2021

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Recommended Citation

Bishop, G. A. (2021), Does California's EMFAC2017 Vehicle Emissions Model Under-predict California Light-duty Gasoline Vehicle NOx Emissions?, J. Air & Waste Manage. Assoc., 71:5, 597-606. DOI: 10.1080/10962247.2020.1869121

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Does California's EMFAC2017 Vehicle Emissions Model Under-predict California Light-duty Gasoline Vehicle NO_x Emissions?

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Abstract

On-road remote sensing measurements of light and medium-duty gasoline vehicles collected within California's South Coast Air Basin since 1999 generally fall within the range of observed summer ambient molar NO_x/CO measurements collected during morning rush hours. Compared with ambient and on-road emissions, the California Air Resources Board EMFAC model under predicts 2018 gasoline vehicle NO_x emission factors by more than a factor of 2.6. Contributing to these differences is that vehicles older than model year 2006 have NO_x emission deterioration rates that are up to 4 time's higher on-road than predicted by the EMFAC model. A fuel-based inventory using the 2018 on-road gasoline emission factors for CO and NO_x results in total CO emissions similar to the basin inventory but NO_x emissions that are 74% higher than the inventory. The higher NO_x emission estimates from on-road gasoline vehicle measurements makes their contribution to the inventory slightly larger than heavy-duty diesel vehicles. We have found LEV I (1994 - 2003) gasoline vehicles are a major source of these on-road emissions and that significant NO_x reductions in the South Coast Air Basin are being overlooked by not targeting the high emitters for removal.

Keywords

Emissions, Automotive; Emissions, Oxides of Nitrogen; EMFAC2017

Introduction

With the introduction of the National Ambient Air Quality Standards (NAAQS) in the early 1970's as part of the Clean Air Act the United States began establishing networks of ambient air monitors in urban areas across the country.(U. S. Environmental Protection Agency)The NAAQS were intended to limit common pollutants found in outdoor air that were considered to be harmful to public health. The species to be monitored were carbon monoxide (CO), lead, nitrogen dioxide (NO_2), particulate matter, ozone and sulfur dioxide. California's South Coast Air Basin (SoCAB), which includes Los Angeles, has over the years experienced elevated levels of most of these species but through tremendous reductions in mobile and stationary source emissions over the last 60 years now only exceeds levels for particulate matter and ozone.(Warneke et al., 2012; Pollack et al., 2013; United States Environmental Protection Agency, 2020b, a)

Since Haagen-Smit first documented the presence of ozone in Los Angeles air, reducing it to healthful levels has proven to be a difficult task.(Haagen-Smit et al., 1953; Haagen-Smit and Fox, 1954) Unlike most of the criteria pollutants, ozone is not directly emitted but is a secondary product formed in a nonlinear reaction involving volatile organic compounds (VOC), carbon monoxide (CO) and oxides of nitrogen ($NO_x \equiv NO + NO_2$) in sunlight.(Finlayson-Pitts and Pitts, 2000; Stedman, 2004) Despite significant reductions in ozone levels in the SoCAB, with the 2015 revisions of the NAAQS 8 hour ozone rule that lowered the standard to 70ppb (fourth-highest daily maximum, averaged across three consecutive years), compliance is not expected for many decades.(Fujita et al., 2013; U. S. Environmental Protection Agency, 2015; Parrish et al., 2017)

Because of the interplay between VOC and NO_x emissions, ozone production can be limited by either species depending on the atmospheric chemistry at a particular location.(Stedman, 2004) HC limited ozone formation, as documented by increases in ozone formation on weekends when NO_x emissions from diesel vehicles decrease significantly, has previously been the predominate mechanism in the SoCAB; however, some recent observations and models are predicting this to be changing in some areas of the basin to a NO_x limited regime.(Chinkin et al., 2003; Pollack et al., 2012; Baidar et al., 2015; Fujita et al., 2016; South Coast Air Quality Management District, 2017; Laughner and Cohen, 2019) This has shifted the focus of California regulatory agencies to significantly lowering NO_x emissions (45% reduction beyond current control measures by 2023) as called for in the SoCAB 2016 State Implementation Plan.(South Coast Air Quality Management District, 2017)

Achieving the desired NO_x reductions relies on an accurate local emissions inventory. On-road vehicles in 2018 were estimated to emit approximately 50% of the SoCAB NO_x emissions, with diesel trucks estimated to contribute the larger share (60%) of this total (see supporting material).

In California, on-road vehicle emissions contributions to the inventory are estimated using the California Air Resources Board developed EMission FACtors (EMFAC) vehicle emissions model. The current version, EMFAC2017, combines vehicle emission factors (grams per mile and per start) for selected pollutants with vehicle activity (miles driven and starts) to estimate total emissions.(California Air Resources Board, 2020b)

Using SoCAB ambient air monitor measurements and on-road vehicle emission measurements, we examine the SoCAB on-road mobile source NO_x emission inventory, and in particular the on-road gasoline and diesel apportionment. The apportionment of NO_x emissions is particularly important since any policies aimed toward reducing NO_x emissions need to be targeted appropriately in order to be effective. Absent this the expected reductions will not materialize and the needed improvements in ozone levels will be pushed even further into the future.

Experimental Methods

Ambient Measurements. Long-term (1960 – 2010) ambient molar NO_x/CO trends in California's SoCAB were described by Pollack et al. for a number of field measurement campaigns and two basin surface network monitoring sites (Azusa and Upland).(Pollack et al., 2013) Hassler et al. extended this NO_x/CO trend through 2015 and added eight additional surface network monitoring sites (La Habra, Long Beach, Magnolia, Mira Loma, North Main, Pomona, Reseda and Rubidoux).(Hassler et al., 2016) The NO_x/CO ratios were determined by bivariate least squares linear regression using only summer (May – September), non-holiday, weekday morning (0500 – 0900 local time) hourly ambient measurements. Ratios were only reported for sites where two thirds of the possible number of hourly data existed and where the resulting NO_x/CO correlation coefficient was greater than or equal to 0.5 ($r^2 \geq 0.5$) helping to restrict the measurements to fresh local motor vehicle emissions. We have extended this data record using this approach through 2018, using data from nine of the ten sites as the Magnolia site ceased operation at the end of 2014.

On-road Measurements. On-road vehicle tailpipe exhaust measurements have been collected with a remote sensor developed at the University of Denver named Fuel Efficiency Automobile Test (FEAT).(Bishop and Stedman, 1996) FEAT is composed of an infrared (IR) and ultraviolet (UV) light source placed across a single lane roadway from four non-dispersive IR and one (NO only for the pre-2008 data sets) or two dispersive UV detectors (includes NO_2 starting in 2008) that allow the measurement of vehicle exhaust gases as a molar ratio to exhaust CO_2 (i.e., CO/CO_2, NO/CO_2, NO_2/CO_2 etc.).(Burgard et al., 2006a) Each measured species ratio is scaled using certified (± 2% accuracy) gas cylinder ratios measured daily as needed at each site by FEAT. This corrects for variations in instrument sensitivity and most importantly ambient CO_2 levels caused by changes in atmospheric pressure, temperature and background pollutants. The molar ratios can also be converted into fuel-based emission factors of grams of pollutant per kg of fuel by the carbon balance method. This uses a carbon mass fraction for the fuel of 0.86 and a doubling of the HC/CO_2 ratio to normalize the reading with a flame ionization detector and compensate for the weak IR absorbance of many aromatic compounds.(Singer et al., 1998) Each measurement includes a video image of the license plate of the vehicle that is manually transcribed and used to retrieve non-personal vehicle information (i.e. age and type) from the California registration records that is combined with the emission measurements into a final database for analysis.

On-road emission measurements have been collected using FEAT from light and medium-duty vehicles in California's SoCAB since 1989. However, NO measurements were not collected until the late 1990's when the instrumentation for collecting those measurements was developed.(Popp et al., 1999) Beginning in 1999, emission measurements have been collected at the on-ramp from southbound La Brea Ave. to eastbound I-10, about midway between downtown LA and Santa Monica. To date there have been eight data sets collected at this West Los Angeles location (1999, 2001, 2003, 2005, 2008, 2013, 2015 and 2018) that includes more than 165,000 vehicle emission measurements.(Bishop and Stedman, 2008; Bishop et al., 2010; Bishop and Stedman, 2015) The 1999 - 2005 measurements were made in the fall of each year. Beginning with the 2008 measurements we began collecting emission measurements of NO_2, allowing for the reporting of vehicle NO_x emissions, and the measurement dates switched to the spring.(Burgard et al., 2006b) Though only a single site within the basin, other researchers have shown that these measurements are representative of basin-wide emissions.(Nowak et al., 2012; Pollack et al., 2013; Hassler et al., 2016; Kim et al., 2016)

Seven additional data sets, also collected within the basin since 1999, in Riverside (1999, 2000 and 2001), at the intersection of I-710 and SR91 (1999), Van Nuys (2010) and at two sites in

Lynwood (2018) are included in this analysis.(Bishop and Stedman, 2008; Bishop et al., 2012; Bishop, 2019) Heavy-duty diesel trucks with either elevated or ground level exhaust emissions were also measured in the spring of 2017 at the Peralta weigh station on SR 91 in the Anaheim Hills of the SoCAB.(Haugen et al., 2018) All of the databases used in this study, as well as many others compiled by the University of Denver, are available at www.feat.biochem.du.edu.

EMFAC2017 Modeling. California's EMFAC2017 vehicle emissions factor model was run using the online EMFAC2017 web database v1.0.2 (https://www.arb.ca.gov/emfac/2017/). For comparison with the ambient measurement ratios summer emissions (online model allows summer or annual estimates) for the SoCAB were modeled for years 2001, 2003, 2005, 2008, 2010, 2013, 2015 and 2018 to match the West Los Angeles remote sensing measurement years. This version of the EMFAC model only predicts emissions back to calendar year 2000 so we did not compare modeled emissions to the 1999 measurements.

Running exhaust molar ratios were calculated for each year by summing the model predicted short tons per day for the thirteen gasoline vehicle types output by EMFAC. The tons were converted into grams and the grams into moles of each pollutant and then ratioed. Model years were aggregated by the model for each vehicle type and predictions were generated for two speeds, aggregated over all of the drive cycles included in the model and at a fixed 20mph. The latter most closely matches the average speed observed at the West Los Angeles FEAT on-road measurement site. The supporting material includes a sample output of this process (see Table S6).

Annual running exhaust emission factors (grams per kilogram of fuel) for 2018 were calculated by model year for gasoline powered light-duty passenger vehicles (model type LDA) and trucks (fuel consumption weighted composite of model vehicle types LDT1, LDT2 and MDV) from the tons/day predicted by the EMFAC model. The chosen truck types cover the weight classes observed at the West Los Angeles site and account for more than 87% of the predicted truck gasoline fuel consumption by the model. Aggregated speeds were used for these calculations as the model does not provide fuel consumption at a fixed speed setting. The model predicted tons/day for each model year were converted into grams/day, the predicted gallons of gasoline consumed were converted into kilograms assuming a density of 0.75 g/ml for California reformulated gasoline and the two ratioed for each pollutant. The supporting material includes the 2018 calculations (see Table S9).

