<u>Supporting Information For:</u>

Long-Term Trends in Motor Vehicle Emissions in U.S. Urban Areas

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TABLE S1. Summary of On-road Studies for Gasoline CO Run Exhaust Emission Factors

Location	Type	Date	CO EF [g/kg fuel]	Mean Age [y][a]	Reference
Chicago, IL (Central Ave)	Remote Sensing	Aug/89	154.5	6.1	Zhang et al. (*1*)
		Oct/90	133.3	5.5	
		Jun/92	127.8	6.4	
Chicago, IL (Arlington Heights)	Remote Sensing	Sep/97	55.8	5.0	Bishop et al. (*2*)
		Sep/98	48.3	5.1	
		Sep/99	44.2	5.1	
		Sep/00	32.5	5.2	
		Sep/02	28.1	5.3	
		Sep/04	21.3	5.5	
		Sep/06	15.8	5.7	
Phoenix, AZ	Remote Sensing	Nov/99	41.9	5.9	Bishop et al. (*2*)
		Nov/00	35.5	5.7	
		Nov/02	28.8	5.5	
		Nov/04	24.1	5.5	
		Nov/06	14.4	5.6	
Denver, CO (Speer Blvd)	Remote Sensing	May/89	170.3	6.6	Bishop et al. (*3*)
		Oct/91	116.1	6.3	Bishop et al. (*4*)
		Apr/92	116.5	6.4	
		Jul-Aug/96	84.4	7.0	
		Dec/02[b]	34.4	6.5	

Location	Type	Date	CO EF [g/kg fuel]	Mean Age [y][a]	Reference
Denver, CO (6th Ave)	Remote Sensing	Dec/95-Jan/96	73.8	6.8	Stedman et al. (*5*)
		Dec/96-Jan/97	67.9	6.7	Stedman et al. (*6*)
		Jan/99	59.7	6.6	Bishop et al. (*2*)
		Jan/00	57.7	6.6	
		Jan/01	46.6	6.4	
		Jan/03	46.5	6.6	
		Jan/05	30.4	6.9	
		Jan-Feb/07	25.5	7.2	
Oakland, CA (Caldecott)	Tunnel	Aug/94	105.7	6.4[c]	Kirchstetter et al. (*7*)
		Jul-Aug/95	94.6	6.3	Kean et al. (*8*)
		Jul-Aug/96	74.3	6.5	
		Jul-Aug/97	75.7	6.7	
		Jul-Aug/99	52.0	6.4[c]	Ban-Weiss et al. (*9*)
		Jul-Aug/01	43.0	6.2	
		Jul-Aug/06	24.0	6.3	
		Jul/10	14.3	6.4[c]	Dallmann et al. (*10*)
San Jose, CA	Remote Sensing	Oct/99	49.6	6.9	Bishop et al. (*11*)
		Mar/08	16.9	7.6	
Los Angeles, CA	Remote Sensing	Dec/89	202.1	7.7	Lawson et al. (*12*)
		May-Jun/91	146.4	6.5	Singer et al. (*13*)
		May-Oct/97	108.1	6.6	Singer et al. (*14*)

Location	Type	Date	CO EF [g/kg fuel]	Mean Age [y][a]	Reference
W Los Angeles, CA	Remote Sensing	Nov/99	69.6	7.2	Bishop et al. (*2*)
		Oct/01	56.9	7.3	
		Oct/03	44.3	7.2	
		Oct/05	28.1	6.9	
		Mar/08	21.1	7.0	Bishop et al. (*11*)
Van Nuys, CA	Remote Sensing	Aug/10	20.9	9.1	Bishop et al. (*15*)
	Tunnel	Sep/93	175.7	7.3	Fraser et al. (*16*)
		1995	120.0	11.2	Gertler et al. (*17*)
		Aug/10	21.3	9.1	Fujita et al. (*18*)
Riverside, CA	Remote Sensing	Jun-Jul/99	70.3	7.1	Pokharel et al. (*19*)
		May-Jun/00	65.6	7.3	
		Jun/01	50.9	7.0	

Notes:

a. Age [y] = CY – mean vehicle fleet MY, where CY = Calendar Year and MY = Model Year.
b. This dataset can be found at: http://www.feat.biochem.du.edu/light_duty_vehicles.html.
c. The mean model year was not reported for these studies. Age was calculated as the average of study years where fleet age information was available.

TABLE S2. Summary of On-road Gasoline CO Run Exhaust Emission Factor Regression[a,b]

	Coef.	s.e.	t value[c]
β_0	118.7	11.5	10.3
β_1	0.395	0.046	8.5
β_2	-14.26	0.85	-16.7
β_3	5.10	1.65	3.1
β_4	-2.28	0.58	-3.9
β_5	33.7	6.6	5.1

Notes:

a. Model: $Y_{CO,EF}$ (g/kg fuel) = $\beta_0 + \beta_1*(CY - 1990)^2 + \beta_2*(CY-1990) + \beta_3*Age + \beta_4*CA + \beta_5*CA*(CY-1990)$, where Age [y] = CY – mean vehicle fleet MY, CA = (0: US, 1: CA).

b. $R^2 = 0.95$.

c. All coefficients are statistically significant to the 99% confidence level.

TABLE S3. Summary of On-road Diesel CO and NMHC Run Exhaust Emission Factor Regressions

	Coef.	s.e.	t value
CO EF[a]:			
β_0	19.25	1.58	12.2[c]
β_1	-0.470	0.123	-3.8[c]
NMHC EF[b]:			
β_0	2.083	0.488	4.3[c]
β_1	0.000423	0.035	0.01[d]

Notes:

a. CO Model: $Y_{CO,EF}$ (g/kg fuel) = $\beta_0 + \beta_1$*(CY – 1990); $R^2 = 0.62$.

b. HC Model: $Y_{HC,EF}$ (g/kg fuel) = $\beta_0 + \beta_1$*(CY – 1990); $R^2 < 0.01$.

c. Coefficients are statistically significant to the 99% confidence level.

d. Coefficient is not statistically significant.

TABLE S4. Summary of Gasoline NMHC/CO Ratios from Summertime Ambient Studies

Year	Data	NMHC/CO Ratio [g/g][a,b]	Comments
1987	SCAQS network N. Main site	0.126 ± 0.024 (95% CI) (N=57, r=0.82)	Over-constrained CMB[c,d]
1994-2001 (summer data)	PAMS network N. Main Site	0.093 ± 0.007 (95% CI) (N=357, r=0.83)	Over-constrained CMB[c,d], No trend observed
2002	Warneke et al. (*20*)	0.125 ± 0.054	Ensemble average of scaled up compound ratios[d-f]
2005	Baker et al. (*21*)	0.144 ± 0.044	Ensemble average of scaled up compound ratios[d-f]
2005	Gentner et al. (*22*)	0.104 ± 0.008 (95% CI)	Basic CMB[d]
2010	Warneke et al. (*23*)	0.119 ± 0.059	Ensemble average of scaled up compound ratios[d-f]

Notes:

a. The NMHC/CO ratios presented here include unburnt fuel, products of incomplete combustion in tailpipe exhaust, and fugitive (e.g. evaporative) emissions from vehicles and storage tanks.

b. Ratios are shown with standard deviations unless indicated otherwise.

c. Over-constrained CMB methods are described in detail in Gentner et al. (*24*) and used fuel data from the periods of interest. Tracer compounds used in this analysis were isopentane, 3-methylpentane, 3-methylhexane, methylcyclohexane, and isooctane (& n-butane in 1987).

d. Ambient ratios are scaled up assuming 24% of NMHC emissions are products of incomplete combustion (*7*).

e. Ambient ratios are scaled up assuming 24% of total gasoline-related emissions are non-tailpipe, which was derived from Gentner et al. (*22*) for 2005 and from CMB analysis in this study for 1987 and PAMS 1994-2001.

