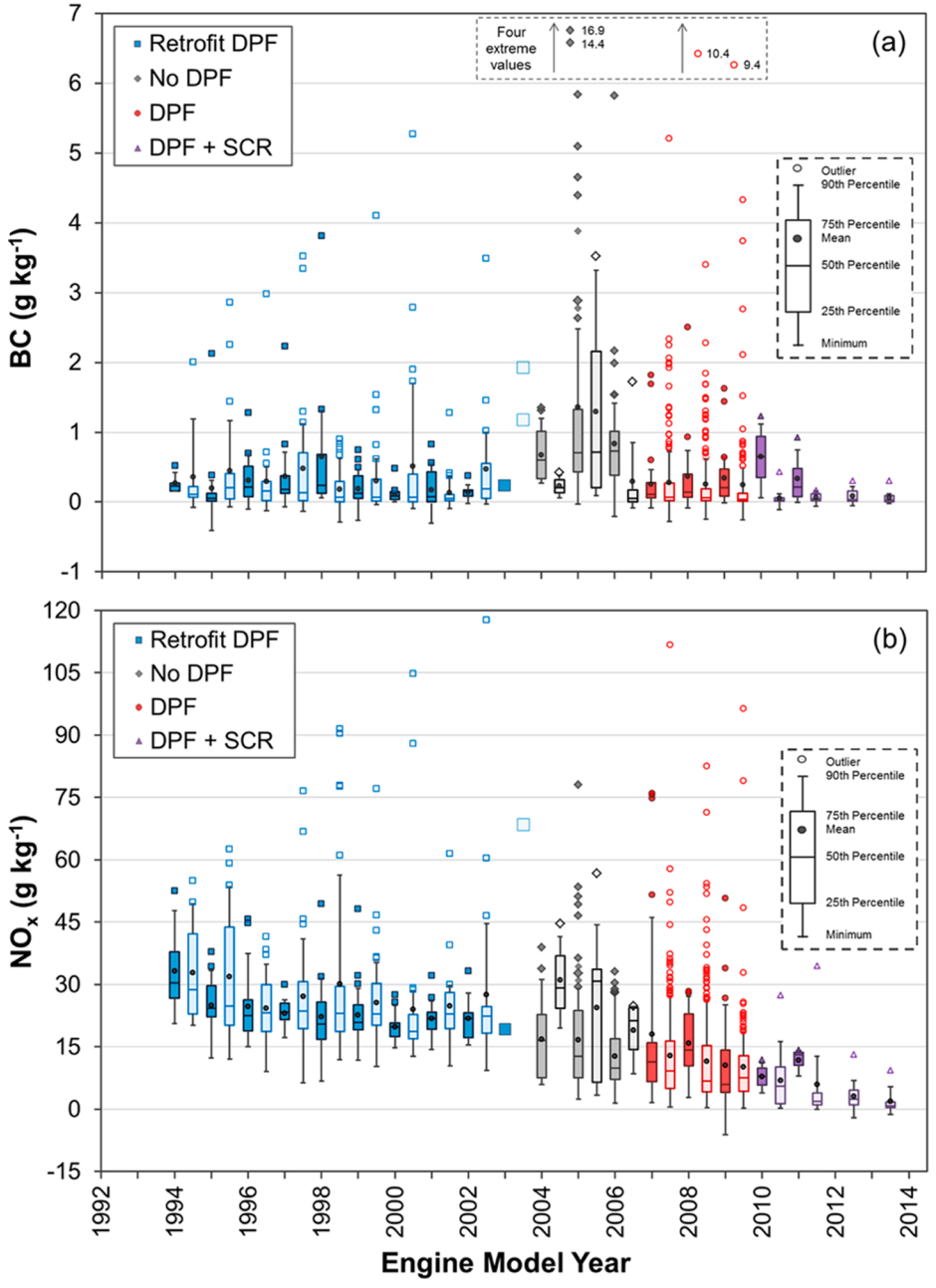

Figure 4. Distribution of (a) BC and (b) NO_x emission factors across engine model years for each individually measured truck. The truck categories as measured in 2011 are shown in shaded boxes and whiskers, and those measured in 2013 are shown in transparent boxes and whiskers. The larger number of outliers for 2013 measurements does not reflect a greater fraction of higher emitting trucks, but is instead the result of generally larger sample sizes during that campaign (Table 1). Also, note that there are four extreme outliers for the BC distribution that exceed the range shown.

multiple times to characterize emissions variability at our sampling location.

Repeat measurements of emissions from 207 individual trucks are plotted in Figure 5 and SI Figure S14 against corresponding average emission factors for each truck. Scatter about the indicated 1:1 line is due to variability in emission factors. Emissions rates for NO_x and BC were found to vary less than NO_2 and PN. Measurement uncertainties contribute to variability in NO_2 emission factors especially, as NO_2 was calculated by difference (i.e., $NO_2 = NO_x - NO$). While fleet-average results for PN can still be evaluated, variability in emissions of this pollutant limits the usefulness of single snapshot measurements to characterize emissions from individual trucks.

Emissions Representativeness. Measurements made in this study do not include trucks traveling at highway speeds, where engine load and exhaust temperatures are high. This is relevant because these parameters can affect emission control system performance. For example, SCR systems are ineffective when exhaust temperature is low, which can occur during cold starts and at low load/low speed.[35] However, NO_x emissions from the SCR-equipped trucks in this study were very low (Figure 4b), which suggests that SCR systems were likely functioning when emissions were sampled. Increased UFP emissions from DPF-equipped trucks have been reported during highway driving when exhaust temperatures were high and during active DPF regeneration events.[18,32] Active filter regeneration can also increase emitted PM mass.[32] In this

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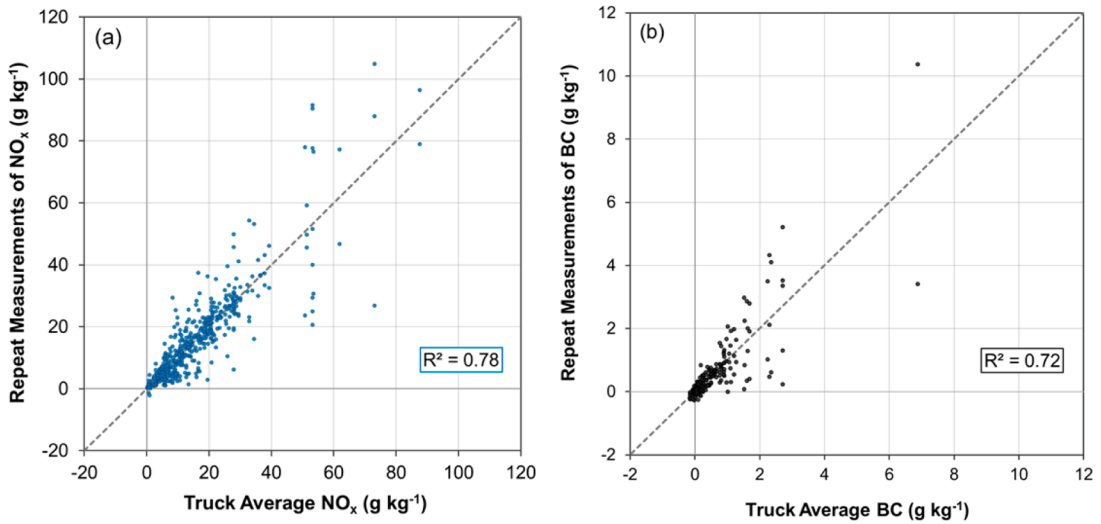

Figure 5. Repeat (a) NO_x and (b) BC measurements for 207 individual trucks, with each individual measurement plotted on the vertical axis against the corresponding average emission factor for each truck on the horizontal axis. Repeatability of the measurement is indicated by the degree of scatter of data points about the dashed 1:1 line.

study, exhaust temperatures were not measured and it was not possible to determine the extent to which filters were actively regenerating.