Annual fuel specific NO_x emission factors for the 2017 heavy-duty diesel truck measurements were calculated using EMFAC2017 in a similar manner. A fuel weighted composite of diesel trucks included all medium-heavy duty diesel trucks greater than 26,000 lbs. (EMFAC model type T6) and all heavy-heavy duty diesel trucks (EMFAC model type T7) except those using the Truck and Bus rule agricultural provision (T7 Ag) with aggregated speeds.(California Air Resources Board, 2018) The density of ultra-low sulfur diesel fuel was assumed to be 0.86 g/ml.

Results

NO_x/CO Trends. Maintaining the graphical approached used in Hassler et al., Figure 1 plots the log of the molar NO_x/CO ratios for the California SoCAB surface network sites against measurement year for the 1983 - 2018 time period.(Hassler et al., 2016) The solid lines in Figure 1 represent a quadratic fit applied to the 1983 - 2009 ambient measurement ratios and a best fit straight line for the remaining 2010 - 2018 ratios. The scatter in these measurements reflects the distribution differences between the gasoline and diesel fleets, vehicle operating characteristics and any observed contributions from stationary or non-road sources. It is expected that gasoline vehicles will figure more prominently in the CO emission contributions while diesel vehicles will contribute a larger fraction of the NO_x emissions.

The ambient ratios increase steadily until about 2010 after which they level out and then decrease. The rise is consistent with the observation by Pollack et al. that vehicle CO emissions decreased at about twice the rate of NO_x emissions over the earlier time period.(Pollack et al., 2013) Since 2010, on-road NO_x emissions have seen significant reductions from both the light-duty gasoline and heavy-duty diesel fleets.(Bishop and Haugen, 2018; Haugen et al., 2018) These reductions have been driven by the introduction of LEV II light-duty vehicles in 2009 (phased in between 2004 & 2009 in California) and by the phase-in of selective catalytic reduction systems for NO_x control in heavy-duty diesel trucks beginning with 2011 trucks.

FEAT molar NO_x/CO ratios for the gasoline portion of each site's fleet are plotted against measurement year for the 15 data sets collected in the SoCAB since 1999. For the data sets collected prior to 2008 the molar NO_x/CO ratios only include measurements for the moles of

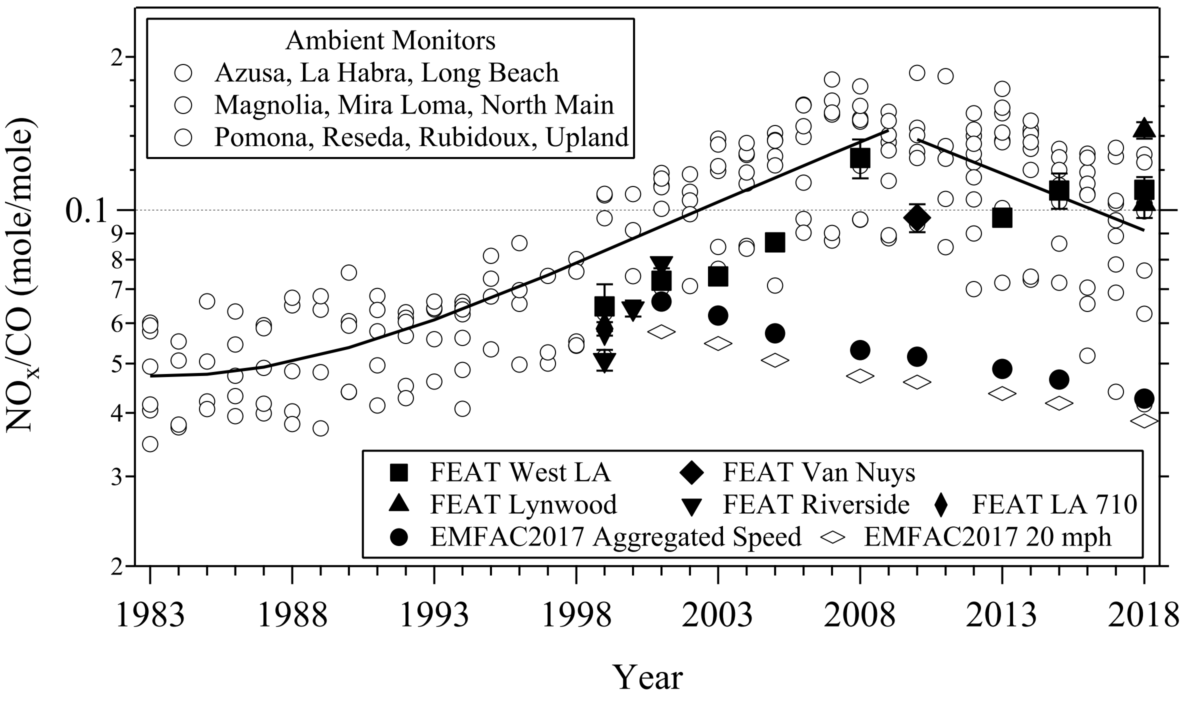

Figure 1. Molar NO_x/CO emission ratios from California's SoCAB ambient monitors (○), on-road measured average ratios for gasoline vehicles from six basin locations and EMFAC2017 running exhaust modeled ratios for gasoline only vehicles with aggregated speed or fixed at 20mph versus measurement year. A quadratic fit is shown for the 1983 – 2009 ambient data and a best fit straight line for the 2010 to 2018 measurements. Uncertainties for the FEAT measurements are standard error of the mean calculated using the daily means.

NO. However, since we have restricted these comparisons to only gasoline powered vehicles this should only slightly (< 1%) underestimate the true NO_x/CO ratios as gasoline engines emit little NO_2. The number of diesel vehicles with ground level exhaust at these on-road sites is small (1.5 to 3%) and their inclusion does, as expected, increase the ratios (~8% on average) but does not significantly change the results (Table S7in the supporting material). Uncertainties displayed are standard error of the mean calculated from the daily means.

This is not a direct comparison with the ambient measurements, as the ratios derived from the on-road measurements exclude diesel powered vehicles. However, in general the on-road measurements fall within the lower range of the observed ambient NO_x/CO ratios, rising along with them until the 2010 peak. The figure indicates that after 2010 the on-road NO_x/CO ratios continue to increase slightly and do not show the decreases observed in the ambient ratios.

As noted, summer EMFAC2017 molar NO_x/CO ratios were calculated from the model for the SoCAB gasoline fleet using two speed selections: 1) a static 20mph and 2) aggregated speeds; these are shown in Figure 1. The aggregated speed setting produces slightly higher NO_x/CO ratios than the static 20 mph speed for each of the years modeled. The EMFAC2017 predicted ratios begin in 2001 with values that are in general agreement with the ambient and the on-road measurements but then steadily decrease in the following years. Noticeably absent from the modeled ratio predictions is the rising ratio values found in the ambient and on-road measurements between 2001 and 2010. The 2018 molar NO_x/CO ratios estimated by EMFAC are 1.8 (aggregated speed) and 2.4 (20mph fixed speed) times lower than the average ambient measurements.

Because we are comparing ratios it is not immediately clear whether the disagreement between the model and the ambient measurements is the result of an under prediction of NO_x or an over prediction of CO. Since the on-road measured ratios generally fall within the range of the ambient measurements we will use the on-road data from the West Los Angeles site to investigate potential differences with the model predictions.

Gasoline Vehicle Emissions Comparison. Figure 2 compares the fuel specific CO emissions for the 2018 West Los Angeles FEAT measurements and the EMFAC2017 predicted emission factors by model year for gasoline light-duty passenger vehicles (top graph) and light-duty trucks (bottom graph). EMFAC2017 light-duty trucks, as previously mentioned, are a fuel use weighted composite emission factor for the model types LDT1, LDT2 and MDV. The model emission factors were calculated on an annual basis for the SoCAB using aggregated speeds since that speed setting predicted the higher ratio values (see supporting material). Uncertainties for the FEAT data are standard error of the mean determined from the daily measurements. Because of the decline in the number of vehicle measurements with age and their increasing uncertainty the on-road measurement plotted at model year 1990 is the average for all 1987 - 1993 vehicles.

The emissions by model year comparisons between the on-road measurements and the model predictions, within the uncertainties, are in generally good agreement. We can calculate an age normalized fleet mean emissions for the EMFAC2017 model output using the fleet CO measurement fractions by model year and vehicle type observed in the 2018 West Los Angeles measurements to further compare the overall agreement (see Table S8). Mean fuel specific

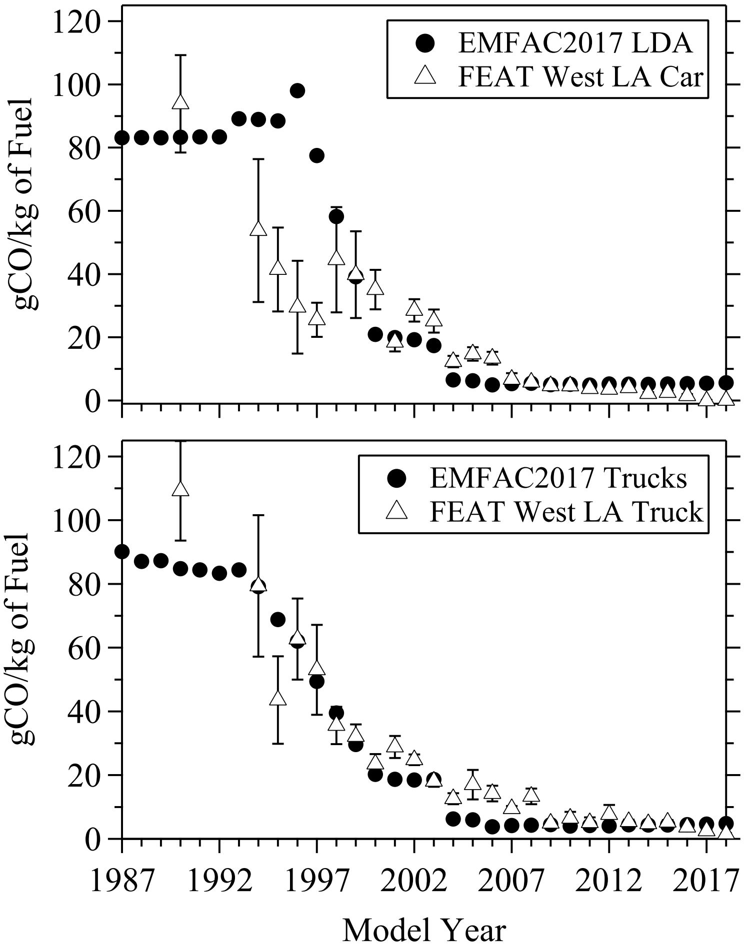

Figure 2. Fuel specific CO emissions for the 2018 West Los Angeles gasoline fleet (Δ) and the EMFAC2017 predicted 2018 SoCAB gasoline fleet (model type LDA) emissions (●) versus model year for light-duty passenger vehicles (top panel) and trucks (bottom panel). EMFAC2017 truck emission factors are a fuel use weighted composite of model types LDT1, LDT2 and MDV. Uncertainties for the FEAT measurements are standard error of the mean calculated using the daily means.

