This is an open access article published under an ACS AuthorChoice License, which permits copying and redistribution of the article or any adaptations for non-commercial purposes.

Environmental Science & Technology

Article

pubs.acs.org/est

High-Mileage Light-Duty Fleet Vehicle Emissions: Their Potentially Overlooked Importance

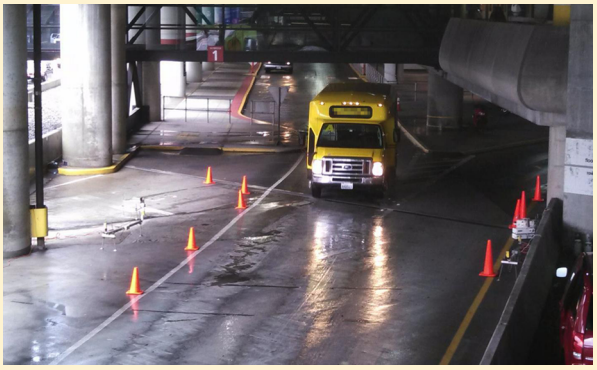
Gary A. Bishop,*[,†] Donald H. Stedman,[†] Daniel A. Burgard,[‡] and Oscar Atkinson[‡]

[†]Department of Chemistry and Biochemistry, University of Denver, Denver, Colorado 80208, United States

[‡]Department of Chemistry, University of Puget Sound, Tacoma, Washington 98416, United States

Supporting Information

ABSTRACT: State and local agencies in the United States use activity-based computer models to estimate mobile source emissions for inventories. These models generally assume that vehicle activity levels are uniform across all of the vehicle emission level classifications using the same age-adjusted travel fractions. Recent fuel-specific emission measurements from the SeaTac Airport, Los Angeles, and multi-year measurements in the Chicago area suggest that some high-mileage fleets are responsible for a disproportionate share of the fleet's emissions. Hybrid taxis at the airport show large increases in carbon monoxide, hydrocarbon, and oxide of nitrogen emissions in their fourth year when compared to similar vehicles from the general population. Ammonia emissions from the airport shuttle vans indicate that catalyst reduction capability begins to wane after 5−6 years, 3 times faster than is observed in the general population, indicating accelerated aging. In Chicago, the observed, on-road taxi fleet also had significantly higher emissions and an emissions share that was more than double their fleet representation. When compounded by their expected higher than average mileage accumulation, we estimate that these small fleets (<1% of total) may be overlooked as a significant emission source (>2−5% of fleet emissions).



INTRODUCTION

State and local vehicle emission control measures in the United States depend upon the preparation of accurate mobile source emission inventories for evaluating effective emission reduction alternatives. Most inventory calculations rely upon a computer model to estimate the fleet's emissions. In California, EMFAC is the model required for on-road emissions, and in the rest of the U.S., it is the United States Environmental Protection Agency's (U.S. EPA) MOVES model.[1,2] Both models in general calculate a mass of emissions emitted through the combination of fleet size, fleet average emissions, vehicle speeds (EMFAC), and fleet average mileage activity. Fleet size and activity are linked with age (model year), while mean emissions generally involve age, initial emission certification levels, and emission level classifications (low to high). Both of these models use different approaches to accomplish the end result, but in both models, vehicle activity levels are most often uniformly applied across all of the vehicle emission level classifications, with the underlying assumption that all vehicles, including high emitters, have the same age-adjusted travel fractions.

Modern vehicle fleet emissions are dominated by a relatively small number of high emitting vehicles, resulting in a skewed emission distribution, which is best described by a gamma distribution.[3] High emitters occur as a result of breakage, mal-maintenance (performance modifications, rechipping, and/or after-treatment removal), and at end of a vehicle's useful life. For example, recent on-road emission measurements in California and other locations have shown that the 99th percentile is responsible for more than a third of the carbon monoxide (CO) and hydrocarbon (HC) emissions and more than 15% of the nitric oxide (NO) emissions on a fuel-specific basis, and these imbalances continue to grow each year.[4,5] These percent contributions assume that the fuel consumption is proportional to the fleet fraction sampled. However, these percentages could increase or decrease if the vehicles represented in the 99th percentile consume significantly less or more fuel than the fleet average, potentially adding an additional compounding skewed distribution to the calculation.

One segment of the on-road fleet that is believed to drive more miles and consume more fuel than the average fleet vehicle is livery (for-hire) vehicles, such as taxis and limousines. A recent news article about a New York City taxi company highlighted the fact that their taxis routinely accumulate 100 000 miles/year operating 7 days a week on two 10 h shifts each day.[6] With such a rapid accumulation of mileage, the question from an air quality perspective is how well these vehicles maintain their new vehicle emission levels and, when

Received: February 10, 2016
Revised: April 18, 2016
Accepted: April 19, 2016
Published: May 3, 2016

 © 2016 American Chemical Society

DOI: 10.1021/acs.est.6b00717
Environ. Sci. Technol. 2016, 50, 5405−5411

Table 1. Fleet Sampling Specifics by Location with Measured Emission Means and Standard Errors of the Mean

location dates sampled	attempts/plates/matched mean model year	mean (g/kg) of fuel emissions and standard errors of the mean[a] CO	HC[b]	$NO^c/NO_2{}^d/NO_x$	NH_3	mean speed (mph) acceleration (mph/s) VSP[e] (kW/tonne)
Chicago, IL 9/8−9/13, 2014	26824/20638/20395 2007.5	9.4 ± 0.8[a]	1.3 ± 0.2	1.5 ± 0.1/−0.04 ± 0.02/2.2 ± 0.1	0.71 ± 0.02	24.0
						0.2
						4.8
Los Angeles, CA 4/27−5/4, 2013	33807/27808/27184 2004.7	16.4 ± 0.6	2.2 ± 0.2	2.1 ± 0.1/0.15 ± 0.02/3.4 ± 0.1	0.58 ± 0.02	21.9
						−0.2
						4.6
SeaTac Int. Airport 9/28−9/29, 2013	3037/1913/1816 2008.0	26.3 ± 0.4	6.5 ± 0.3	7.9 ± 0.2/0.24 ± 0.01/12.3 ± 0.2	0.90 ± 0.02	15.6
						1.2
						7.7

[a]Calculated using a carbon mass fraction of 0.86 for gasoline and diesel, 0.82 for propane, and 0.75 for natural gas. [b]HC grams expressed using a NDIR correction factor of 2 and normalized as described. [c]Grams of NO. [d]Grams of NO_2. [e]Vehicle specific power.

problems arise, how promptly are the vehicles repaired. The Georgia Institute of Technology and the U.S. EPA collected on-road emission measurements of CO and HC from taxi cabs operating at the Atlanta and New York airports in 1993 and 1994.[7] Their results showed that the taxi fleets of both cities had higher average emissions than a sample of typical on-road vehicles. Atlanta taxis were 3 and 36 times higher and New York taxis were 1.5 and 14 times higher for CO and HC, respectively. A similar measurement campaign at the Los Angeles airport in 1992 found similar elevated emissions, and a further investigation exposed an illegal Smog Check certificate generation operation that was successfully prosecuted by the Los Angeles District Attorney's office.[8]

Livery fleets have changed significantly in the past decade, with many city fleets now featuring large numbers of hybrid and alternatively fueled vehicles. There has been little recent research into the emission durability of these new fleets, and in this paper, we examine some recent and historical emission data from the Seattle and Chicago area to obtain a picture of how current livery vehicle emissions compare to the rest of the fleet and examine their importance to fleet emissions.