Each gasoline tracer is scaled to total NMHC by their presence in liquid fuel samples taken over this time period (*24, 25*). These analyses are more uncertain and likely an upper limit due to the contribution of other sources of hydrocarbon emissions.

TABLE S5. Summary of Los Angeles On-road Gasoline Fuel Consumption, Emission Factors, and Emissions

Calendar Year	Fuel[a-c] (10^9 L/yr)	Run CO EF[d,e] (g/kg)	Run CO Emissions[f,g] (t/d)	Cold CO Emissions[h] (t/d)	NMHC EF[i] (g/kg)	Tot NMHC Emissions[j] (t/d)
1990	21 ± 2	179 ± 11	7800 ± 1000	1500 ± 200	21.2 ± 3.6	1100 ± 210
1991	21 ± 2	163 ± 9	7000 ± 900	1400 ± 200	19.3 ± 3.3	990 ± 190
1992	21 ± 2	148 ± 8	6300 ± 800	1400 ± 200	17.6 ± 3.0	900 ± 170
1993	21 ± 2	134 ± 7	5700 ± 700	1300 ± 200	15.9 ± 2.7	830 ± 160
1994	21 ± 2	121 ± 7	5100 ± 600	1200 ± 200	14.3 ± 2.4	750 ± 140
1995	21 ± 2	109 ± 6	4600 ± 600	1200 ± 200	12.9 ± 2.2	690 ± 130
1996	21 ± 2	97 ± 6	4200 ± 500	1200 ± 200	11.5 ± 2.0	630 ± 120
1997	21 ± 2	86 ± 6	3800 ± 500	1100 ± 200	10.2 ± 1.7	580 ± 110
1998	22 ± 2	76 ± 5	3400 ± 400	1000 ± 200	9.0 ± 1.6	520 ± 100
1999	22 ± 2	66 ± 5	3000 ± 400	1000 ± 100	7.9 ± 1.4	470 ± 90
2000	23 ± 3	58 ± 5	2700 ± 400	860 ± 130	6.9 ± 1.2	420 ± 80
2001	23 ± 3	50 ± 5	2400 ± 400	780 ± 120	5.9 ± 1.1	370 ± 70
2002	24 ± 3	43 ± 5	2100 ± 300	710 ± 110	5.1 ± 1.0	330 ± 70
2003	24 ± 3	37 ± 5	1800 ± 300	640 ± 100	4.4 ± 0.9	290 ± 60
2004	24 ± 3	32 ± 5	1600 ± 300	570 ± 80	3.8 ± 0.8	260 ± 50
2005	24 ± 3	28 ± 5	1400 ± 300	500 ± 70	3.3 ± 0.8	230 ± 50
2006	24 ± 3	24 ± 5	1200 ± 300	430 ± 60	2.9 ± 0.7	190 ± 50
2007	24 ± 3	22 ± 5	1100 ± 300	380 ± 60	2.6 ± 0.7	170 ± 40
2008	23 ± 3	20 ± 5	930 ± 280	340 ± 50	2.3 ± 0.7	150 ± 40
2009	23 ± 3	18 ± 6	870 ± 290	320 ± 50	2.2 ± 0.8	140 ± 40
2010	23 ± 3	18 ± 7	850 ± 320	320 ± 50	2.1 ± 0.9	140 ± 40

Notes:

a. Fuel use from McDonald et al. (*26*).

b. Annual daily average shown here. To estimate weekday only, multiply by 1.01 (*27*).

c. Uncertainty is calculated as the propagation of errors in state-level fuel sales reports and spatial apportionment to the urban scale. State-level uncertainty was calculated as the difference between the state's share of national gasoline sales and total vehicle miles traveled. Apportionment error is assumed from speed differences between urban and rural areas. This is estimated at ~10% by comparing average CO_2 emission factors (g/mi) for the South Coast air basin using an urban and rural VMT-weighted speed profile from EMFAC.

d. Running exhaust emission factor shown here is derived from regression model of on-road studies. Uncertainty is denoted as 2σ.

e. The long-term CO emission factor trend is reflective of changes in summertime emissions (see text).

f. Emissions = Fuel x Emission Factor. Uncertainty calculated by error propagation.

g. Running emissions for a given calendar year reflect local vehicle mixes across model years.

h. Uncertainty is estimated as the seasonal variability in the start to running exhaust emission ratio from EMFAC.

i. Calculated as Run CO EF * $NMHC/CO_{ambient}$. Includes evaporative emissions in addition to running exhaust. Start emissions not included. Uncertainty denoted as 2σ, and includes standard error of the mean of ambient data.

j. Calculated as (Run + Cold CO emissions) * $NMHC/CO_{ambient}$. Start emissions are included.

TABLE S6. Summary of Los Angeles On-road Diesel Fuel Consumption, Emission Factors, and Emissions

Calendar Year	Fuel[a-c] (10^9 L/yr)	Run CO EF[d] (g/kg)	Run CO Emissions[e] (t/d)	NMHC EF[d,f] (g/kg)	Run NMHC Emissions[e,f] (t/d)
1990	2.3 ± 0.3	19 ± 3	100 ± 22	2.1 ± 1.1	11 ± 6
1991	2.3 ± 0.3	19 ± 3	98 ± 20	2.1 ± 1.1	11 ± 6
1992	2.4 ± 0.3	18 ± 3	98 ± 20	2.1 ± 1.0	11 ± 6
1993	2.3 ± 0.3	18 ± 3	95 ± 19	2.1 ± 0.9	11 ± 5
1994	2.4 ± 0.3	17 ± 2	96 ± 19	2.1 ± 0.9	12 ± 5
1995	2.6 ± 0.3	17 ± 2	100 ± 19	2.1 ± 0.8	12 ± 5
1996	2.7 ± 0.4	16 ± 2	102 ± 19	2.1 ± 0.7	13 ± 5
1997	2.8 ± 0.4	16 ± 2	103 ± 19	2.1 ± 0.7	14 ± 5
1998	2.9 ± 0.4	15 ± 2	101 ± 19	2.1 ± 0.7	14 ± 5
1999	3.0 ± 0.4	15 ± 2	102 ± 19	2.1 ± 0.6	14 ± 5
2000	3.1 ± 0.4	14 ± 2	103 ± 19	2.1 ± 0.6	15 ± 5
2001	3.1 ± 0.4	14 ± 2	100 ± 19	2.1 ± 0.6	15 ± 5
2002	3.2 ± 0.4	13 ± 2	97 ± 19	2.1 ± 0.6	15 ± 5
2003	3.1 ± 0.4	13 ± 2	93 ± 19	2.1 ± 0.6	15 ± 5
2004	3.3 ± 0.4	12 ± 2	95 ± 21	2.1 ± 0.6	16 ± 5
2005	3.5 ± 0.5	12 ± 2	96 ± 23	2.1 ± 0.7	17 ± 6
2006	3.5 ± 0.5	11 ± 3	92 ± 25	2.1 ± 0.7	17 ± 6
2007	3.5 ± 0.5	11 ± 3	89 ± 26	2.1 ± 0.7	17 ± 6
2008	3.2 ± 0.4	11 ± 3	78 ± 26	2.1 ± 0.8	15 ± 6
2009	2.9 ± 0.4	10 ± 3	68 ± 25	2.1 ± 0.9	14 ± 6
2010	2.9 ± 0.4	10 ± 4	65 ± 27	2.1 ± 0.9	14 ± 6

Notes:

a. Fuel use from McDonald et al. (*26*).
b. Annual daily average shown here. To estimate weekday only, multiply by 1.29 (*27*).
c. Uncertainty reported by McDonald et al. (*26*) for South Coast air basin, using an approach similar to on-road gasoline.
d. Running exhaust emission factor shown here is derived from regression model of on-road studies. Uncertainty is denoted as 2σ.
e. Emissions = Fuel x Emission Factor. Uncertainty calculated by error propagation.
f. Tailpipe emissions only. Evaporative emissions are <u>not</u> included.