EMFAC2017 CO emissions are slightly over-predicted (+11%) for passenger vehicles (10.5 vs. 9.3 ± 0.5 gCO/kg of fuel) and are slightly under-predicted (-20%) for the light-duty trucks (11.1 vs 13.9 ± 0.6 gCO/kg of fuel). When combined, the fleet means are not statistically different at the 95% CI (10.7 vs 11.2 ± 0.6 gCO/kg of fuel).

Figure 3 is the companion graph comparing the fuel specific NO_x emissions by model year for gasoline light-duty passenger vehicles (top) and light-duty trucks (bottom) for the 2018 FEAT West Los Angeles measurements and the EMFAC2017 model predictions. The NO_x comparison is quite good for the 2009 and newer model year vehicles that all have near-zero emissions.

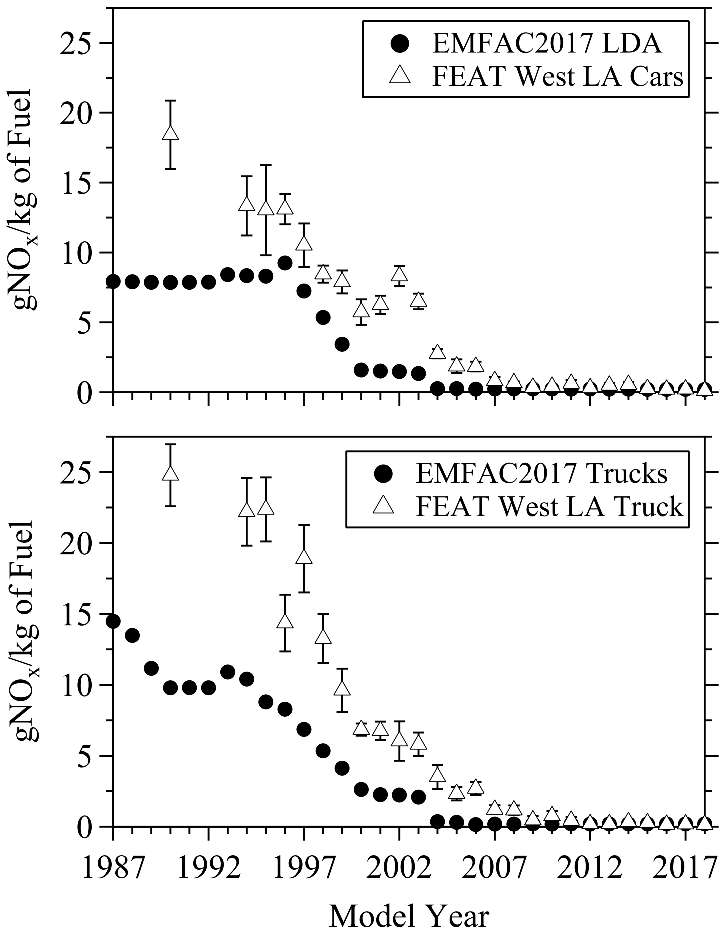

Figure 3. Fuel specific NO$_x$ emissions for the 2018 West Los Angeles gasoline fleet (Δ) and the EMFAC2017 predicted 2018 SoCAB gasoline fleet (model type LDA) emissions (●) versus model year for light-duty passenger vehicles (top panel) and trucks (bottom panel). EMFAC2017 truck emission factors are a fuel use weighted composite of model types LDT1, LDT2 and MDV. Uncertainties for the FEAT measurements are standard error of the mean calculated using the daily means.

Unlike the CO emissions comparison however, the observed on-road gNO$_x$/kg of fuel emissions for both vehicle types are substantially higher than predicted by the EMFAC2017 model for 2008 and older models. For passenger vehicles (0.7 vs 1.8 ± 0.1 gNO$_x$/kg of fuel) and trucks (1.1 vs 3.0 ± 0.1 gNO$_x$/kg of fuel) the on-road age normalized mean emissions are factors of 2.6 and 2.7 times higher than predicted by the model. It is these differences in light-duty gasoline NO$_x$ emissions that are the likely explanation for the differences observed in the EMFAC NO$_x$/CO ratio comparison with the ambient and on-road measurements (see Figure 1).

Fujita et al. reported on an under-prediction of NO$_x$ emissions with an earlier version of the model, EMFAC2007, for measurements collected in a Van Nuys, CA tunnel in the summer of

2010.(Fujita et al., 2012) EMFAC2007 under reported NO_x emissions by factors of 1.2 and 1.4 for the median weekday and weekend measurements respectively. However, the under-prediction was larger (factor of 1.8) for a Sunday morning measurement when the fleet was almost exclusively gasoline powered vehicles (8 diesel vehicles in 1290 total vehicles) while CO emissions were accurately predicted.

A study to model $PM_{2.5}$ levels in California's San Joaquin Valley came to a similar conclusion using the 2014 version of EMFAC.(Kleeman et al., 2019) Observed ambient NO_x emissions in Fresno in the winter of 2013 were significantly higher during the morning rush hour period than predicted by the model even after including soil NO_x emissions. Concentrations of total reactive nitrogen were consistently under-predicted due to insufficient levels of NO_x emissions. The authors concluded that there is an "unknown source of NO_x emissions that is not currently represented in the emissions inventory".(Kleeman et al., 2019)

On-road measurements collected in Fresno in 2008, prior to the recession, found an older light-duty fleet (~8.7 years) than data collected during the same campaign in San Jose (~7.9 years) and West Los Angeles (7.3).(Bishop et al., 2010) The 2008 - 2009 recession increased fleet age by 2 years at the West Los Angeles site as observed in 2013 and we would expect fleets in the San Joaquin Valley to have aged at least this much.(Bishop and Stedman, 2014) This would amplify the number of LEV I vehicles (1994 - 2003 model years) in the San Joaquin Valley during the 2013 measurements that we have found to have NO_x emissions most under-predicted by the EMFAC model which could be the source for the missing NO_x emissions.

One possible explanation for the differences observed in the fuel specific NO_x emissions for the 2005 and older model year vehicles is that emission deterioration rates are actually larger on-road than assumed in EMFAC. Using the multiple years of emission measurements collected at the West Los Angeles site and plotting fuel specific NO_x emissions for the gasoline fleet versus vehicle age at the time of measurement by model year we can estimate the on-road NO_x emissions deterioration rates and compare those with model predictions using the same multi-year approach.(Bishop and Stedman, 2008) The supporting material details the calculation process and Figure S1 shows the graphs for the multi-year on-road and model predicted NO_x emission factors. We have used year 2000 in the EMFAC2017 model for the eighth data point as a substitute for the 1999 on-road measurements since the year 1999 cannot be modeled.

Figure 4 plots the emissions deterioration rates in gNO_x/kg of Fuel/Year versus model year for the gasoline fleet obtained from the fitting results using the data shown in Figure S1. The uncertainties plotted are the standard error of the least squares fit for each model year. The deterioration rate comparison has a similar trend within the uncertainties for the 2007 and newer model years but the on-road deterioration rates, like the fuel specific emissions (see Figure 3), are much higher than the EMFAC estimates for the 2006 and older model year vehicles. The EMFAC2017 estimated emissions deterioration rates do not show any increases until the 2003 model year vehicles. This may be the result of the model assuming California LEV II vehicle emission deterioration rates through the 2004 model year vehicles. This is when their introduction into the California fleet was to begin but we would not expect them to be a significant fraction of the fleet until later model years.(DieselNet, 2018) For the oldest model year vehicles (pre-1999) the final deterioration rates are approximately a factor of 4 higher (~0.5 vs ~0.125 gNOx/kg of Fuel/Year) for the on-road measurements. For perspective 1996 model year vehicles with a 0.49 gNO_x/kg of Fuel/Year emissions deterioration rate represents a 5.8%/year emissions increase or an emissions doubling time of 12 years. The lower rates predicted by the model are likely the result of the NO_x emission factors appearing to be capped around 10 gNO_x/kg of Fuel for 1996 - 1989 model year vehicles (see Figure S1) which we do not observe in the on-road measurements.

Fuel-Based Inventory. The SoCAB inventory is a critical piece of information that is used to shape regulatory policy for future emission reductions toward the goal of achieving compliance with the NAAQS. To estimate the extent of the NO_x under-prediction we have constructed a NO_x fuel-based inventory for the SoCAB using our on-road emission measurements for light-duty gasoline and heavy-duty diesel vehicles. On-road heavy-duty diesel truck emissions were measured at the Peralta weigh station on SR 91 in the spring of 2017.(Haugen et al., 2018) Figure S2 shows the fuel specific NO_x emissions by model year comparison for the on-road heavy-duty diesel measurements and the EMFAC2017 predictions. Annual EMFAC2017 emission factors were calculated using aggregated speeds and a fuel weighted composite emission factor for year 2017 using the diesel truck types previously described. The on-road emissions are generally higher than the model predictions but comparison of estimated mean emissions using the Peralta model year distribution for both sets of emission factors results in only a 20% difference (10 vs 12.5 gNO_x/kg of fuel) which overall is good agreement.

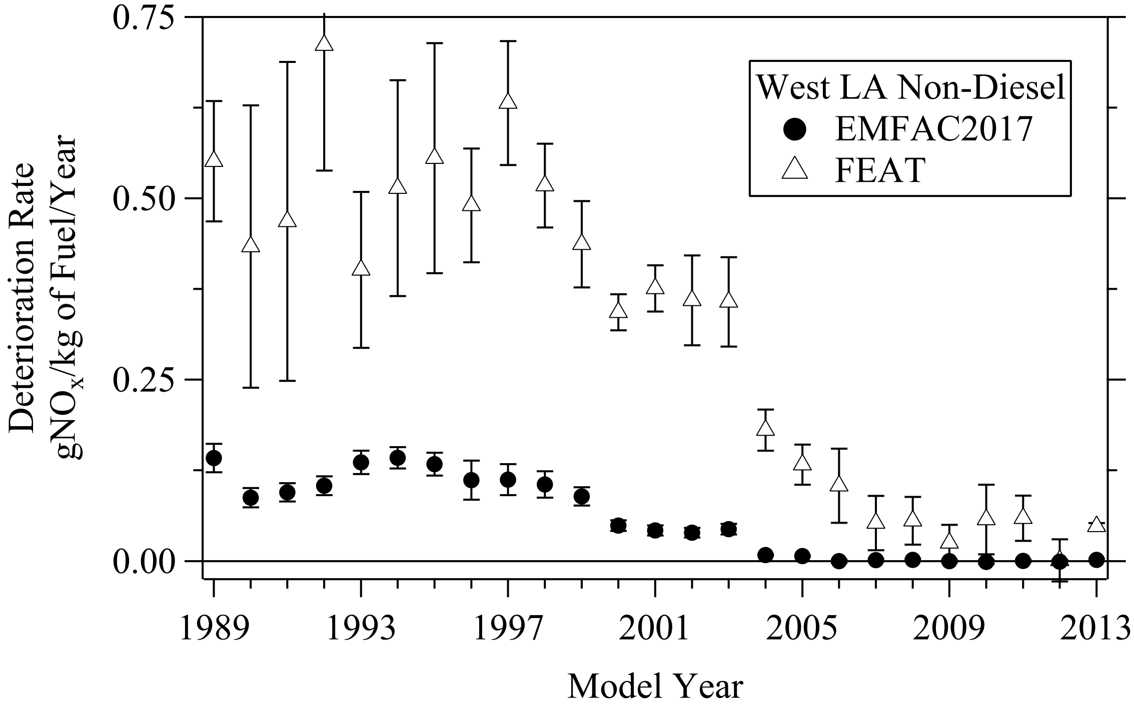

Figure 4. Fuel specific NO$_x$ emissions deterioration rates (gNO$_x$/kg of Fuel/Year) versus model year for the West Los Angeles on-road measurements (Δ) and deterioration rates calculated using the EMFAC2017 model (●) for the gasoline fleet since 1999. The uncertainties are the standard error of the slope for each model year's least squares fit.