EXPERIMENTAL SECTION

This study uses data collected at three U.S. sampling sites (1) in Washington State on the taxi and shuttle loop at the Seattle−Tacoma International Airport (SeaTac, 0° grade), (2) a long-term light-duty site in California located on I-10 between Santa Monica and downtown Los Angeles, CA (WLA, SB La Brea Avenue to EB I-10, 2° grade), and (3) a long-term light-duty site in Illinois located in the northwest suburbs of Chicago (Algonquin Road to SB I-290/SH53, 1° grade), where we have collected emission data since 1997. The SeaTac data were collected on Sept 28 and 29, 2013, as a component of one of the author's senior thesis; the west LA data were collected over 7 days from April 27 to May 4, 2013; and the 2014 Chicago data were collected over parts of six days from Sept 8−13, 2014. The Chicago historical data sets (1997−2000, 2002, 2004, and 2006) have been previously discussed in the literature, and all of these data sets are available for download from our website at www.feat.biochem.du.edu.[9−11]

Two identical remote vehicle exhaust sensors, developed by the University of Denver, named Fuel Efficiency Automobile Test (FEAT), were used to collect the emission measurements (serial 3002 in Chicago and LA and serial 3005 in Seattle). The basic instrument is composed of a source and detector unit aligned across a single-lane road. Collinear beams of infrared (IR) and ultraviolet (UV) light are passed across the roadway into the IR detection unit and are then focused through a dichroic beam splitter that separates the beam into its IR and UV components. The IR detector consists of four non-dispersive infrared (NDIR) detectors for a reference channel (3.9 μm), carbon monoxide (CO, 3.6 μm), carbon dioxide (CO_2, 4.3 μm), and hydrocarbons (HC, 3.3 μm). The UV beam is focused onto a quartz fiber that divides the light between two fiber bundles and directs the incoming light to two dispersive UV spectrometers. One spectrometer measures nitric oxide (NO), sulfur dioxide (SO_2), and ammonia (NH_3) between 195 and 225 nm. The second records nitrogen dioxide (NO_2) spectra between 430 and 450 nm. All of the detectors sample at 100 Hz and have been described in detail in the literature.[12−14]

The detector and source are installed on the shoulders of a single-lane roadway with a beam height of approximately 30−35 cm. When a vehicle blocks the beam, as indicated by a loss of signal on the IR reference channel, the system waits until the beam is reestablished and then collects 0.5 s of data from each IR detector and each spectrometer channel. IR voltages and UV spectra are converted into "concentrations" using an assumed plume path length of 8 cm, which was used initially in the laboratory to generate the detector species response curves. Because the real path length of the plume on-road is unknown, FEAT measures vehicle exhaust gases as a molar ratio to exhaust CO_2. Each species data points collected during the 0.5 s sample are plotted against the CO_2 data points, and a least-squares best fit slope (i.e., CO/CO_2, HC/CO_2, NO/CO_2, etc.) is determined. These ratios are constant for a given exhaust plume. The measured ratio of each species is scaled by its certified gas cylinder ratios, which are measured multiple times each day at each location by FEAT to correct for variations in instrument sensitivity and in ambient CO_2 levels caused by atmospheric pressure, temperature, and ambient pollution differences. Three similar specification calibration cylinders were used at each site containing (a) 6% CO, 0.6% propane, 6% CO_2, and 0.3% NO, balance nitrogen; (b) 0.05% NO_2 and 15% CO_2, balance air; and (c) 0.1% NH_3 and 0.6% propane, balance nitrogen (certified accuracies of ±2%, Air Liquide, Longmont, CO, and Praxair, Tacoma, WA).

FEAT has been demonstrated in double-blind intercomparisons to be accurate to ±5% for CO and ±15% for HC for an individual vehicle measurement.[15,16] The NO spectrometer has

DOI: 10.1021/acs.est.6b00717
Environ. Sci. Technol. 2016, 50, 5405−5411

been determined separately to have a minimum detection limit (3σ) of 24 ppm for NO using a new low-emitting test vehicle.[13] A comparison of fleet average emission by model year versus IM240 fleet average emissions by model year shows excellent correlations, with R^2 values between 0.75 and 0.98, for data from Denver, Phoenix, and Chicago.[17]

A video image of the license plate of each vehicle is recorded, and the transcribed plate is used to obtain non-personal vehicle information, including make, model year, vehicle identification number, and fuel type from the state registration records of California, Illinois, and Washington. Speed and acceleration measurements for each vehicle are attempted using a pair of parallel infrared beams (Banner Engineering) 1.8 m apart and approximately 0.66 m above the roadway.

For this analysis, the measured ratios have been converted into fuel-specific emissions of grams of pollutant per kilogram of fuel by carbon balance using a carbon mass fraction for the fuel of 0.86 and doubling the HC/CO_2 ratio to account for the poor quantification of certain hydrocarbon species by NDIR absorption.[12,18] Uncertainties presented in the tables and graphs are standard errors of the mean calculated from the distribution of the daily measurement means of each site. Because of only 2 days of data collection in Seattle, uncertainties were estimated using a bootstrap resampling technique, which was used to generate multi-day means from which standard errors of the mean could be calculated (see the Supporting Information).

RESULTS AND DISCUSSION

Table 1 contains a summary of the sampling dates, vehicle record information, fleet averaged model year, mean fuel-specific emission measurements with their standard errors of the mean for all of the records, and driving mode parameters collected at each site. Vehicle-specific power has been calculated using the equation from Jimenez et al.[19] HC means have been normalized to the lowest HC emitting subfleet at each site for comparison by eliminating site-specific systematic offsets that arise from either liquid water interferences (Chicago, 2014) or instrument-created offsets (see the Supporting Information).

SeaTac Airport Fleet. The SeaTac International Airport has a vehicle service fleet composed of many advanced fuel vehicles, which are not commonly found in high enough numbers in the general fleet to study. Table S2 of the Supporting Information lists the number of measurements (1816) and number of unique vehicles (524) by fuel type for the SeaTac database. Because of the nature of their business, there are many repeat measurements on the same vehicle. The measurements were collected on the flat covered taxi and shuttle bus loop road exit that parallels the terminal. Only taxis, limousines, and shuttle buses are permitted on this road. The shuttle buses were constantly circulating, stopping only long enough to pick up or drop off passengers. Taxi driving modes are less predictable, with longer stops a distinct possibility, although each taxi had to have driven more than a quarter of a mile before reaching our measurement location. Measurements were collected between 10:30 and 19:00 h over the 2 days with temperatures between 12 and 17 °C.

Slightly more than a quarter of the measurements (506; see Table S2 of the Supporting Information) were collected on Toyota Prius models, whose fleet fractions have increased at the airport as a result of a relaxed 2006 city rule, which allowed gasoline hybrid vehicles to substitute for natural-gas-fueled vehicles for taxi services at the airport.[20] The airport taxis are operated by a single company, and we collected measurements on about 60% of its Prius fleet. Figure 1 plots the mean fuel-specific emissions by model year for CO (top), HC (middle), and NO_x (bottom) for the Prius taxis measured at SeaTac (●) and Prius vehicles measured a few months earlier at the WLA location (△). Prius measurements make up approximately 2.3% of the 2013 WLA database (619/27247 with 509 unique vehicles), but only 24 measurements are from taxis. Concerns about whether the airport taxis have engines and catalytic converters at operating temperature are allayed by the fact that, if our airport measurements had a large fraction of cold-start vehicles, we would expect NO_x emissions to be depressed in all model years and CO and HC emissions to be similarly increased when compared to the WLA site, where there is little possibility for cold-start vehicles. Figure 1 shows that CO and HC emissions are statistically similar for the two data sets for the first 4 model years, after which the SeaTac Prius emission values rise rapidly. NO_x emissions for the airport taxis are higher during the first 4 model years than the WLA vehicles but also rise to higher levels starting with the 2009 models. The SeaTac Prius NH_3 emissions also show an increase (see Figure S1 of the Supporting Information) after the first 4 model years. The drop in all species emissions with the 2006 SeaTac models may or may not be representative because there were only four measurements from two vehicles collected for that model year.

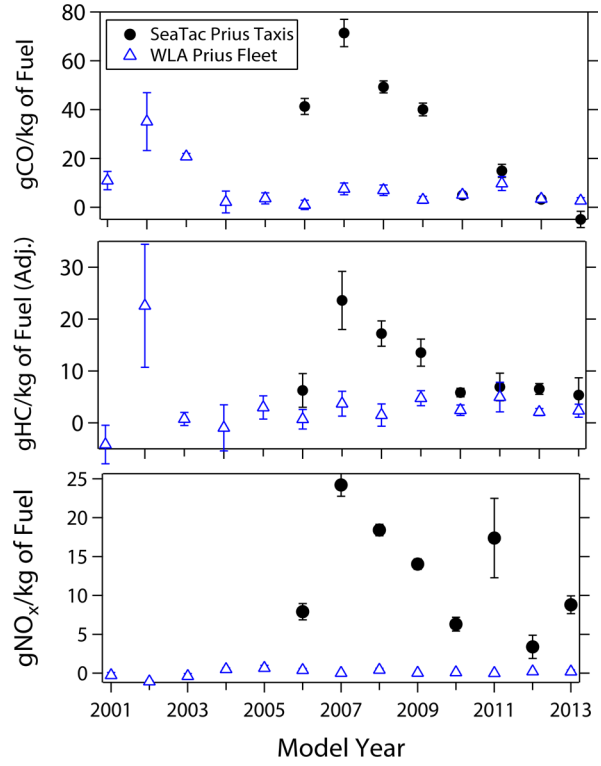


Figure 1. SeaTac Toyota Prius taxis mean grams of pollutant per kilogram of fuel emissions (●) by model year compared to Toyota Prius model emissions measured on-road at the west Los Angeles measurement site (△). Uncertainties are standard error of the means determined from the daily samples for the west LA data and by bootstrap resampling techniques for the SeaTac measurements.