TABLE S7. Summary of New York City and Houston On-road Gasoline Fuel Consumption, Emission Factors, and Emissions

	NYC			HOU		
Calendar Year	Fuel[a,b] (10^9 L/yr)	Run CO EF[c,d] (g/kg)	Run CO Emissions[e,f] (t/d)	Fuel[a,b] (10^9 L/yr)	Run CO EF[c,d] (g/kg)	Run CO Emissions[e,f] (t/d)
1990	17 ± 2	141 ± 9	5000 ± 600	5.4 ± 0.9	138 ± 9	1520 ± 270
1991	17 ± 2	127 ± 7	4400 ± 600	5.2 ± 0.8	125 ± 8	1320 ± 230
1992	16 ± 2	115 ± 7	3800 ± 500	5.2 ± 0.8	112 ± 7	1190 ± 210
1993	16 ± 2	103 ± 6	3400 ± 400	5.4 ± 0.9	100 ± 7	1120 ± 200
1994	17 ± 2	92 ± 6	3100 ± 400	5.0 ± 0.8	89 ± 6	920 ± 160
1995	17 ± 2	82 ± 5	2800 ± 400	5.2 ± 0.8	79 ± 6	850 ± 150
1996	17 ± 2	72 ± 5	2600 ± 300	5.6 ± 0.9	69 ± 6	800 ± 140
1997	18 ± 2	64 ± 5	2300 ± 300	5.7 ± 0.9	61 ± 5	710 ± 130
1998	18 ± 2	56 ± 5	2000 ± 300	6.2 ± 1.0	53 ± 5	670 ± 130
1999	18 ± 2	49 ± 5	1800 ± 300	6.0 ± 1.0	46 ± 5	570 ± 110
2000	18 ± 2	42 ± 5	1500 ± 200	6.1 ± 1.0	40 ± 5	500 ± 100
2001	18 ± 2	37 ± 5	1400 ± 200	6.5 ± 1.1	34 ± 5	450 ± 100
2002	18 ± 2	32 ± 5	1200 ± 200	6.9 ± 1.1	30 ± 5	420 ± 100
2003	20 ± 2	28 ± 5	1200 ± 200	7.0 ± 1.1	26 ± 5	380 ± 100
2004	20 ± 2	25 ± 6	1000 ± 300	6.8 ± 1.1	24 ± 6	330 ± 100
2005	20 ± 2	23 ± 6	930 ± 280	6.7 ± 1.1	22 ± 7	310 ± 100
2006	20 ± 2	22 ± 8	880 ± 330	6.9 ± 1.1	22 ± 8	300 ± 120
2007	20 ± 2	21 ± 9	860 ± 380	7.1 ± 1.1	21 ± 9	310 ± 140
2008	19 ± 2	21 ± 9	840 ± 370	7.4 ± 1.2	21 ± 9	330 ± 150
2009	19 ± 2	21 ± 9	800 ± 360	7.6 ± 1.2	21 ± 9	340 ± 150
2010	20 ± 2	21 ± 9	840 ± 380	7.7 ± 1.3	21 ± 9	340 ± 150

Notes:

a. Annual daily average shown here. To estimate weekday only, multiply by 1.01 (*27*).
b. See footnotes in Table S5 for details on how uncertainty was calculated.
c. Uncertainty denoted as 2σ, and based on running exhaust emission factor regression model.
d. The long-term CO emission factor trend is reflective of changes in summertime emissions (see text).
e. Emissions = Fuel x Emission Factor. Uncertainty calculated by error propagation.
f. Running emissions for a given calendar year reflect local vehicle mixes across model years.

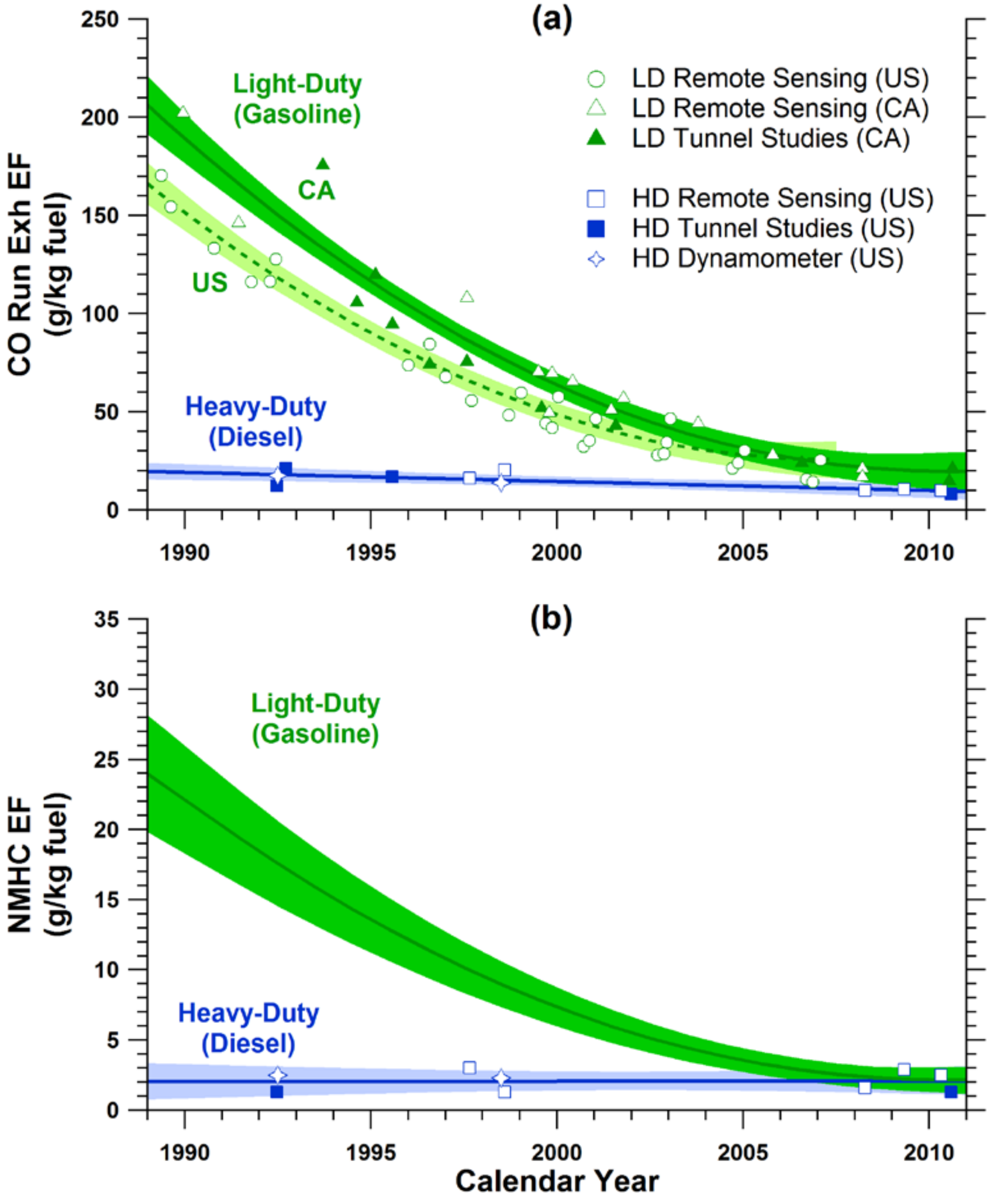

FIGURE S1. (a) Carbon monoxide running exhaust emission factor trends for light- and heavy-duty vehicles. Results are based on regression analyses of on-road studies (see text). Light-duty vehicles are shown separately for California and US. (b) NMHC emission factor trends for gasoline (running exhaust + evaporative) and diesel-powered vehicles (running exhaust). Light-duty NMHC emission factors are calculated as the product of CO emission factors in panel (a) and the NMHC/CO ratio derived from ambient analysis (see text). Start emission are not included. The heavy-duty NMHC emission factor trend is based on regression analysis. All uncertainties are shown as 95% confidence interval in both panels. See footnotes of Tables S4-S7 for basis of uncertainty.