Following the methodology of Hassler et al. we have calculated the 2018 daily gasoline and diesel fuel consumption for the SoCAB in kilograms/day using annual fuel sales data for the State of California (Table S10 in the supporting material).(Hassler et al., 2016) These values are multiplied by the mean on-road emission factors measured at the West Los Angeles site (for the gasoline fleet) in 2018 and the Peralta weigh station in 2017 (heavy-duty diesel fleet) to estimate short tons/day emissions in the Basin. Table 1 compares the on-road fuel-based inventory with the 2018 Annual California Air Resources Board inventory predicted by the online California Emission Projections and Analysis Model (CEPAM) emission tool for the SoCAB (see supporting material).(California Air Resources Board, 2020a) The fuel-based inventory indicates 74% more NO$_x$ emissions from the light-duty gasoline fleet than accounted for in the inventory despite the fact that the fuel-based inventory does not include idle and starting emissions. The light-duty CO and heavy-duty diesel NO$_x$ emissions inventory comparisons are in better agreement with the fuel-based results.

Table 1. 2018 Inventory Comparison for California's South Coast Air Basin

South Coast Air Basin	West Los Angeles (Gasoline)	Peralta Weigh Station (Diesel)
kg Fuel/day[a]	$(4.8 \pm 0.3) \times 10^7$	$(8.1 \pm 0.7) \times 10^6$
On-road gNOx/kg of Fuel[b]	2.3 ± 0.1	12.5 ± 0.6
Fuel-Based NO_x tons/day[c,d]	122 ± 13	111 ± 15
CEPAM[e] NO_x tons/day[c]	70	111
On-road gCO/kg of Fuel[b]	11.2 ± 0.2	5.9 ± 0.9
Fuel-Based CO tons/day[c,d]	592 ± 47	53 ± 13
CEPAM[e] CO tons/day[c]	620	24

[a] derived from State annual fuel sales from Hassler et al., (2016) see supporting material.

[b] uncertainties are standard error of the mean derived from the daily measurements.

[c] short tons; 1 ton = 0.907 metric tons.

[d] uncertainties are the combined uncertainty from the fuel and emission factors.

[e] California Emission Projections and Analysis Model

The fuel-based inventory estimates increase the light-duty gasoline NO_x emissions to being on par, or slightly higher than, the heavy-duty NO_x emissions. This is not a new observation as Kim et al. previously reported this for the 2010 SoCAB fleet.(Kim et al., 2016) It does however, raise the question as to whether the extra NO_x emissions increases the total NO_x inventory or are they offset by over estimates in other categories? The 2018 total inventory estimates of 1741 tons CO/day and 356 tons NO_x/day result in a molar NO_x/CO ratio of 0.12 which is within the spread of the morning ambient measurements and supports the two totals (see Table S5 in the supporting material). In addition Morris et al. showed good agreement between OMI satellite NO_2 observations and the 2018 basin NO_x inventory again supporting the NO_x total and suggesting that over estimates exist in other inventory categories.(Morris et al., 2019)

Figure 5 is a plot of the percent of the total gNO_x/kg of Fuel emissions contributed by vehicle type and fuel, compared with the vehicle age distribution, by model year for the 2018 West Los Angeles on-road measurements. The newest model LEV II vehicles account for the largest percentage of vehicles in the fleet (2009 & newer ~62% of fleet) but only a small minority of the

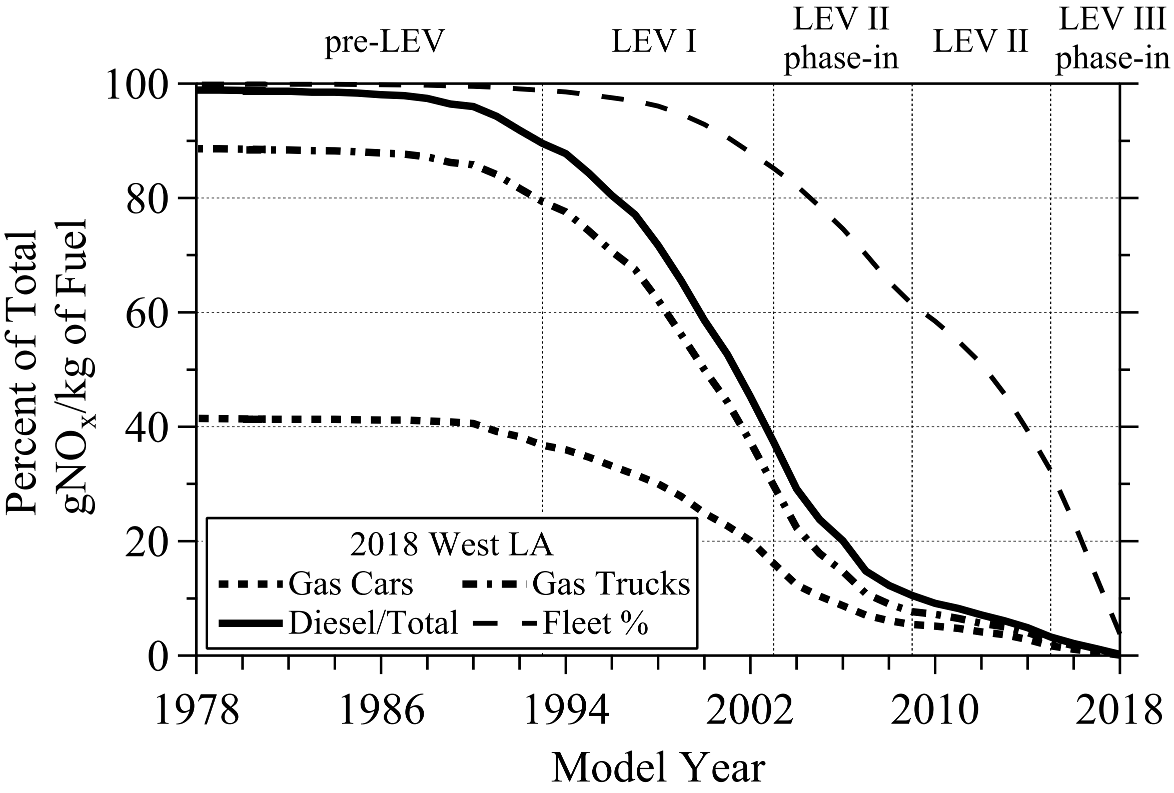

Figure 5. 2018 West Los Angeles light-duty vehicle percent of total gNO$_x$/kg of fuel emissions by vehicle type and fuel and the 2018 fleet age distribution percentage by model year. Along the top of the plot are the approximate model years for the California vehicle certification levels.

emissions (~11% of fuel-based NO$_x$). The simple addition of lower emitting new vehicles, even if they are zero emitting vehicles, will not appreciably change the light-duty NO$_x$ emissions distribution and will not provide any significant changes to the NO$_x$ inventory. The majority of the light-duty NO$_x$ emissions are found in the older LEV I vehicles. LEV I vehicles (1994 - 2003) are approximately 16% of the fleet observed at the West Los Angeles site but account for half of the fuel specific NO$_x$ emissions. The highest emitting 10% of the LEV I vehicles are responsible for more than half of these emissions. Even small reductions in this segment of the fleet will yield large reductions in NO$_x$ emissions. Currently 2001 model year vehicles, as an example, have a year over year percentage removal from the fleet of 8.3% (~8.5 year half-life). The desire for large reductions in the NO$_x$ inventory in the SoCAB will require an extraordinary effort to achieve and LEV I gasoline vehicles appear to be a significant source. It is unlikely that we can expect the elimination of these vehicles from the fleet through natural attrition, especially within the current economic downturn, will occur very fast. This suggests that other methods need to be explored to hasten the removal of high NO_x emitters from the fleet.

Summary

State and regional air quality officials depend on air basin inventories for designing emission reduction plans to help a region comply with the NAAQS. Because the time frames involved in these plans are typically long and the implementation costs are often high any errors in the inventory can result in costly missteps and lost time if the emissions reductions anticipated are not achieved. The SoCAB currently experiences some of the nation's highest ozone levels; as a result local and state regulations are targeting large NO_x reductions from heavy-duty diesel vehicles but admittedly believe that reductions from other sources will be necessary to achieve these targets.

Ambient molar NO_x/CO ratios collected on weekdays during the morning rush hour in the SoCAB were compared with those estimated for gasoline powered vehicles by California's EMFAC2017 emissions model and on-road emission measurements collected from gasoline powered vehicles at sites within the SoCAB. The ambient molar NO_x/CO ratios steadily increase until around 2010 when they level off and then begin to decline through 2018. Both the EMFAC2017 predictions and the on-road emission measurements from 1999 - 2001 have ratios that are along the lower edge of the ambient measurements. However, after 2001 the EMFAC2017 and the on-road NO_x/CO ratios diverge with the model predicted ratios decreasing significantly through 2018 and ending up factors of 1.8 to 2.4 below the 2018 average. The on-road measured ratios increase along with the ambient measurements until their peak and then increase slightly through 2018.

The difference between the two sets of ratios was found to most likely be that the EMFAC2017 model underestimates the NO_x emission factors for the gasoline fleet as the CO emission factor comparison was good. Comparisons with 2018 on-road measurements collected in West Los Angeles found that the fuel specific NO_x emission factor comparison was good for the 2009 and newer model year vehicles that all have near-zero emissions. However, for the 2008 and older models the observed on-road gNO_x/kg of fuel emissions for both passenger vehicles and trucks increase at a higher rate than predicted by the EMFAC2017 model. For gasoline passenger vehicles (0.7 vs 1.8 ± 0.1 gNO_x/kg of fuel) and trucks (1.1 vs 3.0 ± 0.1 gNO_x/kg of fuel) the on-road age normalized mean emissions are factors of 2.6 and 2.7 times higher than predicted by the model. For the oldest model year vehicles (pre-1999) we found NO_x emission deterioration rates that were approximately a factor of 4 higher (~0.5 vs ~0.125 gNOx/kg of Fuel/Year) for the on-road measurements, a likely cause of the under prediction.