Acceleration of Emissions Reductions. Emission reductions observed in this study at the Port of Oakland over ~3.5 years are nearly double the 39 and 30% reductions in BC and NO_x emission factors measured for trucks at the nearby Caldecott Tunnel over a period of 9 years.[8] Trucks using the Caldecott Tunnel were not subject to any retrofit/replacement requirements during the period from 1997 to 2006 considered by Ban-Weiss et al.[8] Emission reductions at the Port have clearly occurred at a pace well beyond what can be achieved by natural fleet turnover alone. Programs requiring accelerated replacement of older trucks now extend beyond ports and rail yards to include heavy-duty trucks operating anywhere in California, regardless of origin or destination. Results of these local, accelerated changes at the Port of Oakland therefore provide a preview of how diesel truck emissions are likely to change across the state in the next few years, as well as nationally as larger numbers of newer trucks with advanced emission control systems enter into service.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information (SI) includes additional details about the sampling location and method, laboratory assessments of sampling artifacts, and supplementary results, as specified in the manuscript. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b01117.

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Notes

The authors declare no competing financial interest.

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Effects of Particle Filters and Selective Catalytic Reduction on Heavy-Duty Diesel Drayage Truck Emissions at the Port of Oakland

SUPPORTING INFORMATION

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INTRODUCTION

In California, the introduction of DPFs has been accelerated by the Statewide Drayage Truck Regulation. Over a three year period—from Jan 1, 2010 to Dec 31, 2012—all drayage trucks were required to be equipped with DPFs, either via retrofit or engine replacement.[1] Table S1 outlines the implementation schedule of this regulation.

Table S1. Phased implementation schedule for the Statewide Drayage Truck Regulation.

Phase of Regulation	Deadline	Engine Model Year	Requirement
Phase 1	January 2010	1993 & Older	Banned
		1994 - 2003	Retrofit with DPF or replace with newer engine
	January 2012	2004	
	January 2013	2005 - 2006	
Phase 2	January 2014	1994 - 2006	Replace with 2007 or newer engine

METHODS

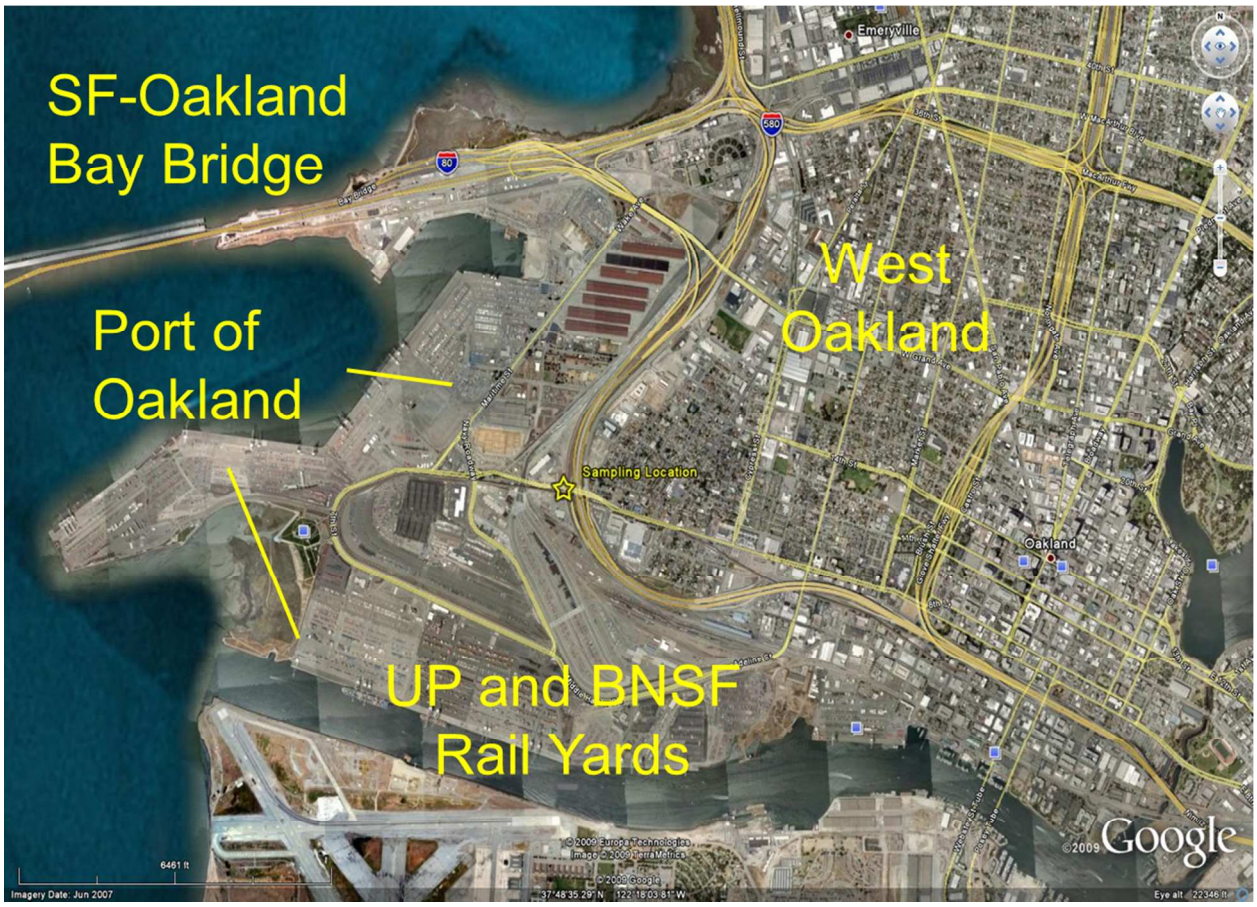

Figure S1. Map of the Port of Oakland, Union Pacific and BNSF rail yards, nearby community of West Oakland, and sampling location (noted by a star). Map data/image provided by Google, Europa Technologies, and Terrametrics © 2009.

Figure S2. Instrumented van positioned on an overpass sampling the exhaust from a truck en route to the Port of Oakland via a flexible aluminum duct (7.6 cm diameter).

Table S2. Instrumentation used to measure truck exhaust emissions in this study.

Parameter	**Instrument**	**Time Resolution**
CO_2 concentration	Nondispersive infrared gas analyzer (LI-COR LI-7000)	2 Hz
NO, NO_x concentrations (the difference of which gives NO_2 concentration)	Two chemiluminescent analyzers (ECO Physics CLD-64)	2 Hz
BC concentration	Aethalometer (Magee Scientific AE16)	1 Hz
BC concentration	Photoacoustic absorption spectrometer (PAS) with reciprocal nephelometer (custom)	1 Hz
PN concentration*	Ultrafine, water-based condensation particle counter (TSI 3788); lower size limit of 2.5 nm and noted as water CPC	2 Hz
PN concentration	Ultrafine, butanol-based condensation particle counter (TSI 3776); lower size limit of 2.5 nm and noted as butanol CPC	10 Hz
PN concentration, dilution factor	Two general purpose water-based condensation particle counters (TSI 3787 and 3783); lower size limit of 7 nm, with a high concentration CPC used upstream of in-line dilution system and standard CPC used downstream	2 Hz
PN concentration, size distribution	Fast mobility particle sizer (TSI 3091); lower size limit of 5.6 nm and noted as FMPS in above text and in Table S3 and Figure S9	1 Hz

*The reported PN emission factors in Table 1 and Figures 3, S10b, S12b, and S14d are derived from this CPC.