These observed emission differences between the Prius vehicles measured at SeaTac and in WLA are likely connected with how the two fleets are used and maintained. At the time of

DOI: 10.1021/acs.est.6b00717
Environ. Sci. Technol. 2016, 50, 5405−5411

these measurements, emission inspections were not required in Seattle nor California for any Prius models; however, Seattle taxis are restricted to being only 7 years old or newer, and beginning in the spring of 2015, California brought 7 year and older Prius models into their onboard diagnostic-only testing program.[21] One major difference between the two fleets is that we expect the rate at which mileage is accumulated on the airport taxis to be significantly higher than the largely public owned WLA vehicles. Anecdotal data from talking with some of the SeaTac taxi drivers was that 200 miles/day was common. This would quickly put 50 000 miles or more on a taxi in a year's time, which is about 4 times the national average mileage accumulation rate of 13 500 miles/year.[22]

One way to gauge mileage accumulation rates is through its negative effects on three-way catalyst activity levels. It is well-known that catalyst age and mileage accumulation is linked and catalysts deteriorate with age.[23,24] We can gauge three-way catalyst activity levels for non-diesel-fueled vehicles by their ability to catalyze the reduction reaction of NO to NH_3. We have previously reported that fleet NH_3 emissions rise with vehicle age until the catalyst begin to lose their ability to reduce NO, at which point NH_3 emissions decline back to the low levels observed in vehicles not equipped with three-way catalytic converters.[4,10,25,26] In 2013 at the WLA site, the NH_3 emission turnover point occurred between 16 and 19 year old vehicles. Figure 2 plots grams of NH_3 per kilogram of fuel emissions versus model year for the gasoline (●) and propane (△) fueled SeaTac shuttle buses. NH_3 emissions for the shuttle buses peak between 5 and 6 year old vehicles and then begin to decline, indicating that, on average, the catalysts of these vehicles begin to lose their reduction capability about 3−4 times sooner than the on-road WLA fleet. This is another indication that these vehicles accumulate mileage at a higher rate than the typical on-road fleet and that exhaust after-treatment components degrade as a result.

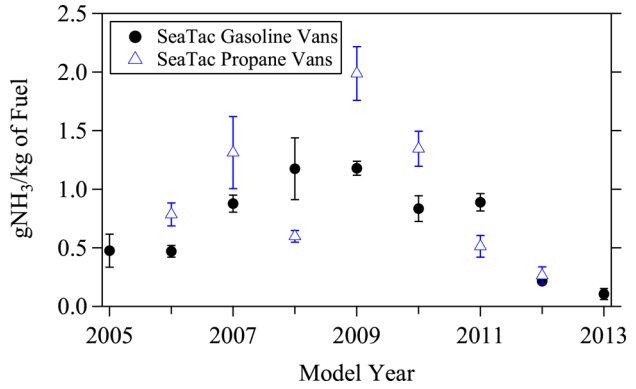


Figure 2. Grams of NH_3 per kilogram of fuel emissions plotted versus model year for gasoline (●) and propane (△) fueled shuttle vans measured at SeaTac International Airport. Uncertainties plotted are standard errors of the mean determined using bootstrap resampling techniques.

Chicago Fleet. At our long-term suburban data collection site northwest of Chicago, we have collected emission data sets in the years 1997−2000, 2002, 2004, 2006, and most recently in 2014.[11] In Illinois, traditional taxi and limousines have dedicated license plate formats (taxi plates end in TX, and limousine plates end in LY) that allow them to be easily distinguished from other vehicles in the databases. Using this information, we analyzed and compared the emissions of the taxi and limousine fleets to the general fleet emission distributions and patterns for each measurement year. Table 2 provides a summary of the number of measurements for each fleet and mean model year by measurement year and shows that the taxi fleet of this site is consistently older than the on-road fleet, while the limousine segment is consistently younger. Figures 3 and 4 plot fuel-specific emissions for CO, HC, and NO emissions for the taxi and limousine fleets and age-adjusted fleet emissions for the data set of each year. Measurements of NO_2 emissions are only available for the 2014 data set; therefore, we have chosen to make all of the comparisons using NO. Because age is the most significant determining factor in fleet emission means, it is important that we eliminate any age differences before making the comparison between the Chicago sites on-road fleet and the taxi and limousine fleets. This is accomplished by using the age distribution of taxi or limousine fleets and applying it to the emission distribution for the on-road fleet, producing an on-road fleet emission mean that now has the same age as the taxi or limousine fleet. This process is detailed in Table S3 of the Supporting Information.

Mean taxi emissions at our northwest Chicago site are consistently higher (Figure 3) than for the similarly age-adjusted on-road fleet for all three pollutants. The one exception is for the HC emissions in the 1998 data set, which is also abnormally low for the limousines. The process of correcting for fleet zero emission offsets in each year's data set (see the Supporting Information) has overcorrected for the 1998 taxi and limousine subfleets. The higher emissions indicate that the taxis have a higher proportion of vehicles in need of emission-related maintenance than the general fleet.

Table 2. Chicago Taxi and Limousine Vehicle Fleet Statistics, Fleet Percentages, and Percent Emission Contribution

			percent emission contribution taxis/limo (multiple of fleet percentage)		
year	measurements of fleet/taxis/limo mean model year (fleet/taxis/limo)	fleet percentages of taxis/limo	CO	HC	NO
2014	20395/268/183 (2007.5/2007.3/2011.3)	1.3/0.9	3.2/0.6 (2.4/0.7)	3.1/0.6 (2.4/0.6)	2.4/0.3 (1.8/0.4)
2006	22200/341/106 (2001/2000/2002.8)	1.5/0.5	2.7/0.5 (1.8/1)	3.2/0.6 (2.1/1.3)	2.4/0.3 (1.6/0.3)
2004	21838/141/70 (1999.2/1997.7/2000.9)	0.6/0.3	1.2/0.5 (1.9/1.4)	1.6/0.4 (2.5/1.3)	1.5/0.4 (2.3/1.2)
2002	22320/160/104 (1997.4/1994.9/1999)	0.7/0.5	1.8/0.6 (2.5/1.2)	2.1/0.5 (2.9/1.1)	1.8/0.6 (2.5/1.3)
2000	22065/154/106 (1995.5/1993.1/1997.1)	0.7/0.5	1.6/0.5 (2.3/1.0)	1.7/0.3 (2.4/0.6)	1.4/0.5 (2.0/1.0)
1999	23088/146/182 (1994.6/1992.6/1996.4)	0.6/0.8	1.8/0.9 (2.8/1.1)	2.0/0.5 (3.2/0.6)	1.2/1.0 (1.9/1.2)
1998	23560/84/151 (1993.6/1992.5/1995.4)	0.4/0.6	0.6/0.5 (1.8/0.8)	0.1/0 (0.2/0)	0.5/0.7 (1.4/1.1)
1997	19682/132/132 (1992.7/1991.1/1994.2)	0.7/0.7	1.5/0.7 (2.2/1.0)	2.4/0.4 (2.4/0.6)	0.9/0.9 (1.8/1.3)
vehicle weighted means		1.0/0.6	2.1/0.6 (2.1/1.0)	2.4/0.4 (2.4/0.6)	1.8/0.6 (1.8/1.0)