A fuel-based inventory for the 2018 SoCAB constructed using the on-road measurements from the West Los Angeles site indicates that there is 74% more NO_x emissions from the light-duty gasoline fleet than represented in the inventory. These estimates imply that NO_x emissions from light-duty gasoline vehicles are comparable or slightly higher than those from heavy-duty vehicles. The majority of the light-duty NO_x emissions are found in the older LEV I (1994 - 2003) vehicles, which make up approximately 16% of the fleet observed in 2018 at the West Los Angeles site but account for half of the fuel specific NO_x emissions. The emissions of these vehicles are as expected skewed with the highest emitting 10% responsible for more than half of the LEV I contribution or 25% of the total NO_x emissions.

The under reporting of NO_x emission factors by the EMFAC model has been reported by other researchers, however the newer iterations continue to carry forward this problem. This issue significantly changes the NO_x emissions distribution for mobile sources in the SoCAB and will lead to an overestimation in the percent reductions that can be achieved in lowering diesel vehicle NO_x emissions. In addition it overlooks the possibility of significant NO_x emission reductions by targeting the removal of older high emitting gasoline vehicles.

Acknowledgment

The author would like to acknowledge the Coordinating Research Council (E-23), the Real World Working Group, the California Air Resources Board and West Virginia University for their financial support in collecting the on-road emission measurements. I would also like to thank Ilana Pollack and Birgit Hassler for sharing their molar ratio data and Brian McDonald for the LA Basin fuel consumption data. I also want to thank David Carslaw, Russell Dickerson and Lew Cohen for their helpful discussions.

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About the author

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Supplemental Data

Supplemental data for this paper can be accessed on the publisher's website.

Supplemental Material for

Does California's EMFAC2017 Vehicle Emissions Model Under-predict California Light-duty Gasoline Vehicle NO_x Emissions?

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Table S1. California's 2018 South Coast Air Basin CO and NO_x Emissions Inventory for Stationary Sources (https://www.arb.ca.gov/app/emsinv/fcemssumcat/fcemssumcat2016.php).

Stationary Sources	AREA	CO	NOx
Fuel Combustion		tons/day	tons/day
ELECTRIC UTILITIES	SOUTH COAST	6.5907	5.177
COGENERATION	SOUTH COAST	0.5742	0.424
OIL AND GAS PRODUCTION (COMBUSTION)	SOUTH COAST	0.7754	1.7425
PETROLEUM REFINING (COMBUSTION)	SOUTH COAST	4.9539	8.2539
MANUFACTURING AND INDUSTRIAL	SOUTH COAST	16.1448	14.1926
FOOD AND AGRICULTURAL PROCESSING	SOUTH COAST	0.3039	0.2268
SERVICE AND COMMERCIAL	SOUTH COAST	16.1491	10.8974
OTHER (FUEL COMBUSTION)	SOUTH COAST	2.632	3.219
Total Fuel Combustion		48.124	44.1332
Waste Disposal			
SEWAGE TREATMENT	SOUTH COAST	0.0064	0.007
LANDFILLS	SOUTH COAST	0.5176	0.6491
INCINERATORS	SOUTH COAST	0.5618	1.7638
SOIL REMEDIATION	SOUTH COAST	0	0
OTHER (WASTE DISPOSAL)	SOUTH COAST	0	0
Total Waste Disposal		1.0858	2.4199
Cleaning and Surface Coatings			
LAUNDERING	SOUTH COAST	0	0
DEGREASING	SOUTH COAST	0	0
COATINGS AND RELATED PROCESS SOLVENTS	SOUTH COAST	0.0099	0.0148
PRINTING	SOUTH COAST	0	0
ADHESIVES AND SEALANTS	SOUTH COAST	0	0
OTHER (CLEANING AND SURFACE COATINGS)	SOUTH COAST	0.0604	0.027
Total Cleaning and Surface Coatings		0.0703	0.0418
Petroleum Production and Marketing			
OIL AND GAS PRODUCTION	SOUTH COAST	0.0185	0.0381
PETROLEUM REFINING	SOUTH COAST	5.161	1.3006
PETROLEUM MARKETING	SOUTH COAST	0.0066	0.0057
OTHER (PETROLEUM PRODUCTION AND MARKETING)	SOUTH COAST	0.0006	0.0009
Total Petroleum Production and Marketing		5.1867	1.3453
Industrial Processes			
CHEMICAL	SOUTH COAST	0.0314	0.0072
FOOD AND AGRICULTURE	SOUTH COAST	0	0.0001
MINERAL PROCESSES	SOUTH COAST	0.1625	0.3991
METAL PROCESSES	SOUTH COAST	0.1853	0.0422
WOOD AND PAPER	SOUTH COAST	0	0
GLASS AND RELATED PRODUCTS	SOUTH COAST	0.0029	0
ELECTRONICS	SOUTH COAST	0.0004	0.0004
OTHER (INDUSTRIAL PROCESSES)	SOUTH COAST	0.1257	0.0225
Total Industrial Processes		0.5082	0.4715
Total Stationary Sources		54.98	48.41

Table S2. California's 2018 South Coast Air Basin CO and NO_x Emissions Inventory for Areawide Sources (https://www.arb.ca.gov/app/emsinv/fcemssumcat/fcemssumcat2016.php).

Areawide Sources	AREA	CO	NOx
Solvent Evaporation		tons/day	tons/day
CONSUMER PRODUCTS	SOUTH COAST	0	0
ARCHITECTURAL COATINGS AND RELATED PROCESS SOLVENTS	SOUTH COAST	0	0
PESTICIDES/FERTILIZERS	SOUTH COAST	0	0
ASPHALT PAVING / ROOFING	SOUTH COAST	0	0
Total Solvent Evaporation		0	0
Miscellaneous Processes			
RESIDENTIAL FUEL COMBUSTION	SOUTH COAST	46.7507	14.5577
FARMING OPERATIONS	SOUTH COAST	0	0
CONSTRUCTION AND DEMOLITION	SOUTH COAST	0	0
PAVED ROAD DUST	SOUTH COAST	0	0
UNPAVED ROAD DUST	SOUTH COAST	0	0
FUGITIVE WINDBLOWN DUST	SOUTH COAST	0	0
FIRES	SOUTH COAST	3.0231	0.0751
MANAGED BURNING AND DISPOSAL	SOUTH COAST	6.3053	0.191
COOKING	SOUTH COAST	0	0
OTHER (MISCELLANEOUS PROCESSES)	SOUTH COAST	0	0
Total Miscellaneous Processes		56.0791	14.8238
Total Areawide Sources		56.08	14.82

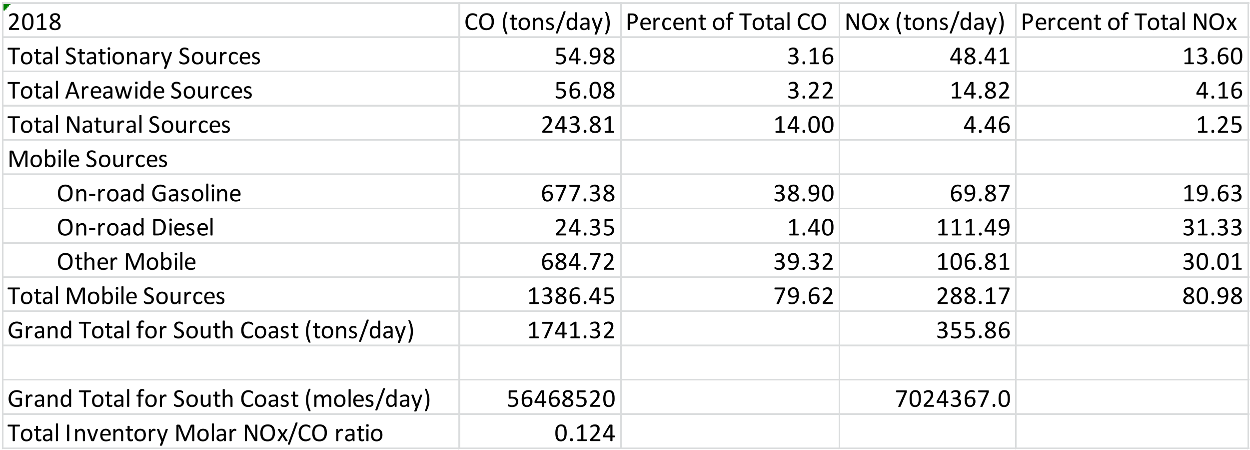
Table S3. California's 2018 South Coast Air Basin CO and NO_x Emissions Inventory for Natural Sources (https://www.arb.ca.gov/app/emsinv/fcemssumcat/fcemssumcat2016.php).

Natural (Non-Anthropogenic) Sources	AREA	CO	NOx
Natural Sources		tons/day	tons/day
BIOGENIC SOURCES	SOUTH COAST	0	0
GEOGENIC SOURCES	SOUTH COAST	0	0
WILDFIRES	SOUTH COAST	243.8116	4.4644
Total Natural Sources		243.81	4.46

Table S4. California's 2018 South Coast Air Basin CO and NO_x Emissions Inventory for Mobile Sources (https://www.arb.ca.gov/app/emsinv/fcemssumcat/fcemssumcat2016.php).

