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A Decade of On-road Emissions Measurements

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A multiyear, on-road emission measurement program carried out in the cities of Chicago, Illinois; Denver, Colorado; Los Angeles (LA), California; and Phoenix, Arizona shows large, fuel-specific tailpipe emissions reductions at all of the sites for carbon monoxide (CO), hydrocarbons (HC), and nitric oxide (NO). CO emissions decreased between 56% (Denver) and 71% (Chicago), HC emissions decrease between 27% (Phoenix) and 63% (Denver), and NO emissions have dropped between 48% (West LA) and 68% (Chicago). Three observed factors common to all of the sites are that the emission reductions are occurring in vehicles of all ages, that the influence of engine load on fuel-specific emissions, especially for CO and NO, is reduced and that fleet-averaged emission deterioration is near zero for model years newer than 2001 and older than 1990. These nationwide data sets imply that the majority of these on-road emissions reductions are the result of continued improvements in function and durability of vehicle emission control systems and that inspection and maintenance and fuel reformulation programs have only played a minor role.

Introduction

Since the early 1950s when A. J. Haagen-Smit first linked automobile exhaust with the formation of urban ozone, the study of vehicle exhaust emissions has provided an important foundation for developing public health policy in the United States (*1, 2*). Those policies have resulted in ever-shrinking national tailpipe and evaporative new vehicle emissions standards. Fuel reformulation mandates, state-controlled vehicle inspection and maintenance (I/M) programs, and other transportation control measures, such as high occupancy vehicle lanes, have also been implemented. These efforts have been primarily directed at reducing levels of carbon monoxide (CO) and ground-level ozone, a major component of urban smog that is produced by the photochemical reaction of nitrogen oxides (NO_x) and volatile organic hydrocarbons (VOCs). As of 2002, on-road vehicles were estimated to be the single largest source for these major atmospheric pollutants, contributing 82% of the CO, 45% of the VOCs, and 56% of the NO_x to the national emission inventory (*3*).

In the mid-1990s the Coordinating Research Council proposed the collection of on-road tailpipe emissions data from various groups with the purpose of monitoring the long-term on-road emission trends of the U.S. vehicle fleet (*4*). The idea was to establish comparable sites around the country where approximately 20 000 tailpipe measurements of CO, hydrocarbons (HC), NO, vehicle information, speed, and acceleration could be collected at the same location over regular intervals on fully warmed up vehicles using on-road vehicle remote sensing systems (*5*). The University of Denver began collecting measurements at a site in the Northwest suburbs of Chicago, Illinois in 1997 and in 1998 at sites in Denver, Colorado; Los Angeles (LA), California; and Phoenix, Arizona (*6*).

Experimental Section

Criteria for site selection in all of the cities required a single lane roadway, preferably with a positive grade; sufficient traffic volume to allow the collection of 20 000+ vehicle records in a week with no prospects for site reconstruction in the foreseeable future. All of the sites listed in Table 1 are curved interchange or on-ramps adjacent to major freeways. It should be noted that both Los Angeles and Phoenix required changes in the initial sampling locations. In Los Angeles, construction projects surrounding our Riverside measurement site made data collection after 2001 unfeasible, and the site in Phoenix was moved between the 1998 and 1999 measurements to a site with more positive acceleration patterns. Sampling was originally slated to be yearly, but beginning in 2001, it was decided that biannual measurements were sufficient. In addition to the four major cities, we have also explored additional cities to add to the list and have collected measurements in Omaha, Nebraska and Tulsa, Oklahoma. These two cities differ from the other sites in that their vehicle fleets have never been subject to any type of local I/M or fuel reformulation program. All of the 29 data sets including more than $^3/_4$ of a million measurements, and summary reports are available for download from our website at www.feat.biochem.du.edu.

The same University of Denver remote vehicle exhaust sensor (#3002) was used to collect all of the data sets listed in Table 1. The instrument consists of a source and detector unit consisting of a nondispersive infrared (IR) component for detecting CO, carbon dioxide (CO_2), and HC, and a dispersive ultraviolet (UV) spectrometer for measuring NO, arranged bistatically across the roadway, and has been fully described in the literature (*5, 7*). As a vehicle passes between the source and detector, its exhaust plume hopefully intersects with the collinear IR and UV beams, allowing absorption measurements of all the species. Because the path length of the plume is unknown and is highly variable from vehicle to vehicle, the remote sensor can only directly measure the ratios of CO, HC, and NO to exhaust CO_2. These ratios are constant for a given exhaust plume. With a small correction for the fact that an NDIR HC reading is about one-half of a total carbon FID reading, by doubling the HC/CO_2 ratio one can convert the measured ratios into fuel-specific emissions of grams of pollutant per kg of fuel (*5, 8*).