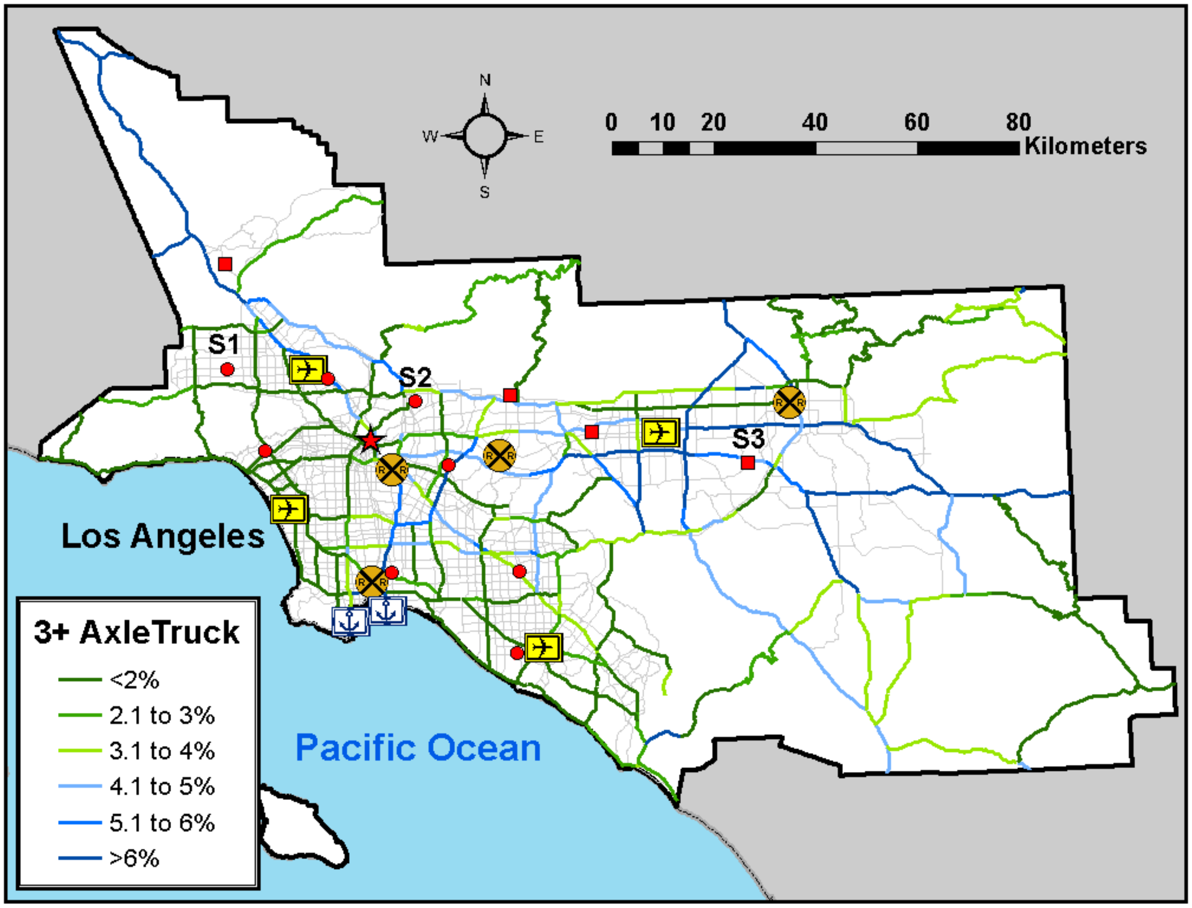

FIGURE S2. Map of South Coast air basin (shown in white) and location of ambient air monitoring sites. CO trends derived only from coastal locations (red circles). Ambient CO/NO_x ratios derived from coastal and inland locations (red circles + squares). The North Main site is part of the PAMS network (red star) and was used to estimate NMHC/CO molar emission ratios from motor vehicles. Ambient monitoring locations were selected to represent traffic emissions and based on their proximity to major highways. The spatial variability in vehicle fleet mix is also shown on the state highway system (*28*), as represented by the fraction of 3+ axle trucks to total vehicular traffic (a spatial surrogate for diesel fuel use). Roadway segments color coded green are more gasoline-dominated than average (~4% of VMT by 3+ axle trucks), and blue segments are more diesel-dominated. See Figure S11 for influence on ambient CO/NO_x (S1 = Reseda, S2 = Pasadena, and S3 = Rubidoux).