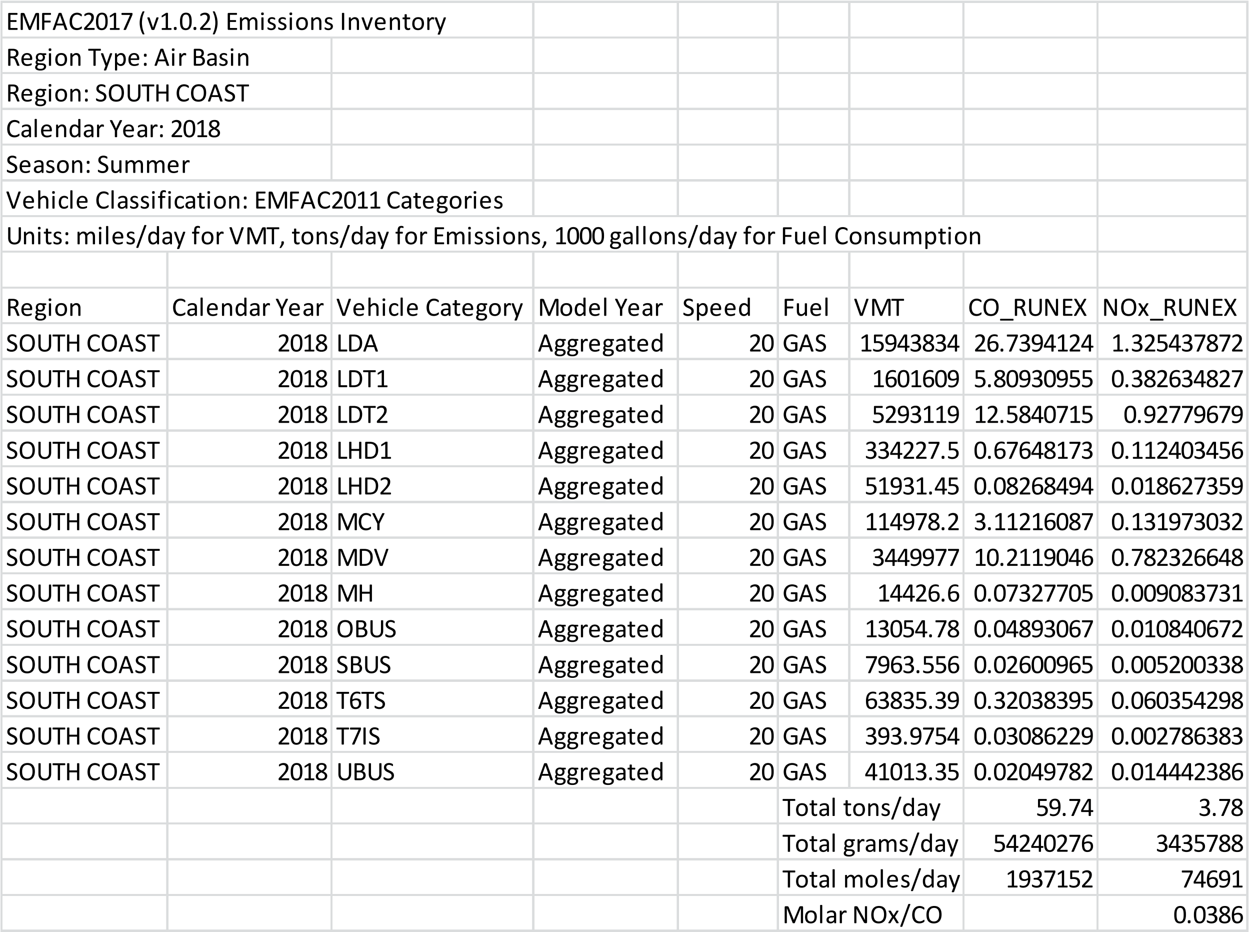
Mobile Sources	AREA	CO	NOx
On-Road Motor Vehicles		tons/day	tons/day
LIGHT DUTY PASSENGER (LDA)	SOUTH COAST	266.4686	22.8899
LIGHT DUTY TRUCKS - 1 (LDT1)	SOUTH COAST	56.4124	5.1051
LIGHT DUTY TRUCKS - 2 (LDT2)	SOUTH COAST	123.6348	13.5449
MEDIUM DUTY TRUCKS (MDV)	SOUTH COAST	144.0057	16.5407
LIGHT HEAVY DUTY GAS TRUCKS - 1 (LHDGT1)	SOUTH COAST	17.3217	5.0034
LIGHT HEAVY DUTY GAS TRUCKS - 2 (LHDGT2)	SOUTH COAST	2.2808	0.7972
MEDIUM HEAVY DUTY GAS TRUCKS (MHDGT)	SOUTH COAST	8.2656	1.4127
HEAVY HEAVY DUTY GAS TRUCKS (HHDGT)	SOUTH COAST	3.4053	0.3647
LIGHT HEAVY DUTY DIESEL TRUCKS - 1 (LHDDT1)	SOUTH COAST	2.292	10.7233
LIGHT HEAVY DUTY DIESEL TRUCKS - 2 (LHDDT2)	SOUTH COAST	0.7253	3.2044
MEDIUM HEAVY DUTY DIESEL TRUCKS (MHDDT)	SOUTH COAST	3.3642	22.4182
HEAVY HEAVY DUTY DIESEL TRUCKS (HHDDT)	SOUTH COAST	8.4102	60.8844
MOTORCYCLES (MCY)	SOUTH COAST	47.986	2.5065
HEAVY DUTY DIESEL URBAN BUSES (UBD)	SOUTH COAST	9.2827	10.2338
HEAVY DUTY GAS URBAN BUSES (UBG)	SOUTH COAST	2.9786	0.5497
SCHOOL BUSES - GAS (SBG)	SOUTH COAST	0.562	0.0707
SCHOOL BUSES - DIESEL (SBD)	SOUTH COAST	0.0975	2.0059
OTHER BUSES - GAS (OBG)	SOUTH COAST	1.9521	0.3753
OTHER BUSES - MOTOR COACH - DIESEL (OBC)	SOUTH COAST	0.1006	0.9907
ALL OTHER BUSES - DIESEL (OBD)	SOUTH COAST	0.0759	1.0301
MOTOR HOMES (MH)	SOUTH COAST	2.1093	0.7132
Total On-Road Motor Vehicles		701.73	181.36
Other Mobile Sources			
AIRCRAFT	SOUTH COAST	37.0541	15.2761
TRAINS	SOUTH COAST	3.9777	17.6595
OCEAN GOING VESSELS	SOUTH COAST	1.3714	13.1882
COMMERCIAL HARBOR CRAFT	SOUTH COAST	2.6125	3.7041
RECREATIONAL BOATS	SOUTH COAST	86.4411	4.8787
OFF-ROAD RECREATIONAL VEHICLES	SOUTH COAST	3.6753	0.0722
OFF-ROAD EQUIPMENT	SOUTH COAST	544.662	49.9524
FARM EQUIPMENT	SOUTH COAST	4.9233	2.0758
FUEL STORAGE AND HANDLING	SOUTH COAST	0	0
Total Other Mobile Sources		684.72	106.81
Total Mobile Sources		1386.45	288.17

Table S5. California's 2018 South Coast Air Basin CO and NO_x Emissions Inventory Totals



2018	CO (tons/day)	Percent of Total CO	NOx (tons/day)	Percent of Total NOx
Total Stationary Sources	54.98	3.16	48.41	13.60
Total Areawide Sources	56.08	3.22	14.82	4.16
Total Natural Sources	243.81	14.00	4.46	1.25
Mobile Sources				
On-road Gasoline	677.38	38.90	69.87	19.63
On-road Diesel	24.35	1.40	111.49	31.33
Other Mobile	684.72	39.32	106.81	30.01
Total Mobile Sources	1386.45	79.62	288.17	80.98
Grand Total for South Coast (tons/day)	1741.32		355.86	
Grand Total for South Coast (moles/day)	56468520		7024367.0	
Total Inventory Molar NOx/CO ratio	0.124			

Table S6. How Molar Ratios are Calculated from the EMFAC2017 Output.



EMFAC2017 (v1.0.2) Emissions Inventory
Region Type: Air Basin
Region: SOUTH COAST
Calendar Year: 2018
Season: Summer
Vehicle Classification: EMFAC2011 Categories
Units: miles/day for VMT, tons/day for Emissions, 1000 gallons/day for Fuel Consumption

Region	Calendar Year	Vehicle Category	Model Year	Speed	Fuel	VMT	CO_RUNEX	NOx_RUNEX
SOUTH COAST	2018	LDA	Aggregated	20	GAS	15943834	26.7394124	1.325437872
SOUTH COAST	2018	LDT1	Aggregated	20	GAS	1601609	5.80930955	0.382634827
SOUTH COAST	2018	LDT2	Aggregated	20	GAS	5293119	12.5840715	0.92779679
SOUTH COAST	2018	LHD1	Aggregated	20	GAS	334227.5	0.67648173	0.112403456
SOUTH COAST	2018	LHD2	Aggregated	20	GAS	51931.45	0.08268494	0.018627359
SOUTH COAST	2018	MCY	Aggregated	20	GAS	114978.2	3.11216087	0.131973032
SOUTH COAST	2018	MDV	Aggregated	20	GAS	3449977	10.2119046	0.782326648
SOUTH COAST	2018	MH	Aggregated	20	GAS	14426.6	0.07327705	0.009083731
SOUTH COAST	2018	OBUS	Aggregated	20	GAS	13054.78	0.04893067	0.010840672
SOUTH COAST	2018	SBUS	Aggregated	20	GAS	7963.556	0.02600965	0.005200338
SOUTH COAST	2018	T6TS	Aggregated	20	GAS	63835.39	0.32038395	0.060354298
SOUTH COAST	2018	T7IS	Aggregated	20	GAS	393.9754	0.03086229	0.002786383
SOUTH COAST	2018	UBUS	Aggregated	20	GAS	41013.35	0.02049782	0.014442386
					Total tons/day		59.74	3.78
					Total grams/day		54240276	3435788
					Total moles/day		1937152	74691
					Molar NOx/CO			0.0386

Table S7. FEAT South Coast Air Basin On-road Campaigns and Statistics.

La Basin				Non-Diesel	Non-Diesel	Non-Diesel		Fleet		Molar
	Total			Mean	Mean	Molar		Molar		NOx/CO
Year/Location	Records	Non-Diesel	%Diesel	gCO/kg	gNOx/kg	NOx/CO	SEM	NOx/CO	SEM	%Difference
2018 Lynwood I-710	14302	14098	1.43	12.42	2.41	0.1028	0.0063	0.1149	0.0043	10.57
2018 Lynwood I-105	7724	7591	1.72	10.52	2.89	0.1435	0.0054	0.1592	0.0068	9.87
2018 West LA	19167	18844	1.69	11.15	2.28	0.1097	0.0065	0.1215	0.0068	9.68
2015 West LA	20100	19774	1.62	13.20	2.59	0.1093	0.0087	0.1185	0.0095	7.74
2013 West LA	26284	25750	2.03	16.61	3.09	0.0966	0.0035	0.1098	0.0049	11.97
2010 Van Nuys	12701	12540	1.27	19.58	3.82	0.0966	0.0061	0.1032	0.0074	6.45
2008 West LA	17866	17557	1.73	21.66	5.43	0.1266	0.0111	0.1364	0.0120	7.19
2005 West LA	19581	19102	2.45	27.58	4.79	0.0865	0.0031	0.0956	0.0035	9.56
2003 West LA	20176	19650	2.61	43.36	6.57	0.0741	0.0021	0.0801	0.0019	7.44
2001 West LA	20234	19614	3.06	55.07	8.43	0.0727	0.0029	0.0783	0.0033	7.15
1999 West LA	18899	18540	1.90	72.03	10.04	0.0647	0.0068	0.0672	0.0071	3.58
2001 Riverside	19783	19039	3.76	49.61	8.07	0.0787	0.0018	0.0861	0.0021	8.68
2000 Riverside	23285	22509	3.33	62.65	8.67	0.0641	0.0023	0.0688	0.0025	6.94
1999 Riverside	18740	17998	3.96	68.17	7.41	0.0508	0.0024	0.0554	0.0025	8.30
1999 LA710/91	12655	12372	2.24	66.69	9.35	0.0585	0.0018	0.06164	0.00177	5.04
									Average	8.01

Table S8. Example Calculation of EMFAC2017 Mean gCO/kg of Fuel for 2018 LDA Type Vehicles Age Normalized to Match the On-road West Los Angeles Gasoline Passenger Fleet.