Quality assurance calibrations are performed as dictated in the field by the atmospheric conditions and traffic volumes. A puff of gas containing certified amounts of CO, CO_2, propane, and NO (Praxair) is released into the instrument's path, and the ratios are measured. The observed ratios are then used in the remote sensor's software to scale the measured emission ratios obtained from the passing vehicles. These calibrations account for day-to-day variations in instrument sensitivity and variations in ambient CO_2 levels caused by atmospheric pressure and instrument path length. Because propane is used to calibrate the instrument, all HC measurements reported by the remote sensor are as propane equivalents.

A freeze-frame video image of the license plate of each vehicle is recorded along with the emission measurements.

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TABLE 1. Summary of Measurement Locations, Collection Month, and Years

city	location/roadway grade (deg)	month	years data collected
Chicago, IL	Algonquin Rd. to EB I-290 (SH 53)/1.0°	September	1997–2000, 2002, 2004, 2006
Denver, CO	NB I-25 to WB Sixth Ave/4.6°	January	1999–2001, 2003, 2005, 2007[a]
LA/Riverside, CA	NB 91 to WB 60/4.4°	June/July	1999−2001
West LA, CA	SB La Brea Ave to EB I-10/2.0°	October	1999[b], 2001, 2003, 2005
Phoenix, AZ[c]	WB Sky Harbor Blvd. to SB 143/1.3°	November	1999, 2000, 2002, 2004, 2006
Omaha, NE	SB I-680 to EB Dodge Rd/2.7°	September	2002
	NB 84th St. to EB I-80/2.2°		2002
	NB I-680 to WB Dodge Rd/3.1°		2004
Tulsa, OK	WB US 64 to SB US 169/2.6°	September	2003

[a] Measurements funded by the University of Denver. [b] Measurements funded by the Steven and Michelle Kirsh Foundation. [c] Measurements were collected in 1998 at WB I-10 to NB US 143 that are not listed here.

The license plate information is used to obtain nonpersonal vehicle information from the local registration records. In addition to emission measurements, a pair of parallel infrared beams (Banner Industries) 6 ft apart and approximately 2 ft above the roadway is used to measure the speed and acceleration of the vehicles. Previous studies have shown that the remote sensor is capable of CO measurements that are correct to within ±5% of the values reported by an on-board gas analyzer and within ±15% for HC (*9, 10*). The NO channel has been estimated to have a detection limit ($\pm 3\vartheta$) of 25 ppm for NO, with an uncertainty of ±5% of the reading at higher concentrations (*7*).

Measurements at each site are generally collected during daylight hours (Phoenix is an exception where some measurements are collected before sunrise during the morning commute) and only during dry roadway conditions. An excess of data is collected at each site so that after the data reduction process, which eliminates invalid gas measurements, unreadable, and out-of-state plates (this typically eliminates 20−30% of the attempted measurements), a database results with approximately 20 000 records that contain at least valid emission measurements for CO and matching vehicle registration data. There is no statistically significant difference between the mean emissions of all valid measurements and of the plate-matched fleet.

Results and Discussion

The fuel-specific mean emissions for CO, HC, and NO are plotted in Figure 1 for Chicago, Denver, Los Angeles (2 sites), and Phoenix for each of the measurement years listed in Table 1. These means have been calculated using all of the valid emissions data (unless noted, the data were not required to include a valid speed and acceleration measurement) in each database and have not been filtered in regards to fuel type (gasoline or diesel), driving mode, vehicle type, or model year. The uncertainty bars plotted for West LA and Chicago are standard errors of the means calculated from each daily measurement mean and are representative of the magnitude of uncertainty for all of the sites. The mean HC values have been adjusted to compensate for a variable systematic offset in the measurements (*11*). The source of this offset, restricted to the HC channel, is thought to be a product of the optical alignment of the HC and reference IR channels combined with the very low HC signals from the lowest emitting HC vehicles today. This setup-specific offset results in the entire HC distributions zero-point being artificially increased or decreased. Site-by-site calculation of the offset is accomplished by computing the mode and means of the newest model year vehicles and assuming these vehicles emit negligible levels of HC, using the lowest of either of these values to adjust the offset. Because we assume these vehicles emit only small amounts of HC, such an adjustment will only err slightly toward clean because the true offset will be