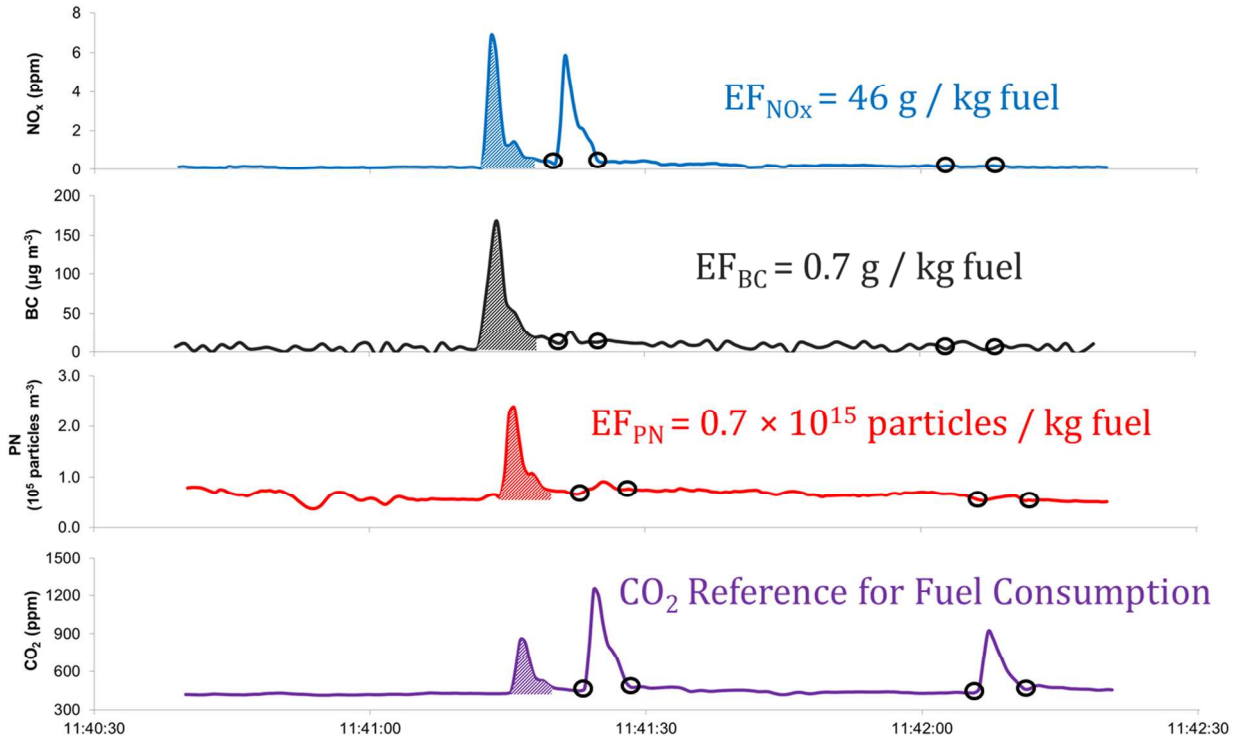

Figure S3. Pollutant concentration time series showing peaks that correspond to the exhaust plumes of three trucks. The first truck emitted appreciable amounts of NO_x, BC, and PN. The shaded peaks correspond to the integrated areas used to compute the emission factors shown in the figure. The second and third trucks emitted much smaller BC and PN concentrations and the third truck emitted essentially no NO_x. The integration boundaries are indicated with open circles for the second and third trucks.

The particle sampling configuration shown in Figure S4 was designed to minimize wall losses by inertial separation and diffusion. Laminar flow was maintained in all sampling lines to minimize particle losses due to turbulence. Diffusional losses of ultrafine particles (UFP) were further minimized by using the high sampling flow rate of the fast mobility particle sizer (FMPS) as a source of transport flow for the condensation particle counters (CPCs) to avoid long runs of slow moving sample flows to all instruments measuring particle number (PN) and size. Using a standard laminar diffusion model, line losses of 10 nm particles were estimated as ~10% for the CPCs, 2% for the FMPS, and 1% for the dilution system's mixing tube.

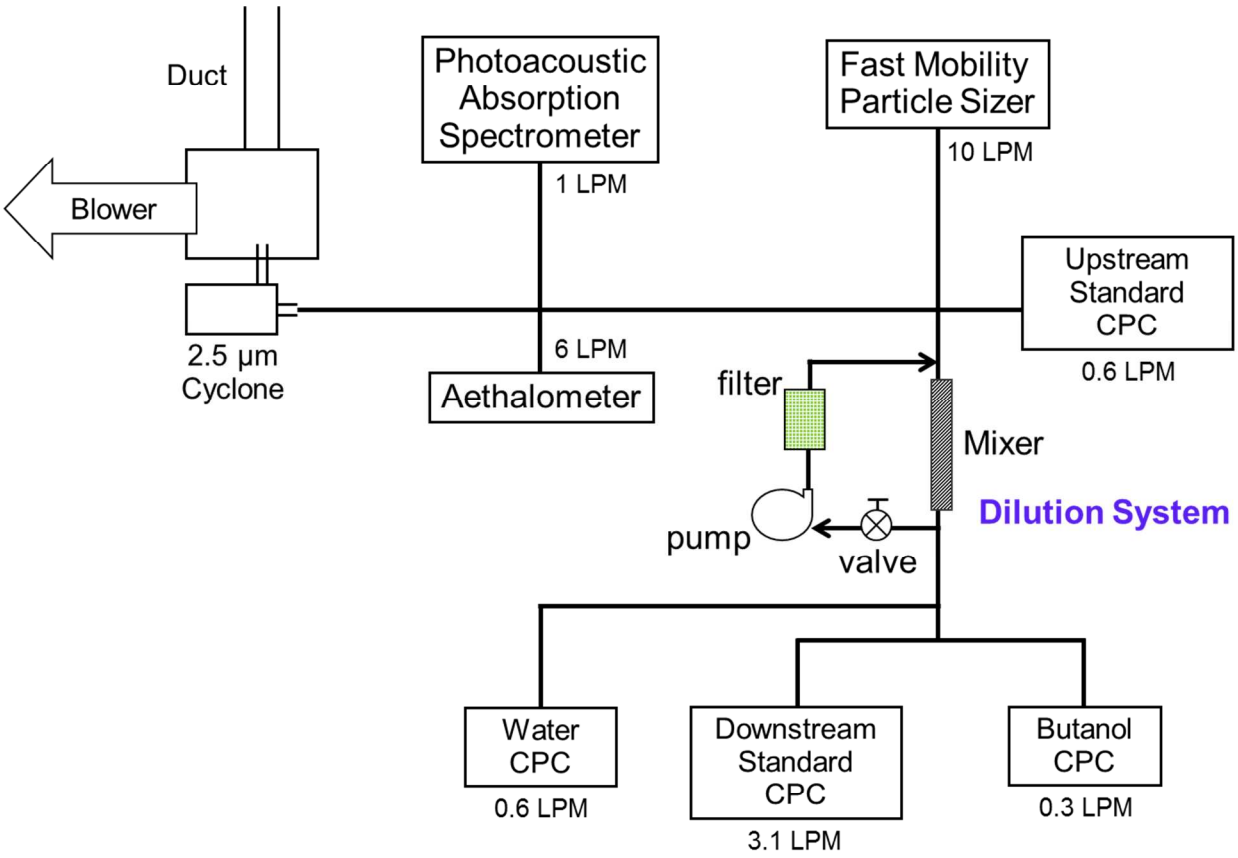

Figure S4. Schematic of particle sampling instruments and their flow rates, and the in-line dilution system.

Moreover, an in-line dilution system was used to avoid exceeding the concentration limits of the CPCs used to measure PN concentrations. The dilutor consisted of a filtered recirculating flow in a closed loop that drew from the outlet side of a standard finned-tubular flow mixer and injected the particle-free flow back into the upstream side of the mixing tube to be combined with the incoming aerosol flow. A matched pair of standard water-based CPCs was placed on the upstream and downstream sides of the dilutor to actively monitor the dilution rate. All downstream particle number concentrations were post-corrected to account for the dilution factor, which was typically ~6. The dilution factor was determined from the regressed slope of

the line forced through zero, with the upstream CPC concentration data as the dependent variable and the downstream CPC concentration as the independent variable. Slight mismatches in data acquisition timing between the independent, asynchronous data streams was handled by averaging down time series with successively larger averaging windows until an asymptotic value was obtained for the regressed slope.

Prior to field measurements, the full setup of instruments and data logging were staged in the laboratory and exposed to the exhaust of an inverted methane-air diffusion flame.[2] The measurement of exhaust plumes of passing trucks was simulated by episodically sampling flame exhaust instead of filtered room air. The analysis of multiple peaks under constant flame exhaust conditions established the precision of the plume capture sampling and carbon balance analysis methods that were used in this study. The NO_2 conversion efficiency of both NO_x analyzers was also evaluated using ozone titration tests to ensure accuracy of total NO_x ($NO + NO_2$) measurements. Multipoint calibrations were conducted for all gas analyzers prior to deployment in the field. At the beginning and end of each sampling day, these calibrations were verified with zero and span checks.