EMFAC2017 (v1.0.2) Emissions Inventory							
Region Type: Air Basin							
Region: SOUTH COAST							
Calendar Year: 2018							
Season: Annual							
Vehicle Classification: EMFAC2011 Categories							
Units: miles/day for VMT, trips/day for Trips, tons/day for Emissions, 1000 gallons/day for Fuel Consumption							
Model Year	EMFAC LDA CO	EMFAC LDA Fuel	EMFAC LDA Fuel kg/day	EMFAC LDA gCO/Day	EMFAC LDA gCO/kg	West LA Gas Pass Fraction	EMFAC Mean CO Fraction
1974	2.077	2.501	7077	1885554	266.43	0.001056	0.28132
1975	0.685	1.429	4043	621547	153.73	0	0
1976	0.658	1.347	3813	597580	156.72	8.8E-05	0.013789
1977	0.692	1.495	4231	628178	148.47	0.000176	0.026127
1978	0.851	1.845	5222	772336	147.91	8.8E-05	0.013015
1979	0.999	2.204	6238	906930	145.38	8.8E-05	0.012792
1980	0.530	1.460	4132	481162	116.45	0	0
1981	0.566	1.468	4154	514208	123.78	8.8E-05	0.010891
1982	0.594	1.566	4432	539750	121.79	0	0
1983	0.645	1.803	5103	585339	114.71	0	0
1984	0.953	2.960	8377	865077	103.27	0.000176	0.018173
1985	0.974	3.968	11229	884026	78.72	8.8E-05	0.006927
1986	1.183	4.564	12915	1074008	83.16	0.000176	0.014635
1987	1.500	5.790	16385	1362192	83.14	0.000352	0.02926
1988	1.710	6.593	18659	1552241	83.19	0.000616	0.051238
1989	2.470	9.539	26996	2242939	83.08	0.000704	0.058484
1990	3.588	13.824	39123	3257913	83.27	0.002552	0.212489
1991	4.563	17.559	49691	4142957	83.37	0.002376	0.198075
1992	4.689	18.037	51044	4257386	83.41	0.0022	0.183473
1993	5.889	21.197	59988	5347265	89.14	0.002992	0.266673
1994	7.676	27.700	78392	6969426	88.90	0.004223	0.375488
1995	11.083	40.204	113777	10063107	88.45	0.004927	0.43581
1996	13.455	44.041	124636	12216926	98.02	0.004839	0.474363
1997	15.318	63.445	179548	13909016	77.47	0.006511	0.504404
1998	14.729	81.181	229742	13374218	58.21	0.010911	0.635155
1999	12.332	101.096	286103	11197877	39.14	0.01575	0.616449
2000	9.399	143.957	407398	8534141	20.95	0.01663	0.348364
2001	10.198	164.348	465106	9260030	19.91	0.017334	0.34511
2002	11.578	192.857	545785	10512871	19.26	0.02015	0.38812
2003	12.927	238.305	674404	11738004	17.41	0.024637	0.428808
2004	5.330	261.768	740803	4839503	6.53	0.029564	0.193138
2005	6.223	319.568	904377	5650410	6.25	0.038187	0.238589
2006	5.488	355.150	1005075	4983398	4.96	0.038275	0.189778
2007	6.618	397.153	1123943	6009151	5.35	0.050418	0.269559
2008	5.904	345.967	979086	5360754	5.48	0.041003	0.224503
2009	4.575	291.160	823981	4154021	5.04	0.032908	0.165903
2010	5.311	332.909	942133	4822137	5.12	0.040827	0.208966
2011	5.498	359.110	1016281	4992395	4.91	0.041795	0.205314
2012	7.771	477.349	1350899	7056384	5.22	0.062209	0.324945
2013	9.525	591.159	1672979	8648665	5.17	0.073559	0.380273
2014	10.167	637.639	1804518	9231821	5.12	0.080422	0.411437
2015	12.773	781.295	2211064	11598252	5.25	0.099956	0.524324
2016	12.526	751.783	2127546	11373948	5.35	0.099692	0.532958
2017	13.983	819.454	2319054	12696314	5.47	0.095996	0.52556
2018	14.315	817.813	2314410	12997642	5.62	0.03546	0.199141
	Age Normalized EMFAC2017 LDA Mean gCO/kg of fuel						10.54

Table S9. Example Calculation of EMFAC2017 Fleet Composite NO_x Emission Factors for 2018.

EMFAC2017 (v1.0.2) Emissions Inventory													
Region Type: Air Basin													
Region: SOUTH COAST													
Calendar Year: 2018													
Season: Annual													
Vehicle Classification: EMFAC2011 Categories													
Units: miles/day for VMT, trips/day for Trips, tons/day for Emissions, 1000 gallons/day for Fuel Consumption													
Gasoline only and aggregated speed. CA RFGII density assumed to be 0.75 g/ml													
	LDA	LDA	LDT1	LDT1	LDT2	LDT2	MDV	MDV					Emissions
	NOx		NOx		NOx		NOx		Fuel/Day	Fuel/Day	NOx		Factor
Model Year	RUNEX	Fuel	RUNEX	Fuel	RUNEX	Fuel	RUNEX	Fuel	1000's gal	Kilograms	Tons/Day	gNOx/Day	gNOx/kg of Fuel
1974	0.184	2.501	0.063	0.785	0.148	1.835	0.112	1.369	6.491	18433.684	0.506	459802.855	24.944
1975	0.110	1.429	0.023	0.293	0.085	1.081	0.125	1.508	4.311	12243.204	0.342	310988.555	25.401
1976	0.071	1.347	0.019	0.365	0.038	0.723	0.205	1.965	4.400	12497.048	0.333	302292.512	24.189
1977	0.057	1.495	0.026	0.497	0.026	0.504	0.397	3.848	6.345	18018.902	0.506	459318.164	25.491
1978	0.070	1.845	0.036	0.704	0.037	0.727	0.133	2.731	6.007	17058.901	0.276	250303.480	14.673
1979	0.084	2.204	0.029	0.746	0.035	0.704	0.131	2.737	6.392	18152.980	0.279	253547.072	13.967
1980	0.047	1.460	0.024	0.616	0.021	0.416	0.052	1.069	3.562	10115.330	0.144	130493.509	12.901
1981	0.049	1.468	0.030	0.878	0.024	0.642	0.025	0.648	3.636	10326.209	0.128	115928.824	11.227
1982	0.053	1.566	0.039	1.094	0.023	0.602	0.026	0.664	3.927	11152.244	0.140	127278.916	11.413
1983	0.058	1.803	0.061	1.177	0.032	0.864	0.023	0.766	4.611	13094.318	0.174	158179.668	12.080
1984	0.090	2.960	0.129	2.544	0.055	1.525	0.052	1.314	8.343	23694.477	0.326	295911.568	12.489
1985	0.093	3.968	0.212	4.156	0.063	1.750	0.049	1.704	11.578	32881.648	0.417	378411.216	11.508
1986	0.113	4.564	0.359	6.955	0.099	2.433	0.059	1.908	15.859	45039.102	0.630	571980.624	12.700
1987	0.143	5.790	0.356	6.951	0.113	2.813	0.060	1.963	17.517	49747.870	0.672	610589.550	12.274
1988	0.163	6.593	0.341	6.970	0.155	4.034	0.087	2.850	20.448	58072.613	0.745	676526.748	11.650
1989	0.234	9.539	0.331	9.679	0.232	6.052	0.120	3.907	29.177	82863.791	0.917	832812.191	10.050
1990	0.339	13.824	0.186	7.881	0.287	7.566	0.133	4.381	33.652	95572.318	0.944	857209.563	8.969
1991	0.431	17.559	0.219	9.461	0.407	10.966	0.137	4.520	42.505	120715.031	1.193	1083584.090	8.976
1992	0.443	18.037	0.200	8.718	0.399	10.845	0.184	6.109	43.709	124132.911	1.226	1113619.878	8.971
1993	0.556	21.197	0.263	10.600	0.571	14.129	0.258	7.411	53.337	151476.425	1.649	1496954.933	9.882
1994	0.720	27.700	0.276	13.336	0.685	17.209	0.461	13.315	71.560	203230.492	2.143	1945970.833	9.575
1995	1.040	40.204	0.283	13.573	0.719	24.669	0.513	17.021	95.467	271127.301	2.555	2319720.730	8.556
1996	1.269	44.041	0.322	14.785	0.787	25.857	0.381	17.097	101.780	289056.381	2.760	2506131.080	8.670
1997	1.432	63.445	0.318	19.491	1.022	44.326	0.589	26.385	153.648	436358.994	3.362	3052248.257	6.995
1998	1.354	81.181	0.320	26.094	0.974	55.427	0.614	32.764	195.466	555124.494	3.262	2961864.169	5.335
1999	1.084	101.096	0.200	24.313	0.880	72.167	0.951	61.515	259.092	735821.623	3.115	2828594.319	3.844
2000	0.715	143.957	0.120	28.854	0.774	108.438	0.933	85.531	366.781	1041656.654	2.543	2308590.843	2.216
2001	0.779	164.348	0.132	33.252	0.749	110.078	0.957	118.645	426.324	1210758.913	2.618	2377186.842	1.963
2002	0.889	192.857	0.124	32.198	0.787	119.010	1.242	158.322	502.386	1426777.530	3.043	2762651.328	1.936
2003	0.995	238.305	0.074	21.045	0.981	145.121	1.237	186.172	590.643	1677427.351	3.286	2984130.124	1.779
2004	0.212	261.768	0.013	17.720	0.120	172.440	0.310	207.198	659.126	1871917.931	0.655	595115.254	0.318
2005	0.260	319.568	0.008	10.782	0.125	185.299	0.231	180.929	696.578	1978281.810	0.623	566121.155	0.286
2006	0.242	355.150	0.011	18.121	0.102	161.207	0.048	177.312	711.790	2021482.276	0.403	365892.555	0.181
2007	0.298	397.153	0.018	27.631	0.103	167.146	0.092	176.958	768.887	2183639.334	0.511	463772.889	0.212
2008	0.260	345.967	0.030	46.032	0.083	132.747	0.068	130.306	655.051	1860346.109	0.442	401414.944	0.216
2009	0.211	291.160	0.022	34.449	0.052	78.651	0.028	51.286	455.546	1293749.564	0.313	284201.795	0.220
2010	0.245	332.909	0.011	16.302	0.084	136.531	0.036	70.690	556.433	1580268.814	0.375	340868.216	0.216
2011	0.253	359.110	0.009	14.939	0.108	172.103	0.052	98.793	644.945	1831643.723	0.422	383562.865	0.209
2012	0.348	477.349	0.017	27.240	0.100	162.788	0.050	96.952	764.329	2170695.333	0.516	468292.148	0.216
2013	0.422	591.159	0.025	39.026	0.145	217.689	0.058	104.306	952.179	2704189.666	0.649	589343.354	0.218
2014	0.445	637.639	0.028	45.891	0.144	216.765	0.093	168.939	1069.233	3036623.016	0.710	644903.976	0.212
2015	0.528	781.295	0.064	108.154	0.177	267.082	0.107	195.308	1351.840	3839224.433	0.877	796365.803	0.207
2016	0.503	751.783	0.062	105.103	0.169	255.396	0.127	232.058	1344.340	3817926.241	0.861	781769.766	0.205
2017	0.548	819.454	0.057	96.710	0.217	317.137	0.130	230.163	1463.464	4156237.290	0.951	863317.787	0.208
2018	0.510	817.813	0.051	93.400	0.207	305.134	0.118	211.430	1427.777	4054886.673	0.886	804312.523	0.198

Calculation of emission deterioration rates from the on-road and EMFAC2017 predicted emission factors.