DOI: 10.1021/acs.est.6b00717
Environ. Sci. Technol. 2016, 50, 5405−5411

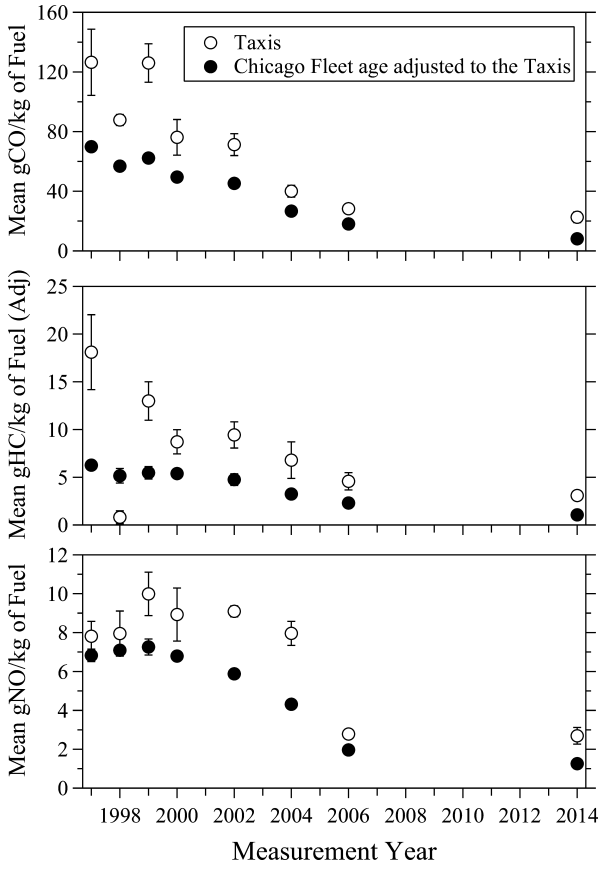


Figure 3. Chicago historical mean grams of pollutant per kilogram of fuel emissions by measurement year for taxis (○) and the entire Chicago fleet that has been age-corrected to match the age of the taxi subfleet (●) for each data set collected at this site. Uncertainties plotted are standard error of the means determined using the daily samples.

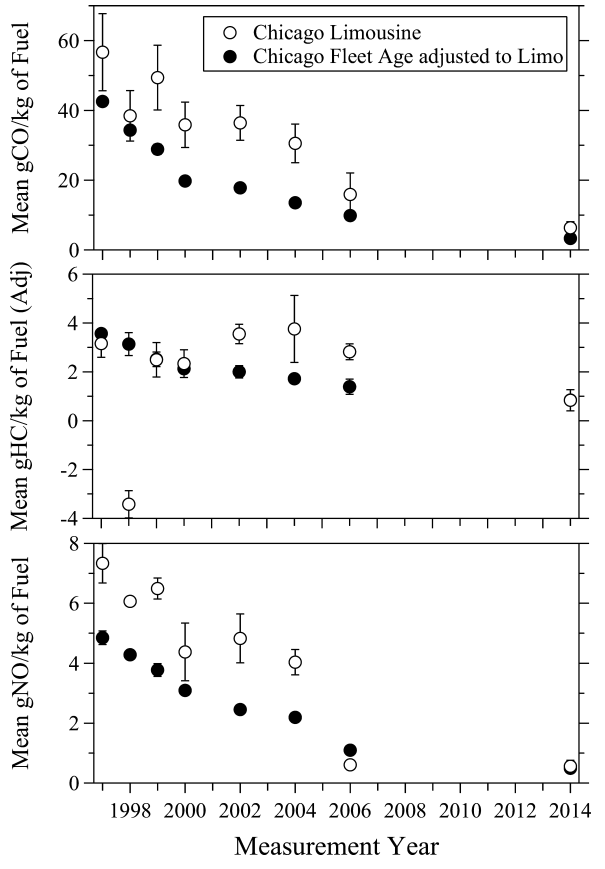


Figure 4. Chicago historical mean grams of pollutant per kilogram of fuel emissions by measurement year for limousines (○) and the entire Chicago fleet that has been age-corrected to match the age of the limousine subfleet (●) for each data set collected at this site. Uncertainties plotted are standard error of the means determined using the daily samples.

The good news is that taxi emissions have decreased along with the general fleet, with large reductions in emissions over the 17 year span of measurements. The historical limousine emission comparison (Figure 4) shows that emissions have also declined over the 17 year period and that CO and NO emissions are not statistically different from the general fleet in 2006 and for all three species in 2014.

Table 2 lists the number of measurements and fleet percentage representation for the taxis and limousines for each measurement year and the percentage of the total fleet emissions that the taxi and limousine fleets contributed. This is not an age-normalized analysis but a check of whether these Chicago subfleets (taxi and limousine) emission contributions are proportional to their fleet representation. For example, the 2014 taxi fleet is responsible for 3.2, 3.1, and 2.4% of the fleet CO, HC, and NO emissions, respectively, even though they only comprise 1.3% of the measured fleet. These percent contributions assume that, on average, the taxis consume the same amount of fuel as the rest of the fleet. If, as we expect, the taxis use 2−4 times more fuel in their travels, then the emission contribution percentages increase by these factors. Overall, the taxi fleet is responsible for, on average, 2.1, 2.4, and 1.8 times more emissions for CO, HC, and NO, respectively, over the past 17 years than would be expected from their fleet percentages, indicating a disproportionate emissions share for the taxis. The limousine fleet over this same time period has averages of 1, 0.6, and 1 for CO, HC, and NO, respectively, indicating that the limousine emissions are generally inline or a little lower than their numerical representation.

The differences between emission contributions are also born out when we compare the two 2014 subfleet emission distributions with the 2014 on-road fleet. Figure 5 is a percentile−percentile plot of the taxi fleet's fuel-specific emissions for CO, HC, and NO compared to the on-road fleet. The diagonal line is the 1:1 line showing where the points would fall if the emission distributions of the two fleets were identical. For both the CO and NO emissions, the taxi fleet starts to have higher emissions than the on-road fleet at very low percentiles. For HC, the 2014 taxi fleet has generally slightly higher emissions across all percentiles until the 99th percentile, where the deviation is more pronounced. The taxis have significantly higher 99th percentile emissions, a percentile that we consider to be fully populated with broken vehicles for all three pollutants. These differences are not likely just the result of age because the 2014 taxi fleet is only 0.2 of a model year older than the on-road Chicago fleet, indicating that other factors are involved in the emission differences seen. A similar plot (Figure S2 of the Supporting Information) compares the 2014 limousine subfleet to the on-road fleet and shows that, for CO and HC, the emission distribution follows the 1:1 line, while NO emissions are significantly lower, which may be a result of the age differences (limousine fleet is 3.8 years

DOI: 10.1021/acs.est.6b00717
Environ. Sci. Technol. 2016, 50, 5405−5411

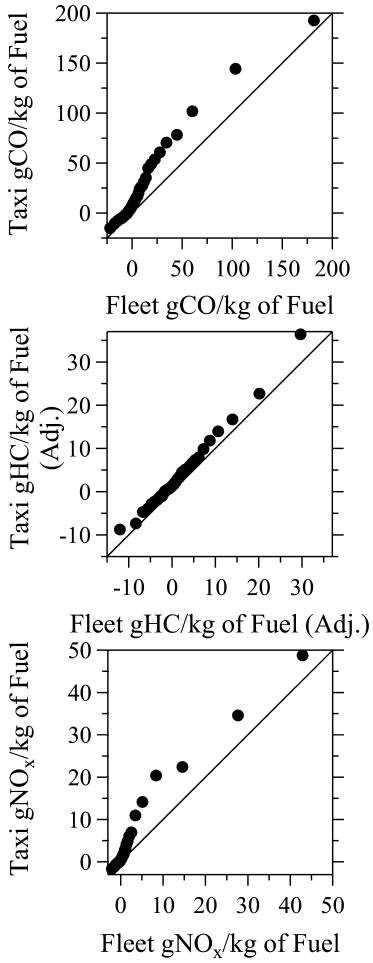


Figure 5. Percentile−percentile plots of the 2014 emission distribution comparison of the Chicago site taxi fleet versus the on-road fleet of fuel-specific emissions for CO (top), HC (middle), and NO (bottom). Lines in each plot are 1:1 lines drawn to indicate a perfect match between the two emission distributions.

younger than the on-road fleet and 4 years younger than the taxi fleet) between these two groups.

Even when corrected for age differences, the 2014 taxi fleet still has significantly higher emissions for CO, HC, and NO than the 2014 limousine fleet. Table S4 of the Supporting Information details the percentage of makes and mean emissions for the taxi and limousine fleets for the 2014 Chicago data set. The limousines are dominated by Lincolns (115/183) and Cadillacs (28/183), with these two groups representing 78% of all of the measurements. These two makes have significantly lower emissions than most of the taxi makes and are the major reason that the limousines have lower overall emissions. The taxi fleet is more diverse, with the two largest makes represented being Toyota (86/268, of which 31 are Prius models) and Dodge (65/268), accounting for 56% of the taxis measured. The emission differences likely come down to the differences in maintenance practices between the two fleets, but without specific information to that effect, we cannot speculate.