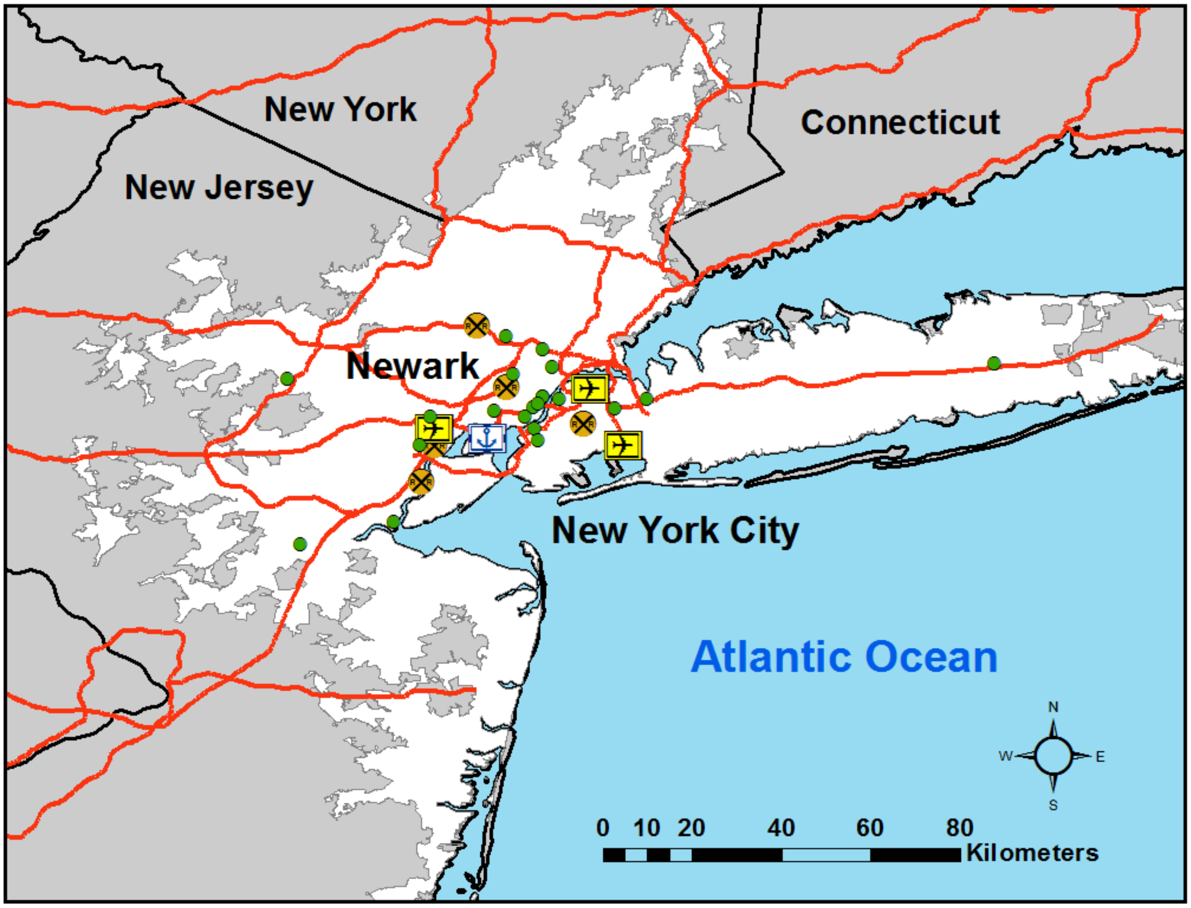

FIGURE S3. Map of New York City-Newark metropolitan area (shown in white) and location of ambient air monitoring sites (green circles). Monitoring sites are generally located near major highways. Monitors located on industrial land uses, in parks, or in areas which may not be representative of traffic emissions were excluded. Measurements are only available from 7-14 sites per year.

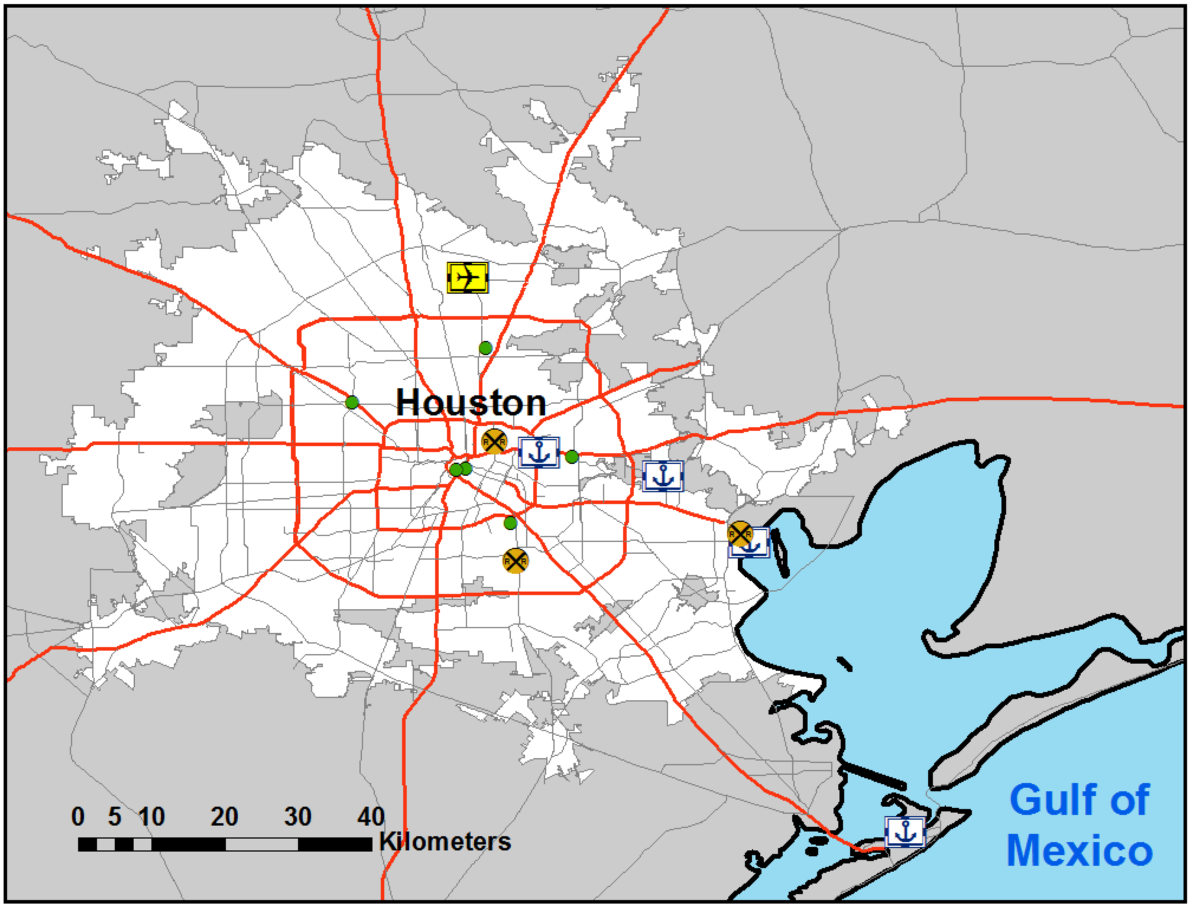

FIGURE S4. Map of Houston metropolitan area (shown in white) and location of ambient air monitoring sites (green circles). Monitoring sites are generally located near major highways. Monitors located in the major ship channel are excluded as these sites also have large industrial sources of emissions. Measurements are only available from 3-4 sites per year.

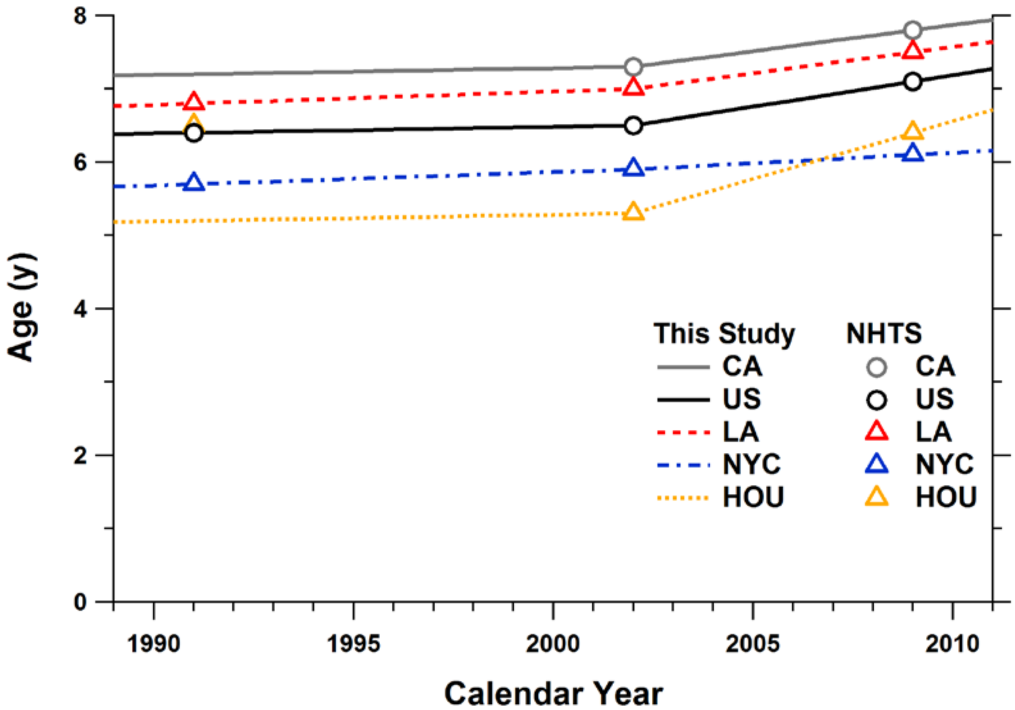

FIGURE S5. Mean light-duty vehicle fleet age for the U.S., California, Los Angeles, New York City, and Houston spatial domains. The mean age is derived from the National Household Travel Survey (*29*-*31*) and weighted by distance traveled. The fleet ages shown here are supplied to the CO emission factor regression for running exhaust for each of the domains specified. A data point for Houston in 1991 is treated as an outlier.