Laboratory testing identified a significant time response issue for the LI-COR model 820 CO_2 analyzer, which tends to overshoot in reporting peak concentrations when rapid transitions occur. Therefore, a faster response LI-7000 instrument was used in the present study, as it did not suffer from the same problem (Figure S5). The LI-820 CO_2 analyzer was used in parallel during the 2011 field study to assess the magnitude of possible biases in earlier measurements. Figure S6 shows a frequency distribution of the ratio of integrated CO_2 peaks measured using LI-820 and LI-7000 analyzers. Use of LI-820 data led to overestimates in the magnitude of CO_2 peak areas by 26 ± 2% (95% confidence interval) for a sample of 389 trucks (Figure S6).

Therefore, baseline pollutant emission factors measured in an earlier field measurement campaign at the Port of Oakland in 2009[3] were multiplied by 1.26 in making comparisons with emission factors measured using the more accurate LI-7000 CO_2 analyzer in the 2011 and 2013 campaigns at the Port.

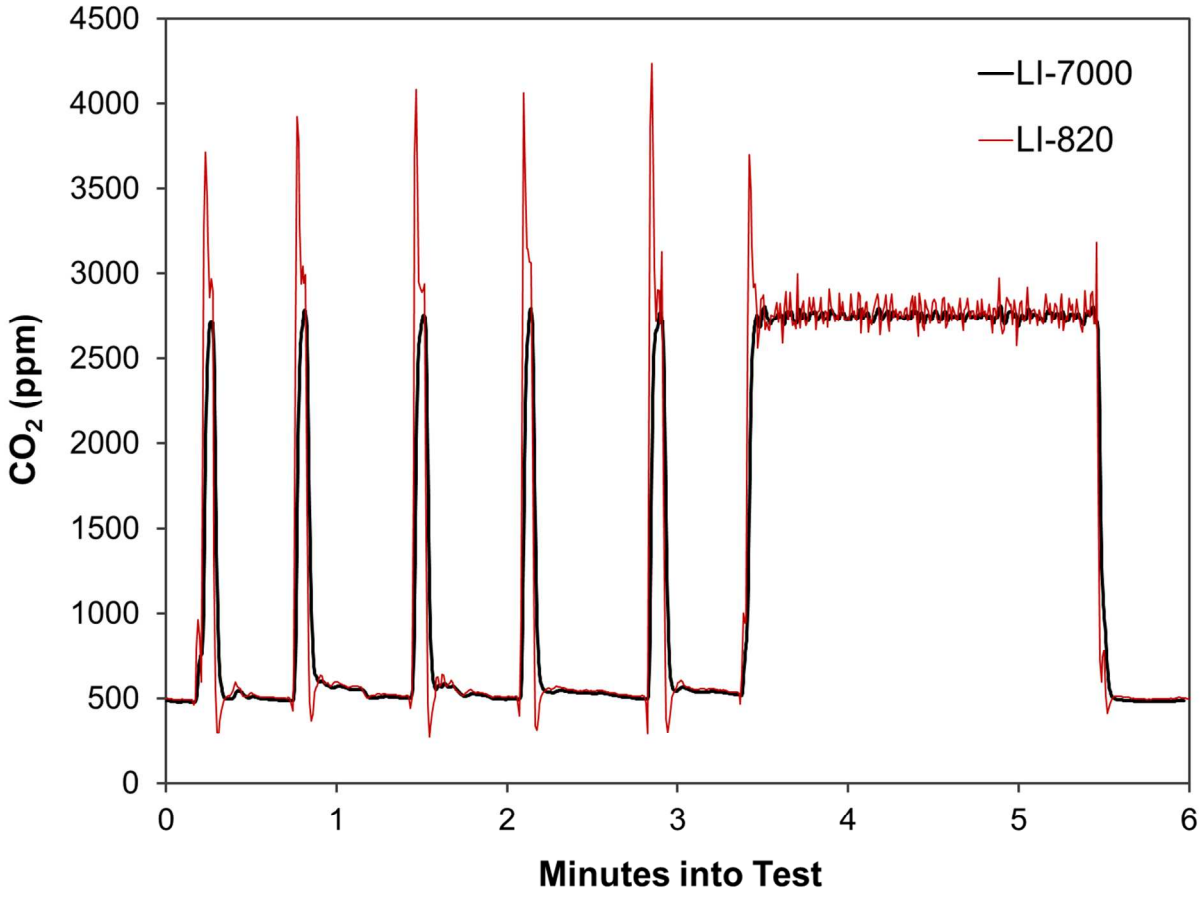

Figure S5. Comparison of time series of CO_2 concentrations measured in laboratory evaluation of two LI-COR CO_2 analyzers: LI-820 and LI-7000. The LI-820 overshoots when concentrations change rapidly, whereas the high performance LI-7000 transitions smoothly to match new peak concentrations.

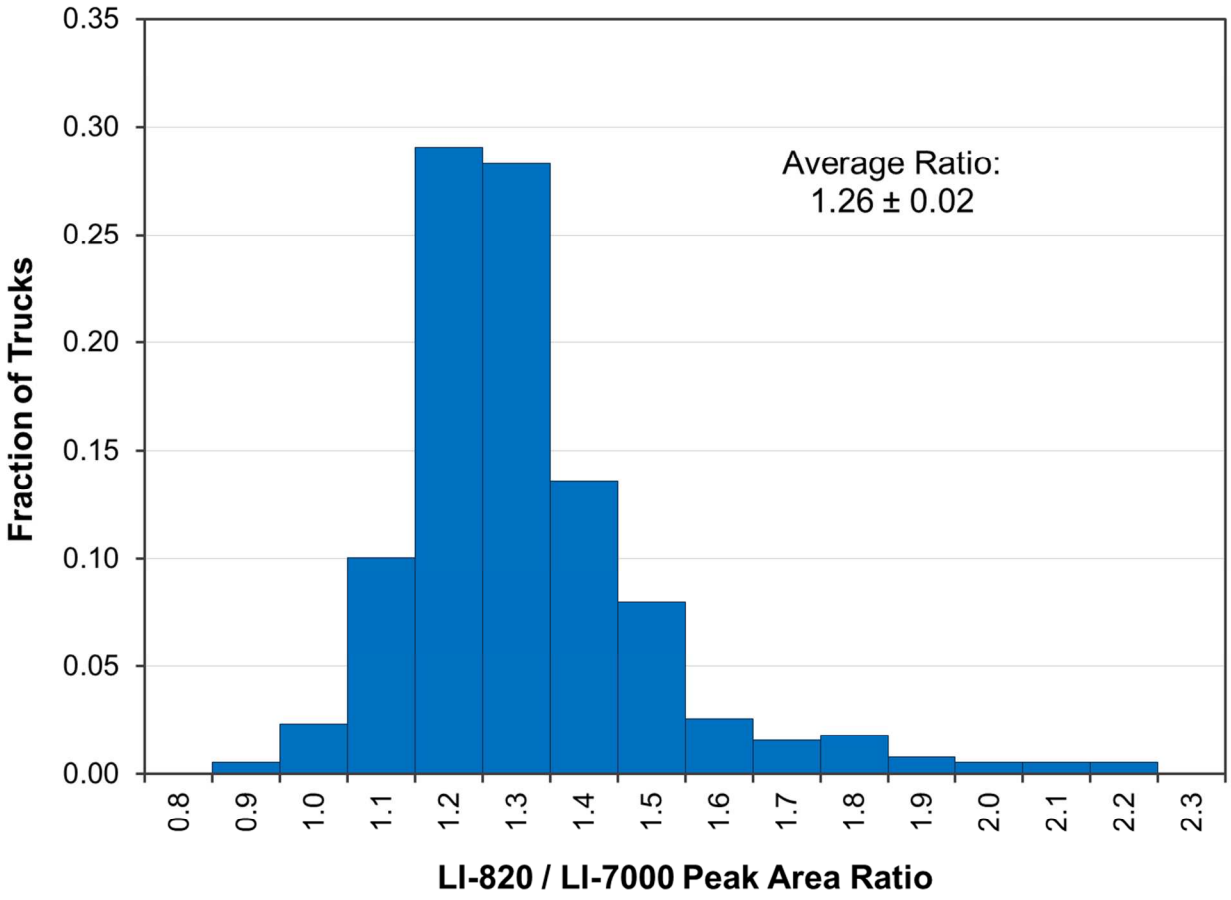

Figure S6. Distribution of average ratio of CO_2 peak areas measured by LI-820 and LI-7000 analyzers, as determined from concurrent measurements during the 2011 study at the Port of Oakland.