The 4 year old Toyota Prius taxis measured at SeaTac showed a pattern of increasing emissions (see Figure 1) when compared to similarly aged models measured in Los Angeles. To check for similar emissions behavior for the Chicago taxis, Figure S3 of the Supporting Information plots the grams of CO, HC, and NO_x emissions per kilogram of fuel by model year for the 2014 Chicago area taxis (all makes and models) and limousine fleets and compares them against the 2014 Chicago on-road fleet. The 2014 taxi (268 measurements) and limousine (183 measurements) subfleets measured in Chicago have fewer measurements, which leads to larger uncertainties in the fuel-specific emission values when broken out by model year. While the 2014 Chicago area taxi fleet in general has higher emissions than the general fleet and the 2014 limousine fleet has similar or lower emissions as previously discussed, there is no clear change in emission levels with increasing age, as observed with the SeaTac airport fleet.

Fleet emission inventory calculations generally rely on some type of age-weighted activity factor, usually model year, for generating the totals. High-mileage vehicles, such as taxis and limousines, are not generally considered to be numerous enough to change these weighting factors, because, for example, in King County, Washington, taxis only account for about 0.6% of the registered vehicles (2000/325 000). If, however, as observed in Chicago, their emission contribution is about twice their fleet percentage and, in addition, they consume more fuel than the county average, then their emission contribution could grow to be 2−5% of the total. In areas where these high-mileage fleets are in higher concentrations, such as airports and downtown business centers, their emission contribution could produce much larger inventory errors if not taken into account. As tailpipe emissions continue to decrease, emission distributions will become more skewed, likely increasing the importance of higher emission high-mileage fleets and their contribution to vehicle emission inventories.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b00717.

Supplementary tables (Tables S1−S4) and figures (Figures S1−S3) referenced in the text (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*Telephone: 303-871-2584. E-mail: gbishop@du.edu.

Notes

The authors declare the following competing financial interest(s): The authors Gary A. Bishop and Donald H. Stedman acknowledge receipt from the University of Denver of a share of the patent royalty payments from Envirotest, an operating subsidiary of Opus Inspection, which licenses the commercial remote sensing technology developed at the University of Denver. Commercial instrumentation was not used in this study.

■ ACKNOWLEDGMENTS

This paper is a result of work supported by the California Environmental Protection Agency Air Resources Board (12-303), the Coordinating Research Council (E-106), the University of Puget Sound, and the University of Denver. Results and conclusions presented here are solely the responsibility of the authors and may not represent the views of the sponsors. The authors gratefully acknowledge Annette Bishop (license plate transcription).

DOI: 10.1021/acs.est.6b00717
Environ. Sci. Technol. 2016, 50, 5405−5411

REFERENCES

(1) Air Resources Board, California Environmental Protection Agency. *EMFAC Emissions Database*; http://www.arb.ca.gov/emfac/ (accessed Feb 2016).

(2) United States Environmental Protection Agency. *Modeling and Inventories; MOVES (Motor Vehicle Emission Simulator)*; http://www.epa.gov/otaq/models/moves/ (accessed Feb 2016).

(3) Zhang, Y.; Bishop, G. A.; Stedman, D. H. Automobile emissions are statistically gamma distributed. *Environ. Sci. Technol.* **1994**, *28*, 1370−1374.

(4) Bishop, G. A.; Schuchmann, B. G.; Stedman, D. H.; Lawson, D. R. Multispecies remote sensing measurements of vehicle emissions on Sherman Way in Van Nuys, California. *J. Air Waste Manage. Assoc.* **2012**, *62* (10), 1127−1133.

(5) Bishop, G. A.; Stedman, D. H. *On-Road Remote Sensing of Automobile Emissions in the Tulsa Area: Fall 2013*; Coordinating Research Council: Alpharetta, GA, 2014; http://www.feat.biochem.du.edu/assets/databases/Oklahoma/CRC-106%20Tulsa%202013%20Final%20report.pdf (accessed Feb 2016).

(6) Berk, B. Taxi tough: A look inside an NYC cab shop. *Car Driver* Feb 2011; http://www.caranddriver.com/features/taxi-tough-a-look-inside-an-nyc-cab-shop-feature.

(7) Vescio, N.; DuBose, R. S.; Rodgers, M. O. Comparison of taxi fleet compositions and emissions behavior in Atlanta, Georgia, and New York City. *Proceedings of the 5th Coordinating Research Council's On-Road Vehicle Emission Workshop*; San Diego, CA, April 3−5, 1995.

(8) Churchill, M. Auto smog program needs repairs. *Press Enterprise* July 26, **1992**.

(9) Bishop, G. A.; Stedman, D. H. Reactive Nitrogen Species Emission Trends in Three Light-/Medium-Duty United States Fleets. *Environ. Sci. Technol.* **2015**, *49* (18), 11234−11240.

(10) Bishop, G. A.; Stedman, D. H. The recession of 2008 and it impact on light-duty vehicle emissions in three western US cities. *Environ. Sci. Technol.* **2014**, *48*, 14822−14827.

(11) Bishop, G. A.; Stedman, D. H. A decade of on-road emissions measurements. *Environ. Sci. Technol.* **2008**, *42* (5), 1651−1656.

(12) Burgard, D. A.; Bishop, G. A.; Stadtmuller, R. S.; Dalton, T. R.; Stedman, D. H. Spectroscopy applied to on-road mobile source emissions. *Appl. Spectrosc.* **2006**, *60*, 135−148.

(13) Popp, P. J.; Bishop, G. A.; Stedman, D. H. Development of a high-speed ultraviolet spectrometer for remote sensing of mobile source nitric oxide emissions. *J. Air Waste Manage. Assoc.* **1999**, *49*, 1463−1468.

(14) Burgard, D. A.; Dalton, T. R.; Bishop, G. A.; Starkey, J. R.; Stedman, D. H. Nitrogen dioxide, sulfur dioxide, and ammonia detector for remote sensing of vehicle emissions. *Rev. Sci. Instrum.* **2006**, *77*, 014101.

(15) Lawson, D. R.; Groblicki, P. J.; Stedman, D. H.; Bishop, G. A.; Guenther, P. L. Emissions from in-use motor vehicles in Los Angeles: A pilot study of remote sensing and the inspection and maintenance program. *J. Air Waste Manage. Assoc.* **1990**, *40*, 1096−1105.

(16) Ashbaugh, L. L.; Lawson, D. R.; Bishop, G. A.; Guenther, P. L.; Stedman, D. H.; Stephens, R. D.; Groblicki, P. J.; Johnson, B. J.; Huang, S. C. On-road remote sensing of carbon monoxide and hydrocarbon emissions during several vehicle operating conditions. *Proceedings of the A&WMA International Specialty Conference on PM10 Standards and Non-traditional Source Control*; Phoenix, AZ, Jan 12−15, 1992.

(17) Pokharel, S. S.; Stedman, D. H.; Bishop, G. A. RSD Versus IM240 Fleet Average Correlations. *Proceedings of the 10th CRC On-Road Vehicle Emissions Workshop*; San Diego, CA, March 27−29, 2000; http://www.feat.biochem.du.edu/assets/presentations/RSD%20verus%20IM240%20Correlations%2010th_CRC_2000.pdf (accessed Feb 2016).

(18) Singer, B. C.; Harley, R. A.; Littlejohn, D.; Ho, J.; Vo, T. Scaling of infrared remote sensor hydrocarbon measurements for motor vehicle emission inventory calculations. *Environ. Sci. Technol.* **1998**, *32*, 3241−3248.

(19) Jimenez, J. L.; McClintock, P.; McRae, G. J.; Nelson, D. D.; Zahniser, M. S. Vehicle specific power: A useful parameter for remote sensing and emission studies. *Proceedings of the Ninth CRC On-Road Vehicle Emissions Workshop*; Coordinating Research Council: Alpharetta, GA, 1999; pp 7-45−7-57.

(20) Murakami, K. First hybrid taxis humming at Sea-Tac. *Seattle Post-Intelligencer* Sept 13, 2007; http://www.seattlepi.com/local/article/First-hybrid-taxis-humming-at-Sea-Tac-1249560.php (accessed Feb 2016).

(21) California Bureau of Automotive Repair. *Q&As: Hybrid Vehicles in the Smog Check Program*; http://www.bar.ca.gov/Industry/Industry_Resources/Q&As_Hybrid_Vehicles_in_the_Smog_Check_Program.html#Question1 (accessed April 2016).