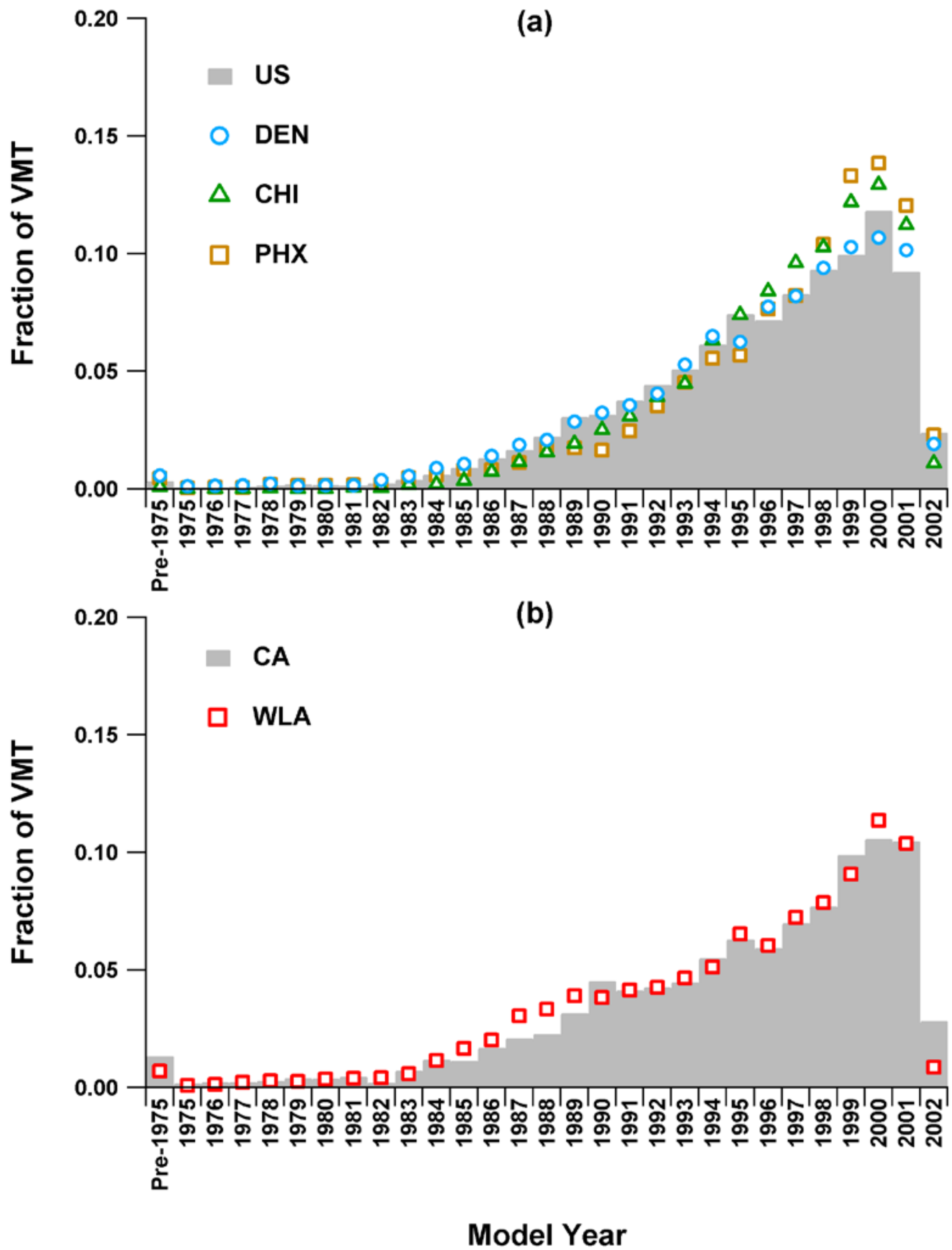

FIGURE S6. Comparison of light-duty vehicle age distributions between remote sensing and the (a) U.S. and (b) California vehicle fleets in calendar year 2001. The age distribution is weighted by vehicle miles traveled.

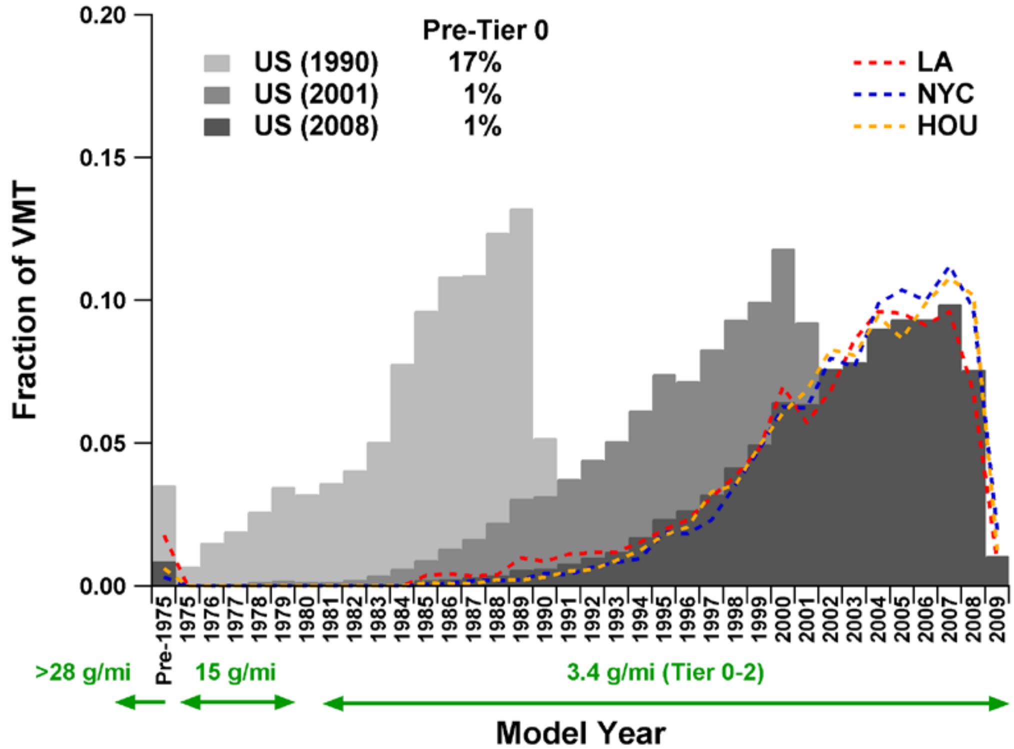

FIGURE S7. Vehicle age distribution of U.S., Los Angeles, New York City, and Houston vehicle fleets over time (*29*-*31*). Successive federal CO emission standards are shown for light-duty vehicles at bottom (*32*). Large changes in emission standards occurred prior to Tier 0 standards, which started with model year 1981.