The laboratory tests also identified a concern associated with particle size distributions measured using the FMPS. Jeong and Evans[4] previously reported an unexpected peak near the lower size limit (below 10 nm) of the FMPS when sampling ambient air in both urban and rural settings. This was posited to be due to either the data inversion algorithm or the calibration of the FMPS electrometers used for particle detection. In our tests, the FMPS overstated the concentration of UFP with diameters less than 10 nm on the trailing side of peaks when particle number concentrations were rapidly decreasing. Figure S7 shows an example of this effect. Accordingly, when analyzing particle size distributions measured at the Port of Oakland,

representative particle size distributions for each truck were chosen from the leading side of the particle number concentration peak.

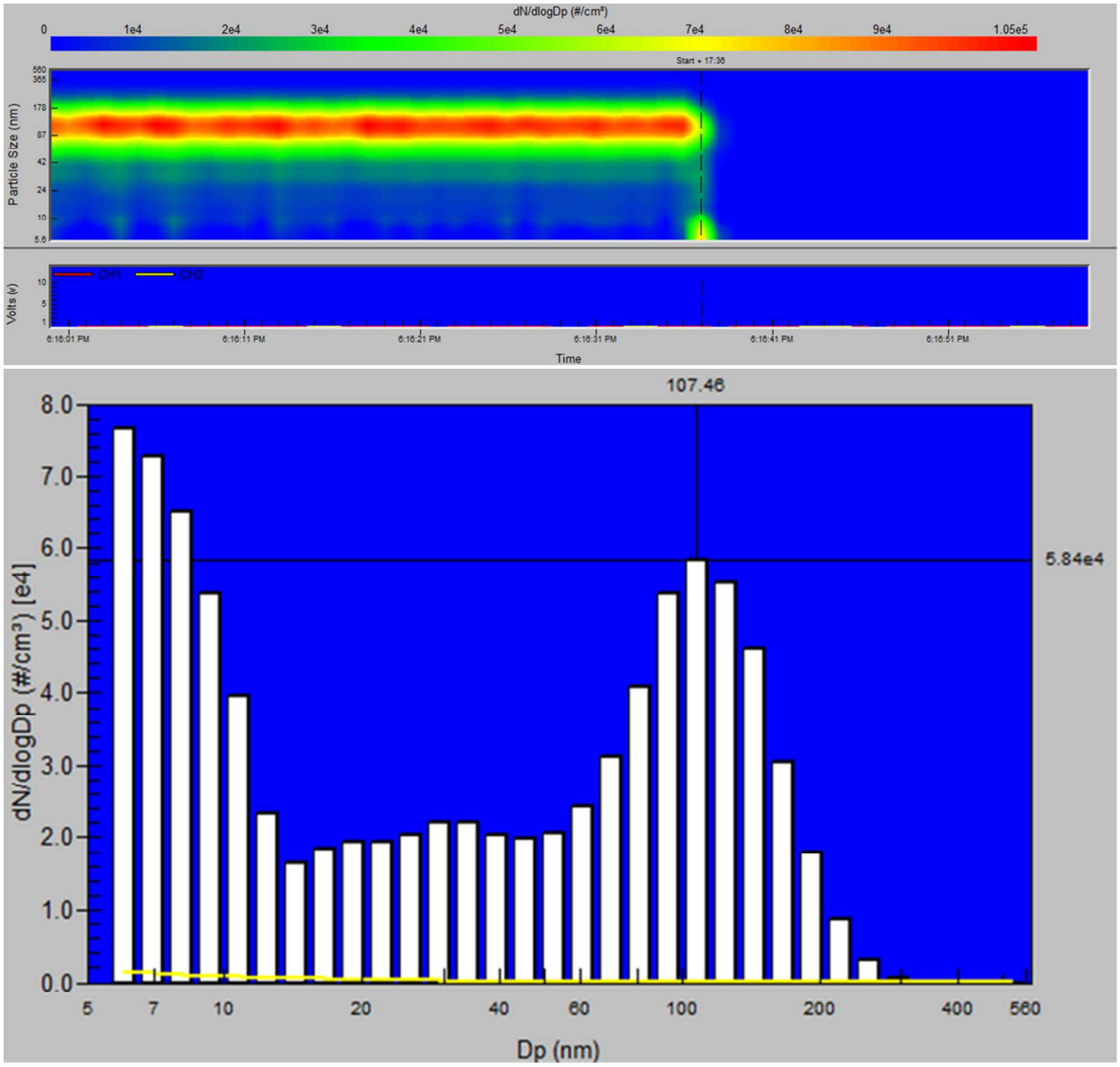

Figure S7. FMPS data showing an example of the artificial increase in UFP number concentration of particles below 10 nm in diameter when particle number concentrations were rapidly decreasing. The top panel shows the measured particle number concentration, sampled from a constant source in the laboratory. The vertical dashed line shows the time at which the number concentration decreased and the artificial pulse of particles <10 nm was reported. The bottom panel shows the corresponding particle size distribution at that time, including an anomalous peak in particle sizes below 10 nm.

Aethalometer BC data were post-corrected for time-dependent behavior that causes the instrument response to decline with increasing filter darkening.[2] A modified version of the correction equation developed by Kirchstetter and Novakov was used to adjust the BC concentration reported by the aethalometer:

$$BC = \left[\frac{BC_0}{a \exp\left(\frac{-ATN}{100}\right) + (1 - a)} \right] \quad \text{(S1)}$$

where BC and BC_0 are the adjusted and unadjusted BC concentrations, respectively, and ATN is the attenuation of light by the filter. The correction parameter, a, adjusts BC_0 such that BC concentrations are independent of filter loading. This correction is validated by plotting the ratio of the light absorption coefficient measured with the photoacoustic absorption spectrometer (PAS) and the BC concentration measured with the aethalometer, as shown in Figure S8. Both instruments respond to light-absorbing PM, but the in-situ PAS measurement is not affected by filter loading effects. As shown, the ratio of absorption coefficient to BC_0 increased by a factor of two as the aethalometer filter progressed from pristine (ATN = 0) to heavily loaded (ATN = 150). In contrast, the ratio of absorption coefficient to BC is approximately constant and independent of ATN. This indicates that adjusted BC concentrations can be up to two times higher than unadjusted BC concentrations, assuming aethalometer measurements as the basis.

Our measurements of diesel truck exhaust at the Port of Oakland indicated $a = 0.66$, whereas BC emission factors reported by Dallmann et al.[3] were corrected with Kirchstetter and Novakov's value of $a = 0.88$.[2] This published value was derived from testing using laboratory-generated soot, while the current value is site-specific. We calculated BC emission factors (n =

1000) for each truck using both values and found that emission factors calculated with $a = 0.66$ were 15 ± 2% lower on average than those calculated using $a = 0.88$. Therefore, BC emission factors measured in the 2009 campaign[3] were multiplied by 0.85 (i.e., total correction with CO_2 = 1.07) before making comparisons to emission factors derived from the 2011 and 2013 field campaigns.