(22) Federal Highway Administration, United States Department of Transportation. *Average Annual Miles Per Driver by Age Group*; https://www.fhwa.dot.gov/ohim/onh00/bar8.htm (accessed April 2016).

(23) López Granados, M.; Larese, C.; Cabello Galisteo, F.; Mariscal, R.; Fierro, J. L. G.; Fernández-Ruíz, R.; Sanguino, R.; Luna, M. Effect of mileage on the deactivation of vehicle-aged three-way catalysts. *Catal. Today* **2005**, *107−108*, 77−85.

(24) Winkler, A.; Ferri, D.; Hauert, R. Influence of aging effects on the conversion efficiency of automotive exhaust gas catalysts. *Catal. Today* **2010**, *155* (155), 140−146.

(25) Bishop, G. A.; Peddle, A. M.; Stedman, D. H.; Zhan, T. On-road emission measurements of reactive nitrogen compounds from three California cities. *Environ. Sci. Technol.* **2010**, *44*, 3616−3620.

(26) Burgard, D. A.; Bishop, G. A.; Stedman, D. H. Remote sensing of ammonia and sulfur dioxide from on-road light duty vehicles. *Environ. Sci. Technol.* **2006**, *40*, 7018−7022.

DOI: 10.1021/acs.est.6b00717
Environ. Sci. Technol. 2016, 50, 5405−5411

Supporting Information For:

High-mileage Light-duty Vehicle Emissions: Their Potentially Overlooked Importance

Gary A. Bishop* and Donald H. Stedman

Department of Chemistry and Biochemistry MSC 9020, University of Denver, Denver CO 80208

Daniel A. Burgard and Oscar Atkinson

Department of Chemistry, University of Puget Sound, Tacoma, WA 98416

*Corresponding author email: gbishop@du.edu; phone: (303) 871-2584

Summary of Supporting Information:

11 Pages (excluding cover):

Tables

S1. HC Normalization Mode Calculation.

S2. SeaTac International Airport Vehicle Measurements by Fuel Type.

S3. 2014 Chicago Fleet Age Normalization Example for CO Emission Comparison with the Taxi Sub-Fleet.

S4. 2014 Chicago Limousine and Taxi Measurements by Make.

Figures

S1. SeaTac Toyota Prius taxis mean grams of NH_3 per kilogram of fuel emissions by model year (●) compared with Toyota Prius models emissions measured on-road at the West Los Angeles measurement site (Δ). Uncertainties are standard error of the means determined from the daily samples for the West LA data and by bootstrap resampling techniques for the SeaTac measurements.

S2. Percentile-Percentile plots of the 2014 emissions distribution comparison of the Chicago site's Limousine fleet versus the on-road fleet of fuel specific emissions for CO (top), HC (middle) and NO (bottom). Lines in each plot are 1:1 lines drawn to indicate a perfect match between the two emission distributions.

S3. Grams of pollutant per kilogram of fuel emissions versus model year for the Chicago 2014 data for the on-road fleet (●), taxis (Δ) and limousines (◆). Uncertainties are standard error of the means determined from the daily samples.

How we estimate standard errors of the mean for our reported uncertainties

Because vehicle emissions from US vehicle fleets are not normally distributed the assigning of uncertainties on fleet emission means involves a process that many readers may not be familiar with. Standard statistical methods that were developed for normally distributed populations when used on a skewed distribution results in uncertainties that are unrealistically too small due to the large number of samples. The Central Limit Theorem in general says that the means of multiple samples, randomly collected, from a larger parent population will be normally distributed, irrespective of the parent populations underlying distribution. Since we almost always collect multiple days of emission measurements from each site, we use these daily measurements as our randomly collected multiple samples from the larger population and report uncertainties based on their distribution. We calculate means, standard deviations and finally standard errors of the mean for this group of daily measurements. We report the means for all of the emission measurements and then calculate a standard error of the mean for the entire sample by applying the same error percentage obtained from the ratio of the standard error of the mean for the daily measurements divided by the daily measurement mean. An example of this process is provided below for the 2014 Chicago gCO/kg of fuel and gNO/kg of fuel measurements. While this example is for a fleet mean we also use this technique when we report standard errors of the mean for individual model years or specific fuel or technology types. For example each model will have its daily mean calculated and then its standard error of the mean for the daily average computed and that percent uncertainty will be applied to that model year's fleet emissions mean.

Chicago 2014

Date	Mean gCO/kg of fuel	Counts	Mean gNO/kg of fuel	Counts
9/8/14	9.56	4126	1.36	4125
9/9/14	10.61	4568	1.34	4568
9/10/14	12.25	2710	1.22	2710
9/11/14	6.74	4151	1.64	4151
9/12/14	8.78	1753	1.70	1753
9/13/14	8.85	3087	1.81	3087
Daily Mean	9.46		1.51	
Standard Error for the daily means	0.76		0.1	
Fleet Mean	9.40		1.49	
Standard Error for the fleet means	0.76		0.1	
As reported in Table 1	9.4 ± 0.8		1.5 ± 0.1	

Because only two days of data were collected at the Seattle-Tacoma Airport we have to use a different method to estimate the uncertainties. We use a technique called bootstrap resampling to take multiple random samples (usually five for the general number of days in a week that we collected data) with replacement from all the measurements we collected at the SeaTac Airport. We usually make these samples to be half the size of the number of measurements that we have so for the overall averages from Seattle we selected about 900 readings. We generate five random samples and calculate the mean for each of those samples and then calculate the standard error of the mean for those five samples and apply the same error percentage to the fleet mean to estimate its standard error of the mean. Below is a sample of that process for gCO/kg of fuel. For the uncertainties on model year emissions we repeat this process using only the measurements for that specific model year and or vehicle type.

SeaTac Airport data

Sample #	Mean gCO/kg of fuel	Counts
1	27.018	908
2	27.874	908
3	27.147	908
4	25.239	908
5	26.773	908

Pseudo-Daily Mean	26.81
Standard Error for the pseudo-daily means	0.43
Fleet Mean	26.25
Standard Error for the fleet means	0.42
As reported in Table 1	26.3 ± 0.4

How we normalize mean gHC/kilogram of fuel emissions for comparisons with other data sets.

The hydrocarbon channel on FEAT has the lowest signal to noise ratio of all the measurement channels in large part because the absorption signals are the smallest (millivolt levels). FEAT uses one detector for the target gas absorption and a second detector for the background IR intensity (reference). These channels are ratioed to each other to correct for changes in background IR intensities that are not the result of gas absorption. These two detectors responses are not perfectly twinned and for the low signal HC channel these intensity corrections can result in systematic offsets (positive or negative) for zero measurements on the HC channel. In addition the region of infrared absorption that we use for HC measurements is overlapped by an absorption band for liquid water. Normally this is not an issue as fully warmed up vehicles emit little if any liquid water at the tailpipe. However, there are times when low temperatures and high dew points can cause water vapor to condense at the tailpipe which will create an increased absorption that is not related to HC emissions. In these cases the normalization value calculated will be larger because it includes the adjustment for the liquid water emissions.

The offset is calculated by computing the mode and means of the newest model year vehicles, and assuming that these vehicles emit negligible levels of hydrocarbons, using the lowest of either of these values as the offset. We then add (for negative offsets) or subtract this value from all of the hydrocarbon measurements. Since it is assumed that the newest vehicles are the lowest emitting this approximation will slightly over correct because the true offset will be a value somewhat less than the average of the cleanest model year and make.

The Chicago 2014 measurement included a correction for both of the previously discussed issues as the first three days of measurements were with normal temperatures and low humidity while the last three days experienced the exact opposite. FEAT ratios are first reported as percent emissions and the normalization calculations are performed using these percent values. Below are the data tables used for estimating the HC normalization value for the 2014 Chicago measurements.

For the Monday through Wednesday time slot Honda's vehicles had the lowest average HC emissions with a mean %HC of 0.0013. In Table S2 the mode calculation has two values that are very close to each other 0.001 and 0.0015. We chose to average those two values and the HC normalization value for the first time period used was 0.00125% which is approximately 0.5 gHC/kg of fuel.

For the Thursday through Saturday time period Honda vehicles again had the lowest HC emission. The average of 2009 – 2014 Honda vehicles is 0.003% which is the same as the mode shown in Table S2. This is approximately 1.25 gHC/kg of fuel.