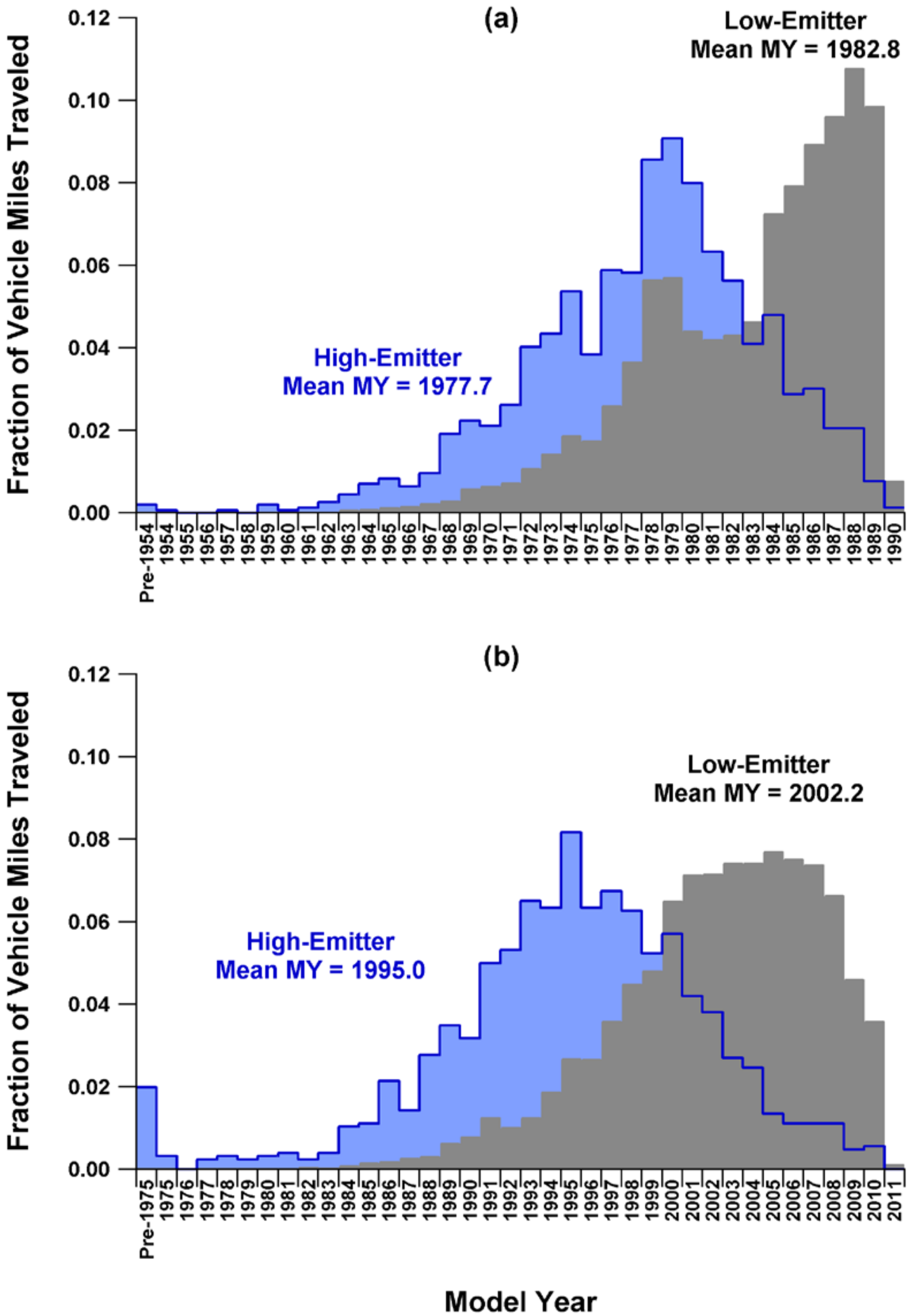

FIGURE S8. Vehicle age distributions of high and low emitting vehicles for running exhaust emissions of CO from remote sensing in Los Angeles for (a) 1989 and (b) 2010. High emitting vehicles are defined as the top 10% and low emitting as the bottom 90%.

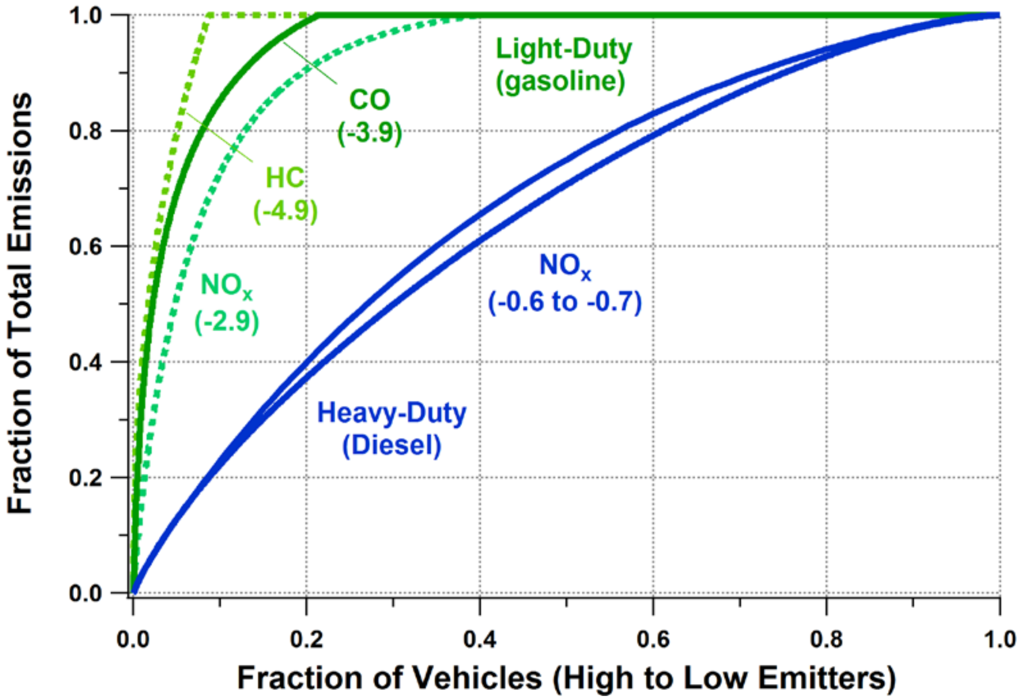

FIGURE S9. Distribution of running exhaust emissions for NMHC, CO, and NO_x in 2010. Light-duty measurements are for the same location in Los Angeles. Similar results for heavy-duty diesel NOx are shown based on remote sensing at Peralta weigh station in Anaheim (lower blue line) and tunnel measurements in Oakland (upper blue line).