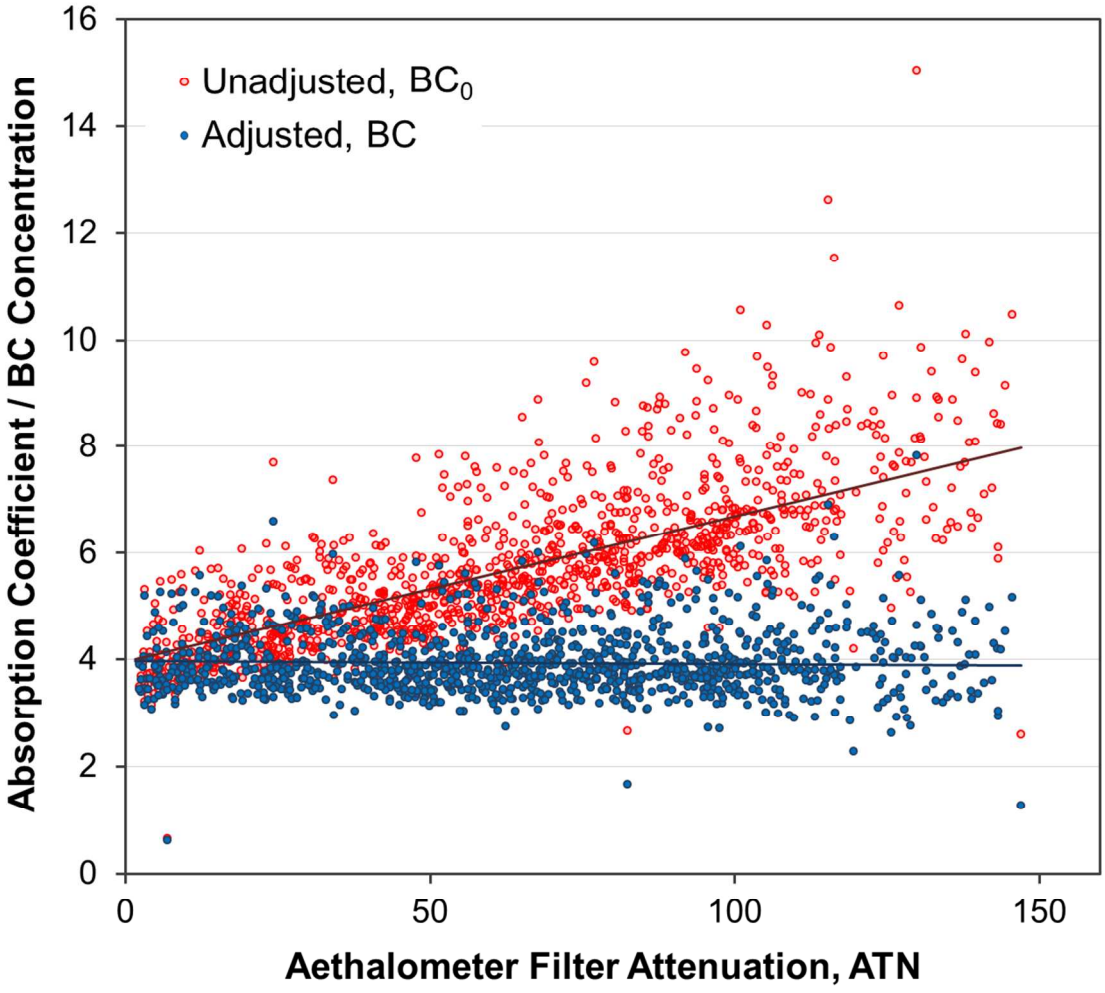

Figure S8. BC concentration measured using an aethalometer and absorption coefficient measured using a photoacoustic absorption spectrometer (PAS). Unadjusted BC concentrations (BC_0) depend on aethalometer filter loading (i.e., ATN) whereas the adjusted BC concentrations (BC) are independent of ATN.

RESULTS

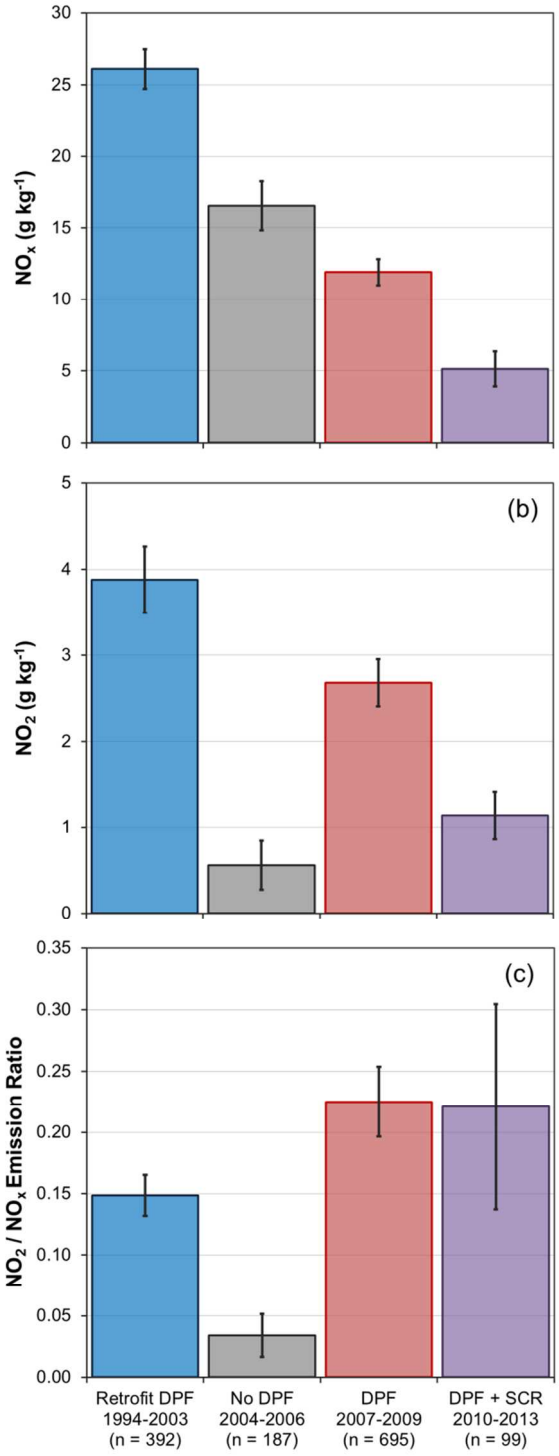

Figure S9. Average (a) NO_x emission factors, (b) NO_2 emission factors, and (c) NO_2/NO_x emission ratios by emission control category, based on combined 2011 and 2013 data. The range of engine model years for each category is indicated and the number of analyzed trucks is noted parenthetically on the category axis. Error bars reflect 95% confidence intervals about the mean.

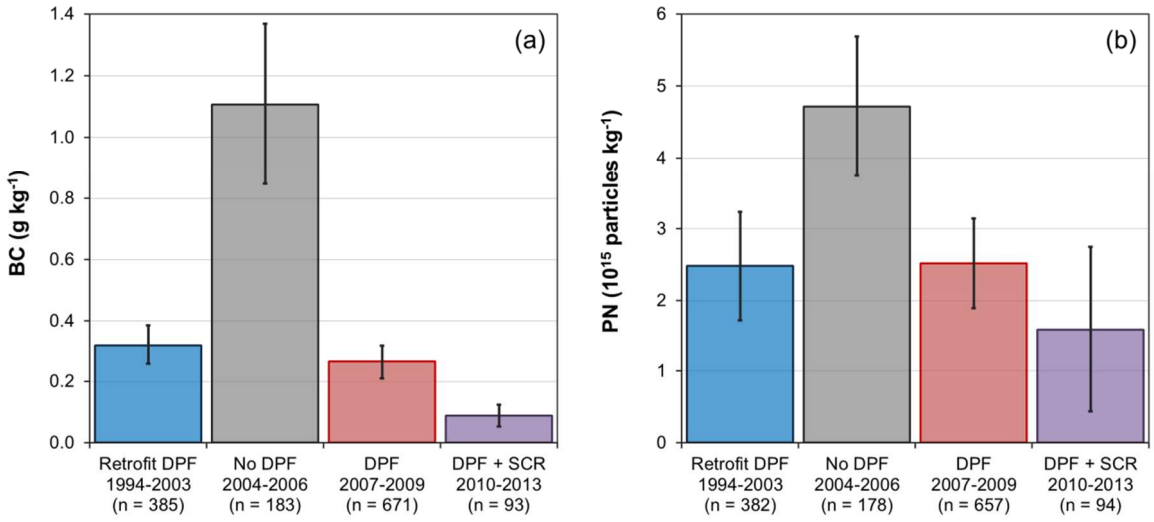

Figure S10. Average (a) BC emission factors and (b) PN emission factors by emission control category, based on combined 2011 and 2013 data. The number of analyzed trucks is indicated in parentheses on the category axis and the error bars reflect 95% confidence intervals about the mean.