Assuming that emissions deterioration can be modeled as a linear process we plot the fuel specific NO_x emission factors for the gasoline fleet versus vehicle age at the time of measurement for each model year. This creates a graph for each model year of emissions versus time (see Figure S1) whose data is then fit with a best fit straight line. The slope of this line is equal to that model year's emissions deterioration rate in gNO_x/kg of Fuel/Year.

Because of the lack of on-road NO_2 measurements prior to 2008 for the emission deterioration rate calculations we have chosen for consistency to only use the NO measurements for the eight West Los Angeles measurement campaigns (1999, 2001, 2003, 2005, 2008, 2013, 2015 and 2018). However, to compare NO_x emissions with the model we have converted the grams of NO to grams of NO_2 using the molecular weights of each species (30 and 46 g/mole; grams NO_2 = grams NO * 46/30) and plotting the data as gNO_x/kg of fuel. Because we have restricted these calculations to only the gasoline fleet the contribution of NO_2 to the total NO_x emissions is not significant. For the EMFAC2017 gasoline fleet we have created fuel weighted composite running exhaust emission factors that include the model vehicle classes LDA, LDT1, LDT2 and MDV (see Table S9 for an example) for each measurement year. Model years older than 2001 have a data point from each campaign while that number decreases in newer model years until the 2009 - 2013 model years have only the minimum necessary to fit a straight line, 3 data points from the 2013, 2015 and 2018 measurements. This results in significantly larger uncertainties in the slope estimates for the on-road measurements for these model years.

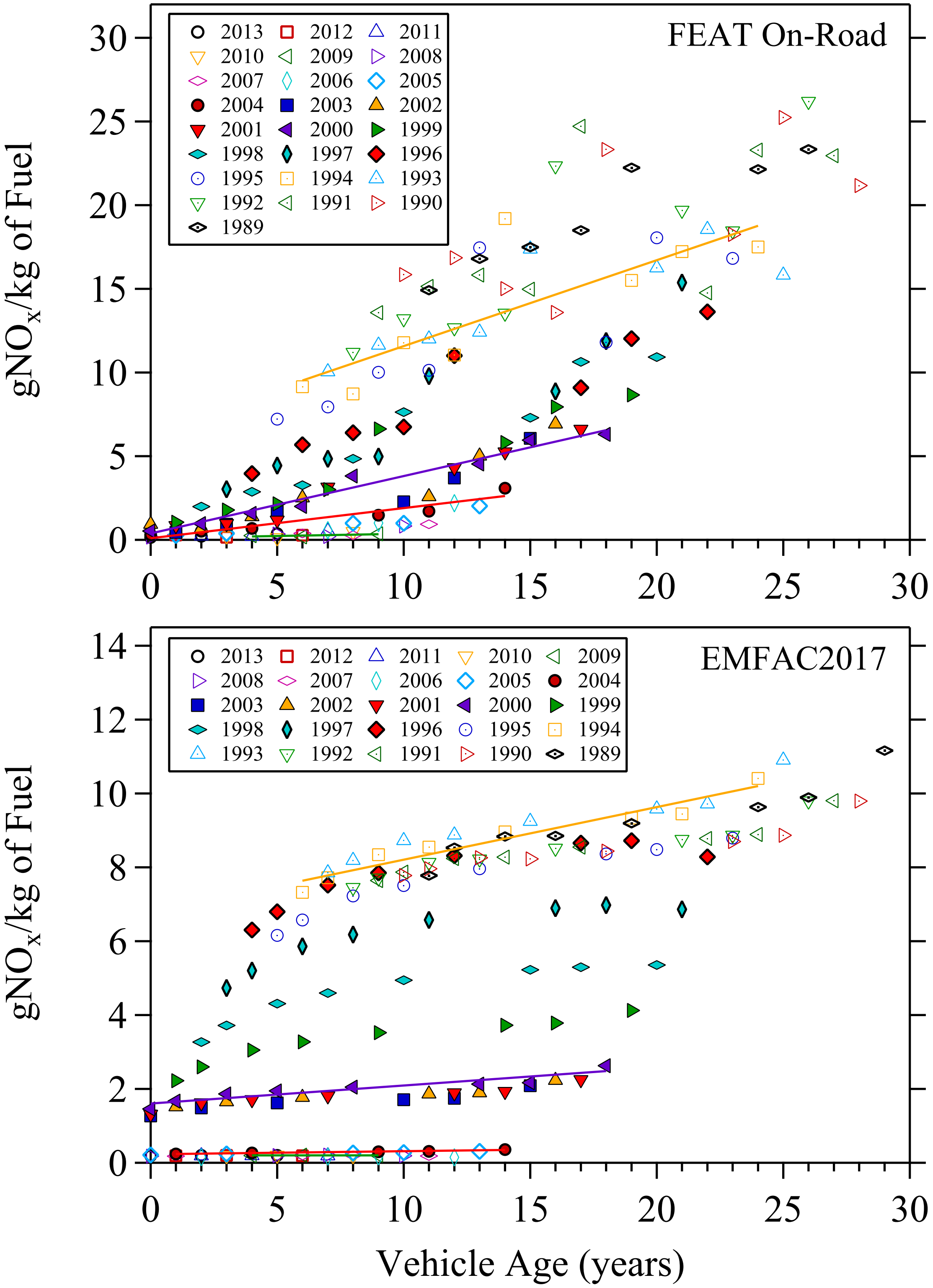


Figure S1. Fuel specific NO_x emission for the gasoline fleet versus vehicle age for 2013 - 1989 model year groupings for the FEAT on-road measurements (top) and EMFAC2017 emission factor predictions (bottom). Best fit lines are plotted for the 2009 (green), 2004 (red), 2000 (purple) and 1994 (orange) model years.

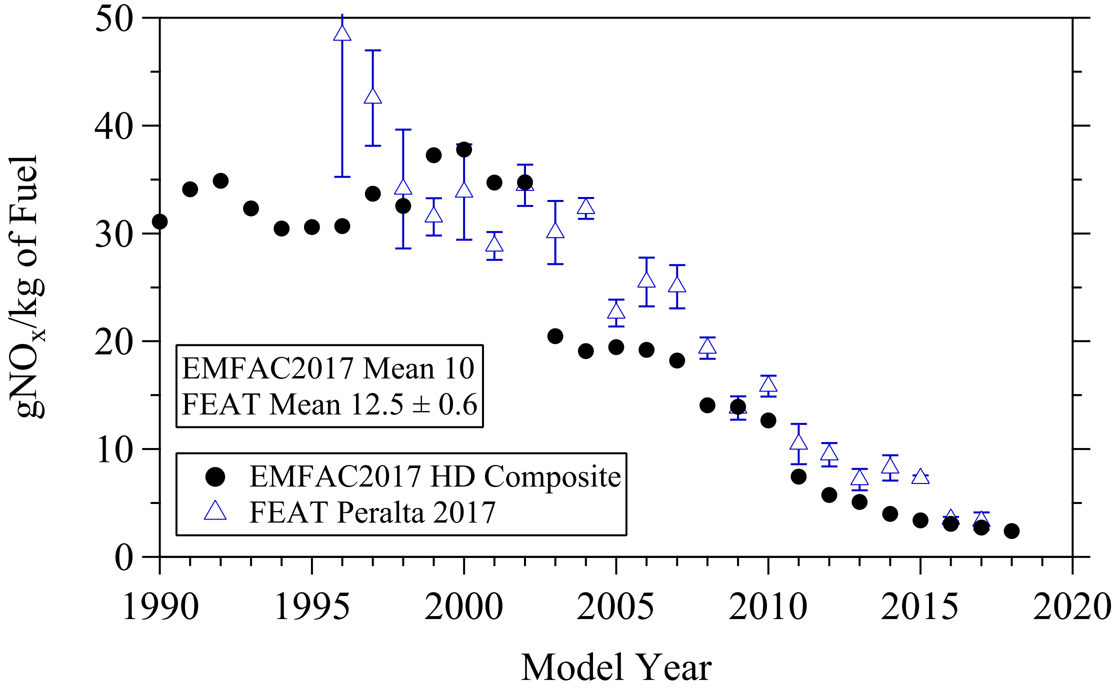


Figure S2. Fuel specific NO_x emissions versus model year for heavy-duty diesel trucks measured in 2017 at the Peralta weigh station on SR 91 in the Anaheim Hills and EMFAC2017 annual running exhaust emission factors with aggregated speed for a fuel weighted heavy-duty diesel composite using all T6 vehicle types with gvwr>26,000 lbs. and all T7 vehicle types except T7 Ag. ULSD density assumed to be 0.86g/ml. Uncertainties are standard error of the mean determined from the daily measurements. EMFAC2017 mean emissions have been calculated using the model year distribution of the Peralta weigh station on-road fleet.

Table S10. Calculation of 2018 South Coast Air Basin Gasoline and Diesel Fuel Use.

Year	Basin Gasoline (kg/day)	State Total Gasoline[b] (kg/day)	Basin Fraction of State Total Gasoline	Basin Diesel (kg/day)	State Total Diesel[b] (kg/day)	Basin Fraction of State Total Diesel
2011	(4.5 ± 0.3) x 10^7[a]	1.1 x 10^8	0.4	(6.8 ± 0.7) x 10^6[a]	2.0 x 10^7	0.34
2012	(4.5 ± 0.3) x 10^7[a]	1.1 x 10^8	0.4	(6.8 ± 0.7) x 10^6[a]	2.0 x 10^7	0.34
2013	(4.5 ± 0.3) x 10^7[a]	1.1 x 10^8	0.4	(7.1 ± 0.7) x 10^6[a]	2.1 x 10^7	0.34
2014	(4.6 ± 0.3) x 10^7[a]	1.1 x 10^8	0.4	(7.2 ± 0.7) x 10^6[a]	2.2 x 10^7	0.34
2018	(4.8 ± 0.3) x 10^7[c]	1.2 x 10^8	0.4	(8.1± 0.7) x 10^6[c]	2.4 x 10^7	0.34

[a] Data from Hassler et al., uncertainties are reported as absolute error.

[b] State gasoline sales minus aviation gasoline derived from annual sales. Source: California Department of Tax and Fee Administration (https://www.cdtfa.ca.gov/taxes-and-fees/spftrpts.htm).

[c] South Coast Air Basin daily fuel consumption using basin fraction calculated from Hassler et al.