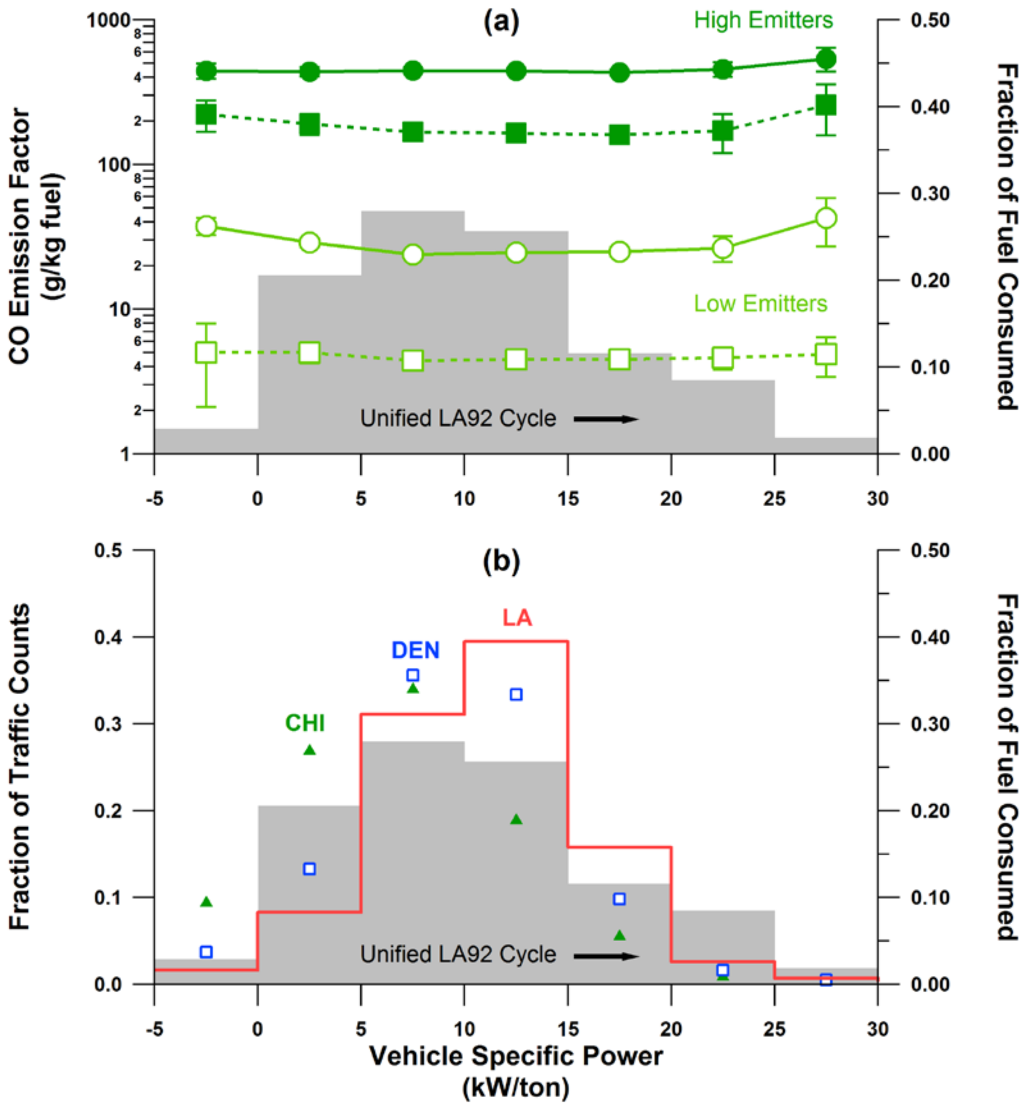

FIGURE S10. (a) Variability of CO emission factors with engine load at remote sensing from West Los Angeles. High-emitters are defined as the top 10% of vehicles. Vehicle specific power is calculated as VSP = 4.39*sin(slope)*v + 0.22*v*a + 0.0954*v + $0.0000272*v^3$, where slope is in degrees, v = velocity in mph, and a = acceleration mph/s (*33*). The remaining vehicles are labeled as low emitters. Emission factors for each subgroup are shown for the years 1999 (solid lines) and 2008 (dashed lines). (b) Distribution of engine loads for remote sensing in West Los Angeles, Denver, and Chicago as compared to the California Unified LA92 drive cycle.

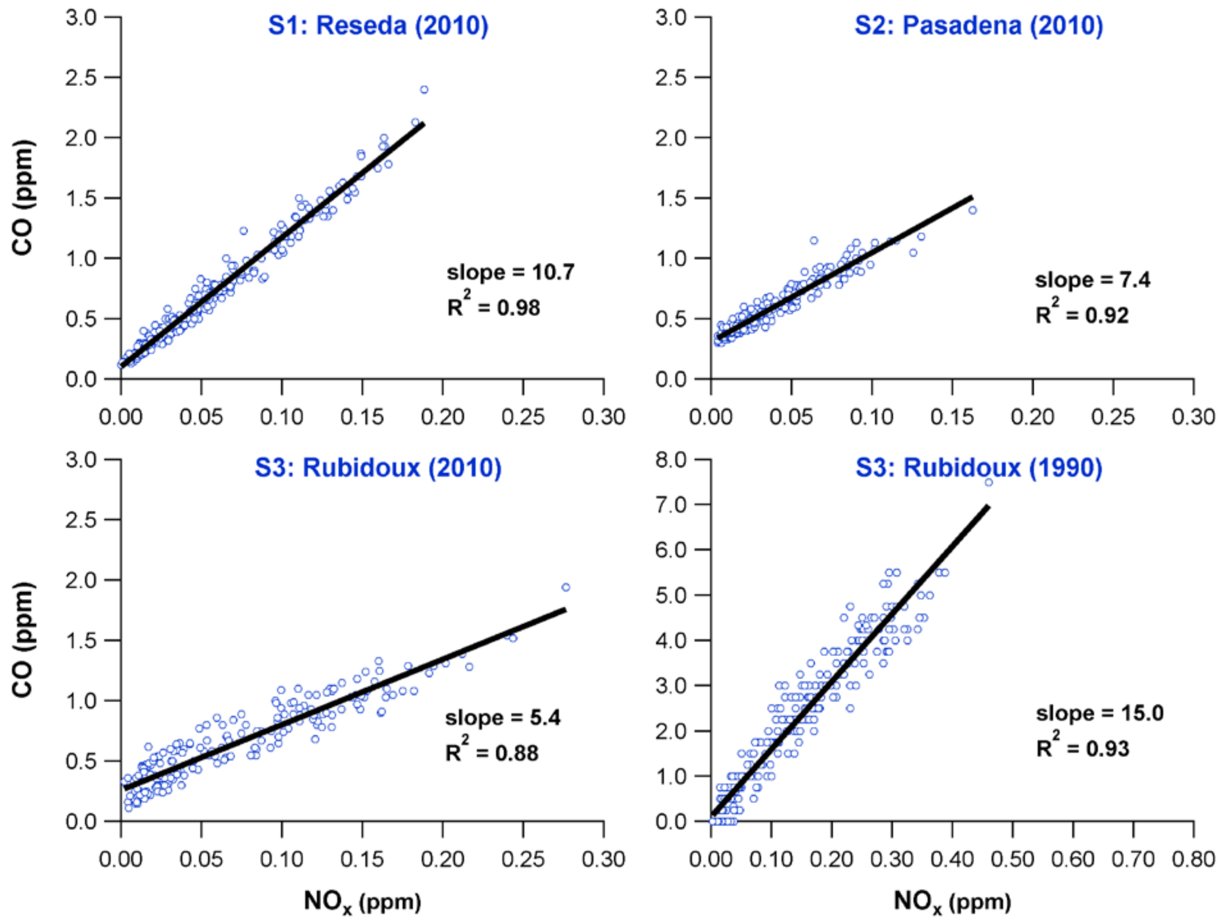

FIGURE S11. Examples of analysis used to determine ambient CO/NO_x ratios during morning peak hours (0500 to 0800 PST). Site locations can be found in Figure S2. The three sites represent the range of ambient ratios found spatially across the South Coast air basin for the year 2010. Rubidoux is shown for the years 1990 and 2010, showing the temporal change in CO/NO_x observed at a single site. Note that the scale for 1990 is larger in the absolute for CO and NO_x.

References

1. Zhang, Y.; Stedman, D. H.; Bishop, G. A.; Beaton, S. P.; Guenther, P. L., On-road evaluation of inspection/maintenance effectiveness. *Environ Sci Technol* **1996,** *30* (5), 1445-1450; DOI 10.1021/Es950191j.
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