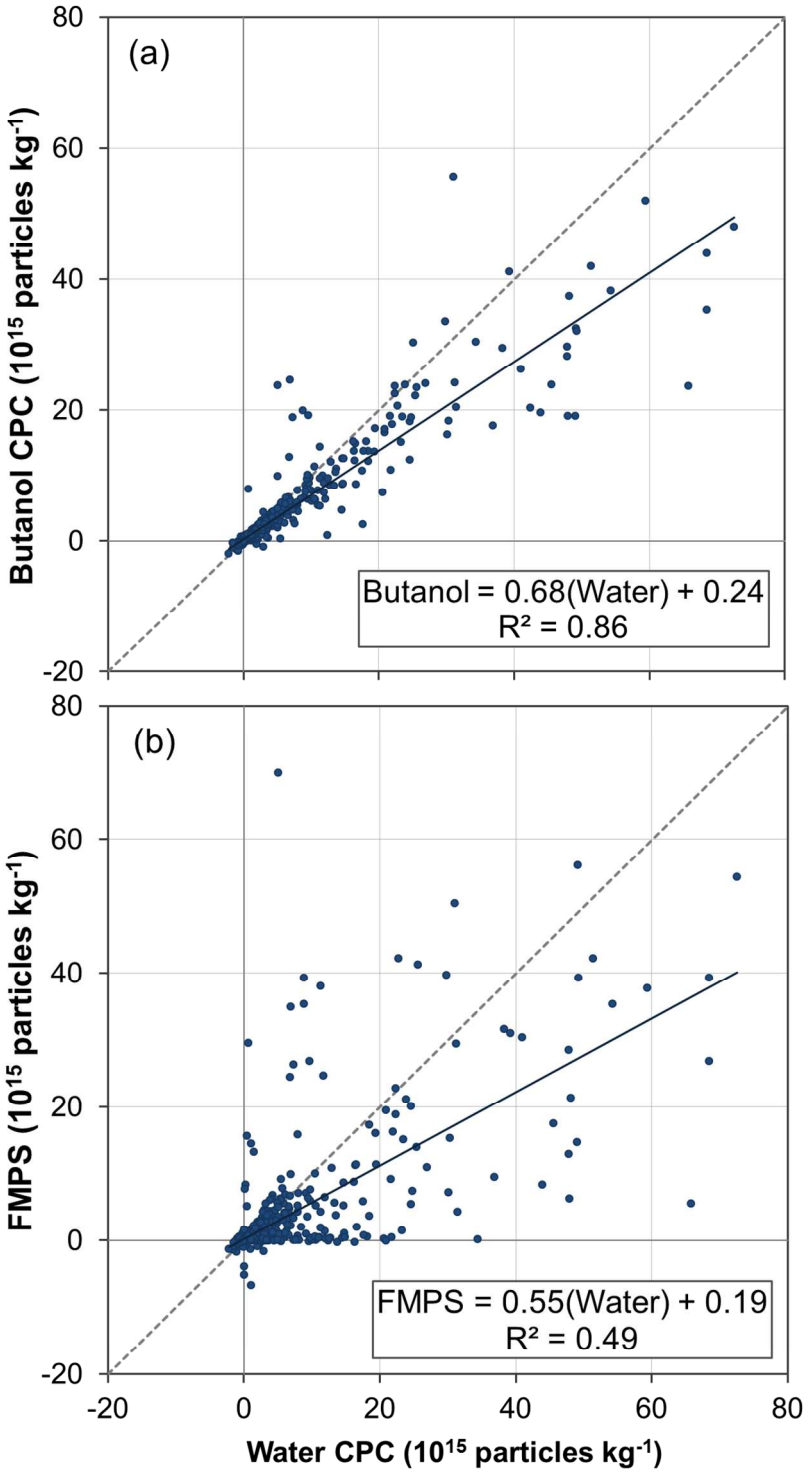

Figure S11. Comparison of PN emission factors determined from the ultrafine water- and butanol-based CPCs and the FMPS. The dashed line is the 1:1 diagonal; linear best fit lines and regression statistics are also shown for each plot.

Table S3. Summary of particle number (PN) emission factors for drayage trucks operating at the Port of Oakland in 2011 and 2013 measured using an ultrafine water-based CPC, an ultrafine butanol-based CPC, and a FMPS. Emission factor units are 10^{15} particles emitted per kg of diesel fuel burned. Uncertainty bounds provide 95% confidence intervals about the mean. The results for the four truck categories are based on the combined data from 2011 and 2013. See Table S2 for more detailed descriptions of the individual CPCs.

Fleet or Truck Category	Number of Trucks[a]	(10^{15} particles kg^{-1})		
		Water CPC	Butanol CPC	FMPS
2011 Fleet (54% DPF, 2% SCR)	359 - 376	3.59 ± 0.81	2.54 ± 0.51	2.91 ± 0.85
2013 Fleet (99% DPF, 9% SCR)	934 - 967	2.47 ± 0.48	1.97 ± 0.37	1.34 ± 0.33
Retrofit DPF (1994-2006)	382 - 390	2.61 ± 0.76	1.79 ± 0.50	1.00 ± 0.41
No DPF (2004-2006)	178 - 187	4.72 ± 0.97	3.87 ± 0.80	4.29 ± 1.19
DPF (2007-2009)	648 - 665	2.52 ± 0.62	2.01 ± 0.47	1.69 ± 0.53
DPF + SCR (2010-2013)	93 - 94	1.59 ± 1.15	1.05 ± 0.60	0.71 ± 0.54

[a]The number of trucks used for each fleet or truck category analysis depended on the data available from each instrument.

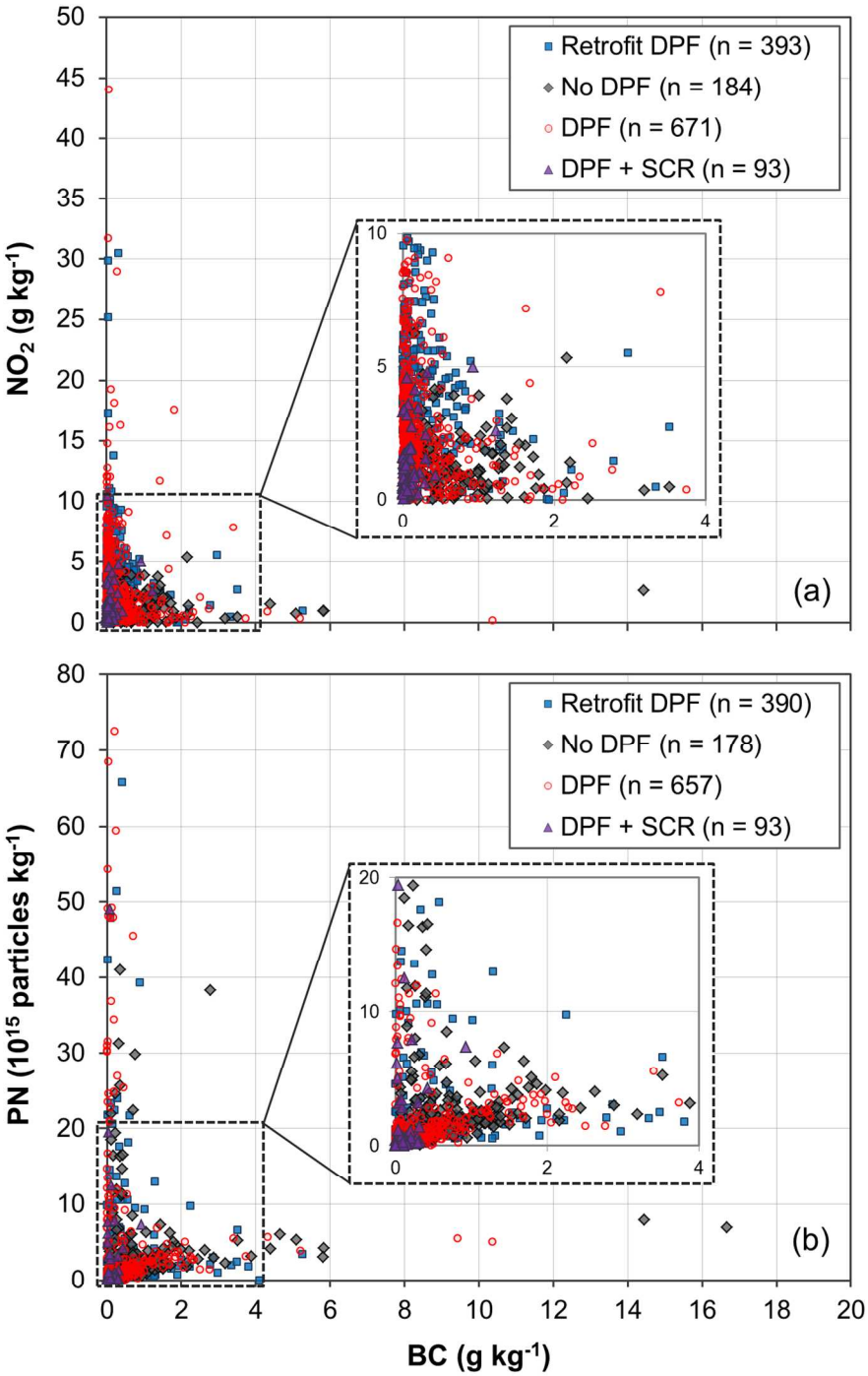

Figure S12. Relationship between emissions of BC to emissions of (a) NO_2 and (b) PN by truck category. The highest emitters of BC tend to have low emissions of PN and NO_2, and vice versa, across engine model years and installed control technologies.