2014 Chicago Mode Calculations
For model year 2009 and newer vehicles

Table S1. HC Normalization Mode Calculation.

Monday – Wednesday		Thursday - Saturday	
%HC	Counts	%HC	Counts
-0.0015	129	-0.0015	73
-0.001	147	-0.001	59
-0.0005	138	-0.0005	75
0	125	0	67
0.0005	126	0.0005	79
0.001	152	0.001	69
0.0015	155	0.0015	75
0.002	143	0.002	85
0.0025	104	0.0025	51
0.003	131	0.003	94
0.0035	129	0.0035	68
0.004	120	0.004	77
0.0045	115	0.0045	80
0.005	124	0.005	88

This method will successfully normalize the fleet HC means but may over or under correct smaller sub-fleets.

Table S2. SeaTac International Airport Vehicle Measurements by Fuel Type.

Fuel Type	Vehicle Measurement Counts (%)	Unique Vehicles
Diesel	52 (2.9%)	9
Gasoline	896 (49.3%)	271
Natural Gas	95 (5.2%)	12
Propane	267 (14.7%)	49
Gasoline-Hybrid	506 (27.9%)	183
Totals	1816	524

Age normalizations are accomplished by adjusting the Chicago fleet means to exactly match the proportion of vehicles by model year in the sub-fleet (Taxis or Limousines). Below is how to calculate the age adjusted mean gCO/kg of fuel for the 2014 on-road Chicago fleet that matches the age of the 2014 Taxi fleet.

Table S3. 2014 Chicago Fleet Age Normalization Example for CO Emission Comparison with the Taxi Sub-Fleet.

2014 (Chicago Fleet as Measured)	Model Year	Mean gCO/kg	No. of Measurements	Total Emissions
	1999	30.878	417	12876.298
	2000	24.189	574	13884.236
	2001	16.903	616	10412.481
	2002	16.474	785	12931.730
	2003	12.996	940	12215.976
	2004	12.660	1129	14292.654
	2005	11.117	1237	13752.253
	2006	7.396	1312	9703.335
	2007	3.768	1423	5362.034
	2008	5.054	1476	7459.151
	2009	5.497	1102	6058.203
	2010	4.931	1226	6045.138
	2011	2.687	1610	4326.126
	2012	1.460	1654	2415.270
	2013	2.053	1843	3783.433
	2014	0.931	1832	1705.581
				137223.901
			Mean gCO/kg of Fuel	**7.2**
2014 (Taxis as Measured)	Model Year	Mean gCO/kg	No. of Measurements	Total Emissions
	1999	7.511	3	22.533
	2000	44.367	13	576.774
	2001	35.338	6	212.028
	2002	32.894	18	592.083
	2003	20.713	18	372.835
	2004	11.351	19	215.678
	2005	43.144	27	1164.884
	2006	57.129	25	1428.217
	2007	14.561	18	262.106
	2008	23.130	14	323.825
	2009	18.463	7	129.238
	2010	18.998	15	284.964
	2011	9.020	25	225.501
	2012	3.640	18	65.517
	2013	2.033	27	54.890
	2014	7.823	15	117.351
			268	6048.423
			Mean gCO/kg of Fuel	**22.6**
2014 (Chicago Fleet Age Adjusted to Match the Taxi Fleet Age)	Model Year	Mean gCO/kg	No. of Measurements	Total Emissions
	1999	30.878	3	92.635
	2000	24.189	13	314.451
	2001	16.903	6	101.420
	2002	16.474	18	296.524
	2003	12.996	18	233.923
	2004	12.660	19	240.532
	2005	11.117	27	300.170
	2006	7.396	25	184.896
	2007	3.768	18	67.826
	2008	5.054	14	70.751
	2009	5.497	7	38.482
	2010	4.931	15	73.962
	2011	2.687	25	67.176
	2012	1.460	18	26.285
	2013	2.053	27	55.427
	2014	0.931	15	13.965
			268	2178.425
			Mean gCO/kg of Fuel	**8.1**

Table S4. 2014 Chicago Limousine and Taxi Measurements by Make.

Make	Limousine Counts (%) Mean Model Year Mean gCO/kg / gHC/kg / gNO/kg	Taxi Counts (%) Mean Model Year Mean gCO/kg / gHC/kg / gNO/kg
Cadillac	28 (15.3%) 2012.7 1.9 / -1.4 / 0.08	
Chevrolet	16 (8.7%) 2012.3 28.9 / -1.4 / -0.06	15 (5.6%) 2005.9 20.2 / 1.1 / 4.1
Chrysler	5 (2.8%) 2012.4 -9.2 / 8.9 / 0.4	22 (8.2%) 2004. 6 37.7 / 2.7 / 4.6
Dodge		65 (24.3%) 2006.3 36.8 / 4.7 / 2.3
Ford	1 (0.5%) 2012 7.2 / 3.1 / 0.9	26 (9.7%) 2007.2 28.7 / 5.7 / 4.4
Honda		17 (6.3%) 2002.9 29.4 / 2.2 / 8.0
GMC	4 (2.2%) 2011 -20.0 / -6.6 / 2.1	
Infinity	1 (0.5%) 2014 1.5 / 5.9 / 0.2	
Lincoln	115 (62.8%) 2011 5.3 / 1.1 / 0.7	8 (3%) 2003.3 16.6 / 3.3 / 1.4
Mazda		4 (1.5%) 2004.25 26.5 / 1.5 / 0.5
Mercedes-Benz	8 (4.5%) 2012.3 9.0 / 4.7 / 0.3	
Mercury	4 (2.2%) 2007.8 14.9 / -0.5 / 0.6	1 (0.4%) 2002 18. 0 / 4.6 / 14.9
Mitsubishi		4 (1.5%) 2009 33.3 / 0.1 / -0.1
Nissan		13 (4.8%) 2009.8 6.8 / 0.6 / 1.6
Pontiac		5 (1.9%) 2001 1.6 / 0.5 / 0.8
Toyota	1 (0.5%) 2013 22.6 / 17.4 / 0.9	86 (32.1%) 2010.3 9.2 / 2.5 / 1.2 (31/86 Prius, 2010.7) 8.4 / 4.0 / 0.2
VPG		2 (0.7%) 2012 2.1 / -1.1 /
Totals	183 2011.3 6.3 / 0.8 / 0.6	268 2007.3 22.6 / 3.1 / 2.7

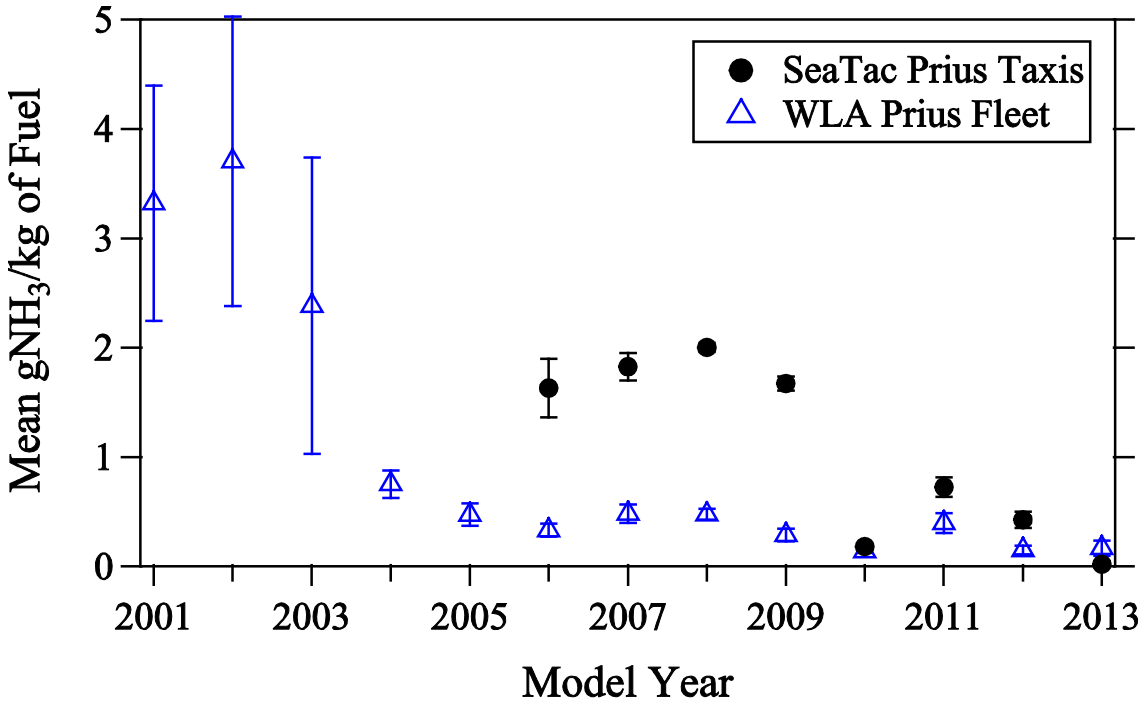


Figure S1. SeaTac Toyota Prius taxis mean grams of NH_3 per kilogram of fuel emissions by model year (●) compared with Toyota Prius model emissions measured on-road at the West Los Angeles measurement site (Δ). Uncertainties are standard error of the means determined from the daily samples for the West LA data and by bootstrap resampling techniques for the SeaTac measurements.