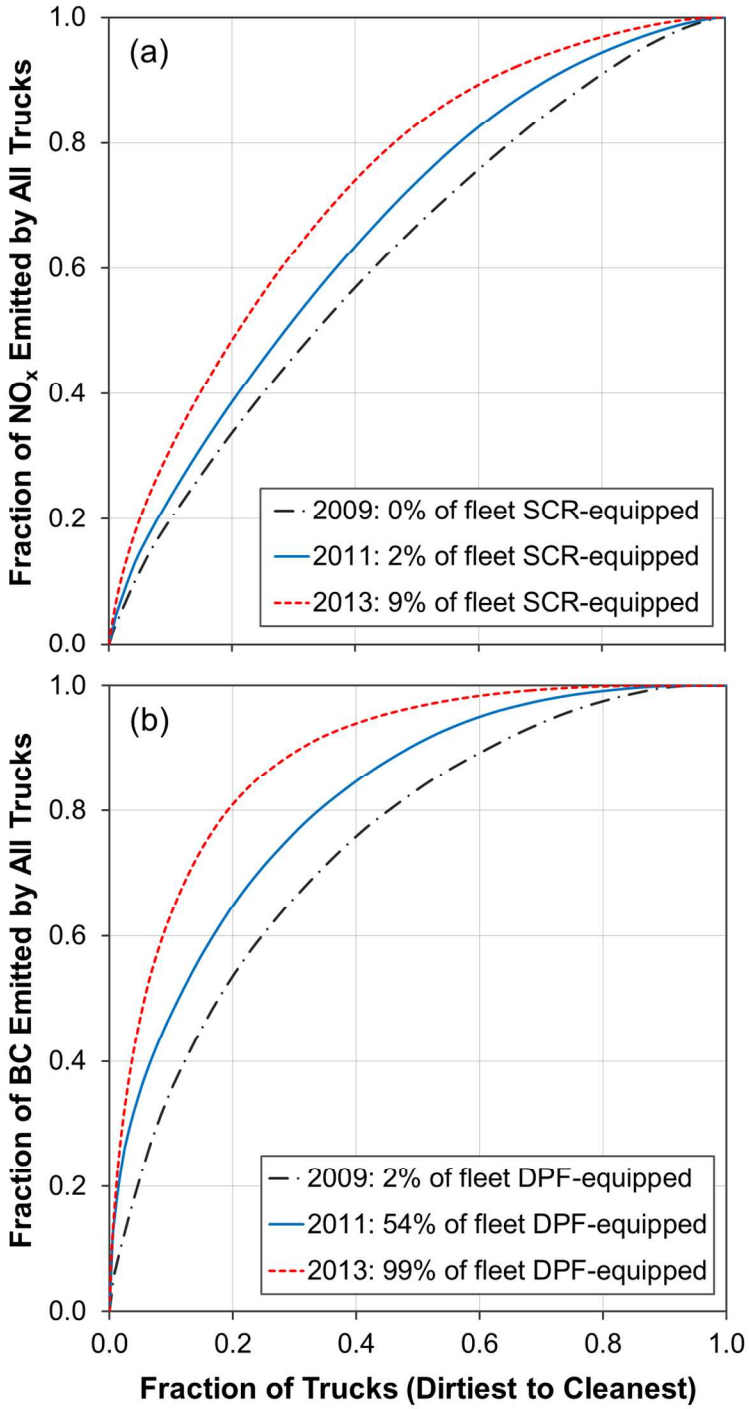

Figure S13. Cumulative emissions for each fleet with trucks ranked from highest to lowest for (a) NO_x and (b) BC emissions over time.

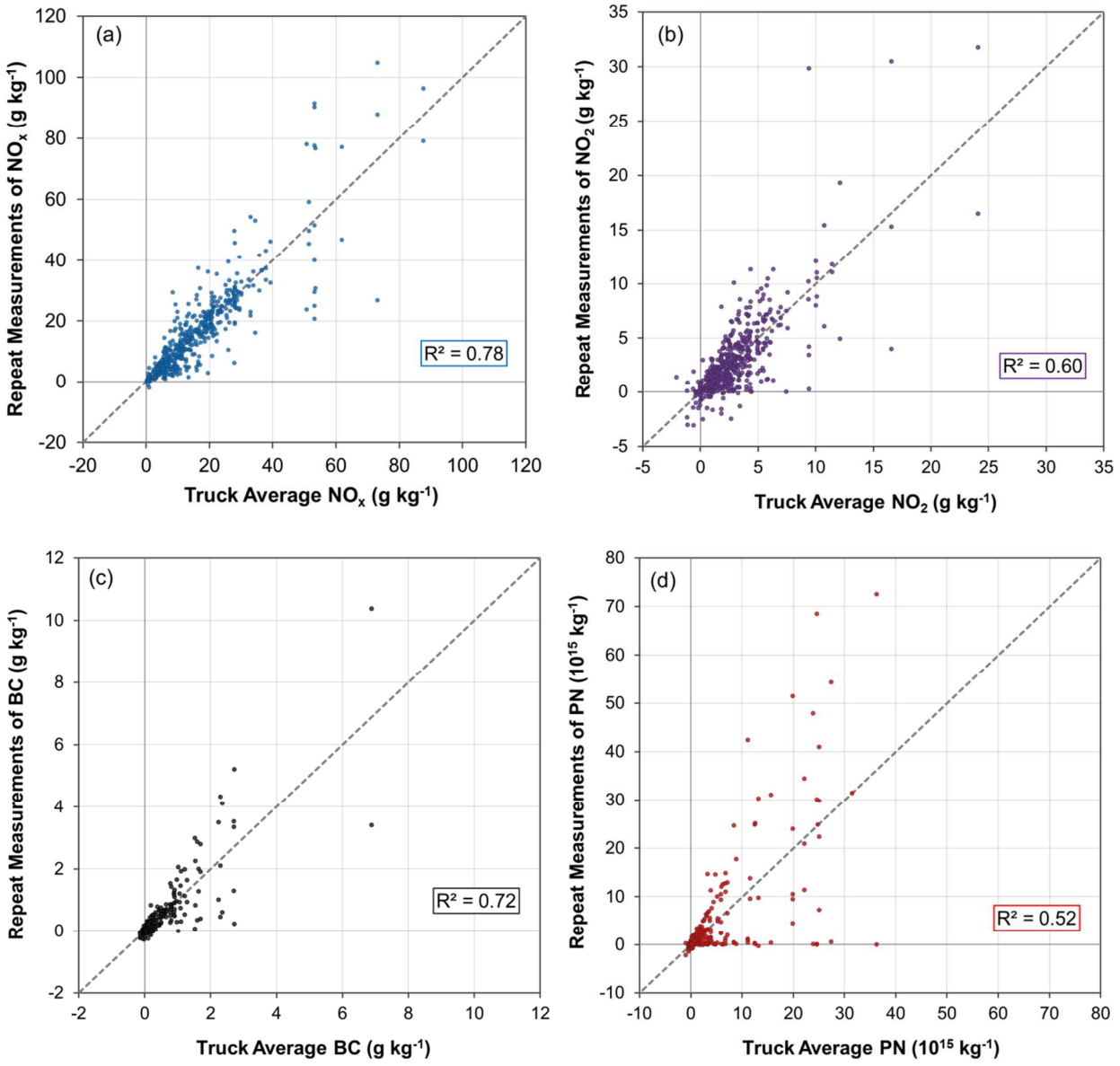

Figure S14. Results of repeat measurements for 207 individual trucks; each individual measurement is plotted on the vertical axis against the corresponding average emission factor for each truck on the horizontal axis. Repeatability of the measurement is indicated by the degree of scatter of data points about the dashed 1:1 line.

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