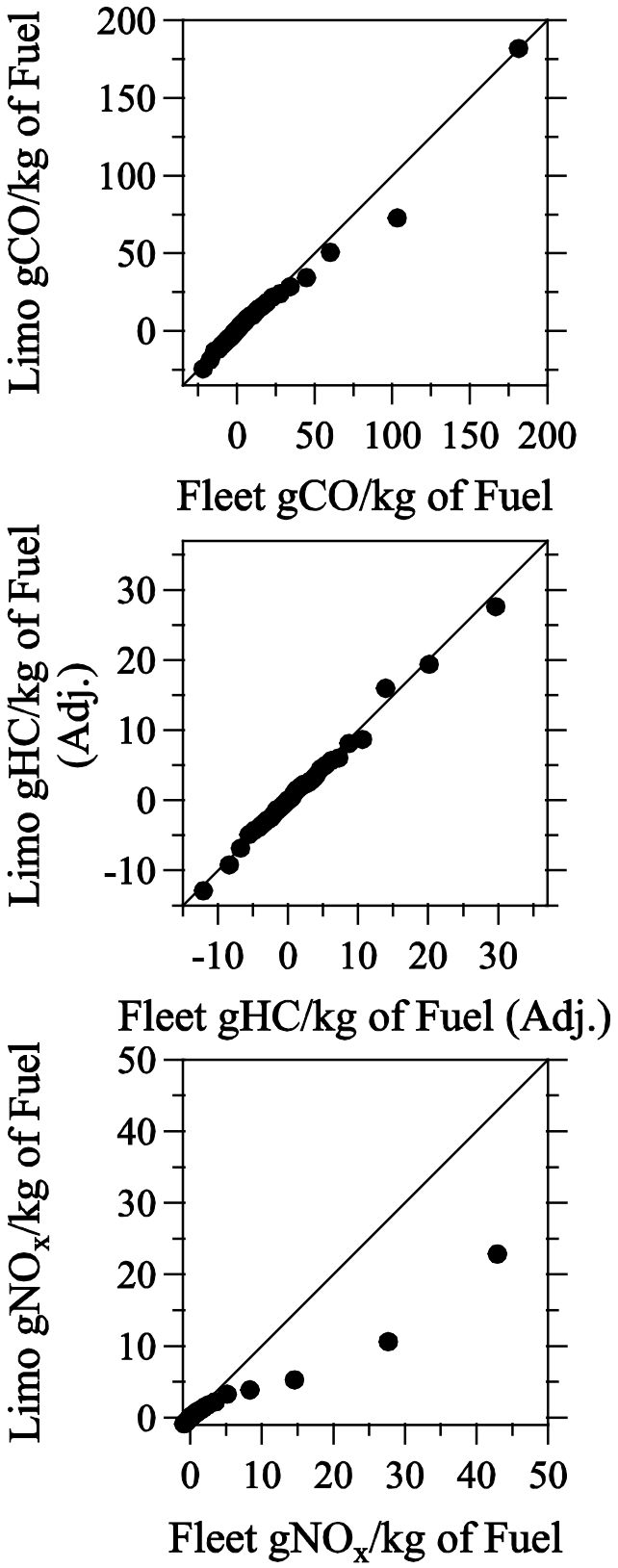


Figure S2. Percentile-Percentile plots of the 2014 emissions distribution comparison of the Chicago sites Limousine fleet versus the on-road fleet of fuel specific emissions for CO (top), HC (middle) and NO (bottom). Lines in each plot are 1:1 lines drawn to indicate a perfect match between the two emission distributions.

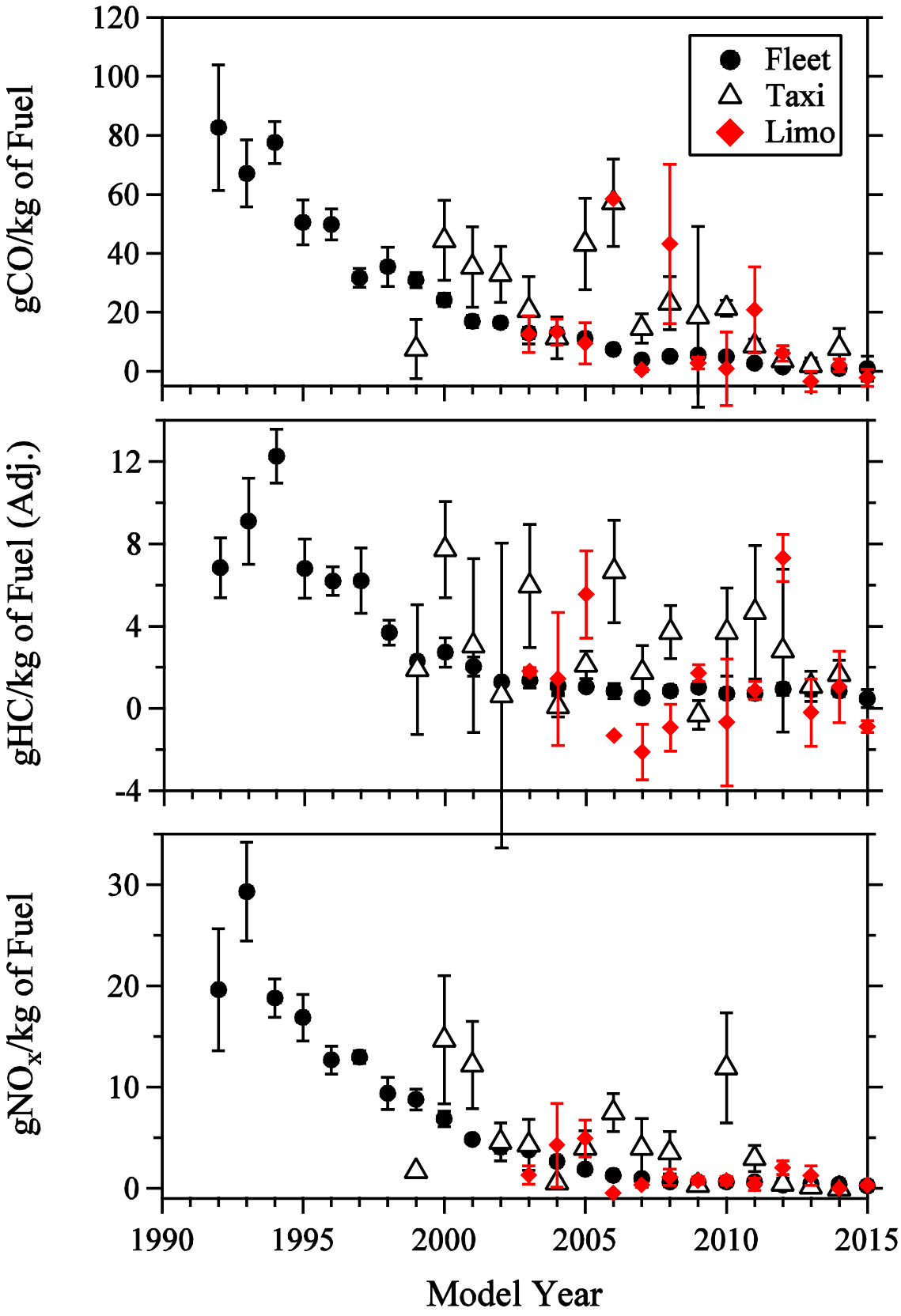


Figure S3. Grams of pollutant per kilogram of fuel emissions versus model year for the Chicago 2014 data for the on-road fleet (●), taxis (268 measurements, Δ) and limousines (183 measurements, ◆). Uncertainties are standard error of the means determined from the daily samples.