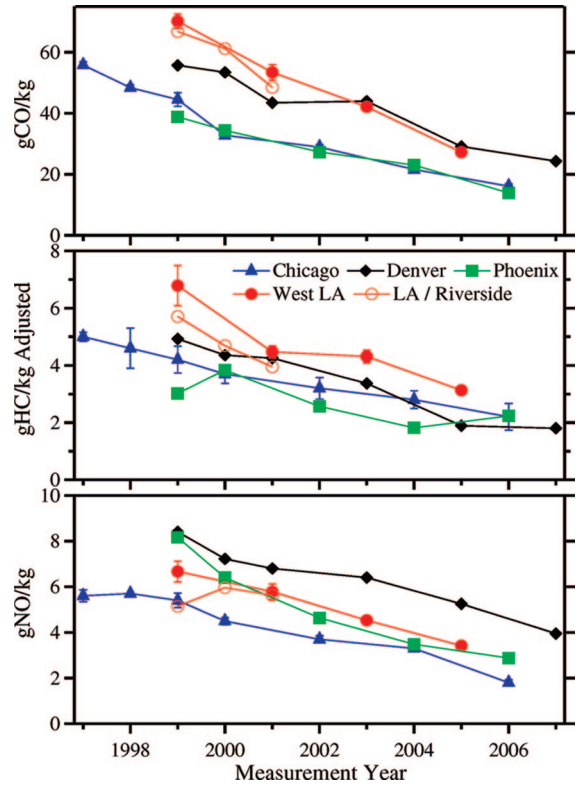

FIGURE 1. Mean, fuel-specific emissions plotted by measurement year. Standard errors of the mean calculated from each site's daily measurement means are plotted for the Chicago and West LA sites and are representative of the uncertainties for all sites.

a value somewhat less than the average of the cleanest model year and make. The offset is only used for comparing sites and is not included in any of the archived databases.

The standardization of measurement sites and sampling times was an attempt to lower year-to-year-variability in the measurements by minimizing changes in fleet characteristics. But because the data in Figure 1 are not age normalized, year-to-year changes in the level of new car purchases changes the age of the vehicle fleets measured, and their mean emission levels vary with these changes despite controlling for location and sampling time. In addition, the weather is another variable that cannot be controlled from year to year and will slightly affect emissions, for example, by changing the amount of vehicle air conditioning that is being used.

Some of the differences in emissions shown in Figure 1 between the four sites can be accounted for by differences in fleet age and driving mode. Chicago and Phoenix have the

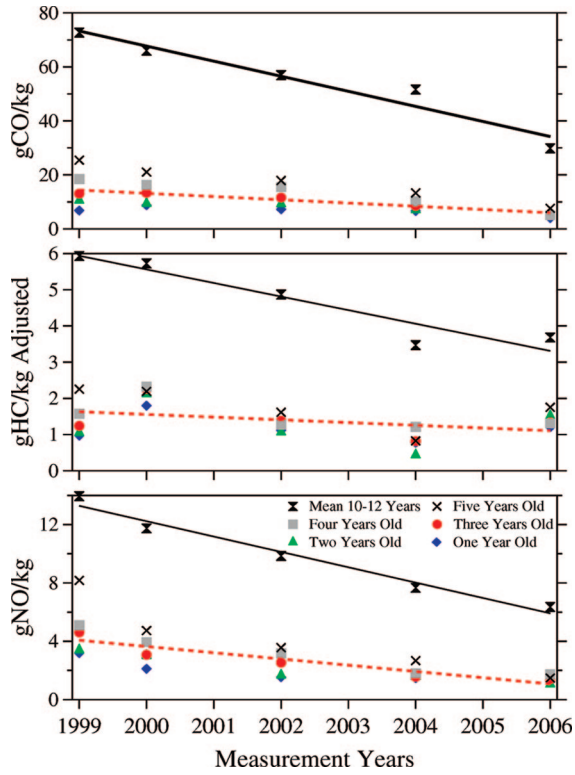

FIGURE 2. Mean, fuel-specific emissions for selected vehicle age groups from the Phoenix measurement site plotted by measurement year. The lines are least-squares fit to all of the data that makes up the subsets.

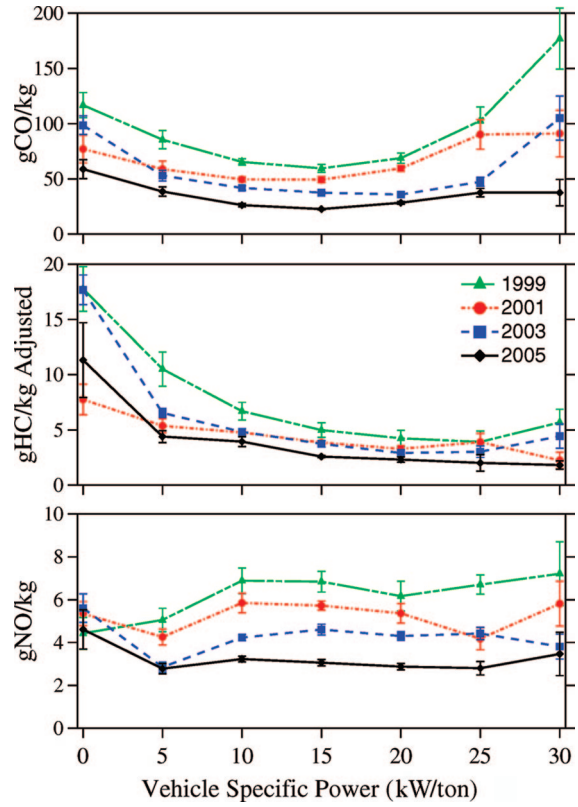

FIGURE 3. Mean, fuel-specific emissions for the four years of data collected at the West LA site plotted as a function of Vehicle Specific Power (VSP). Only emissions data with valid speed and acceleration measurements are plotted, and the uncertainties displayed are standard errors of the means calculated using the daily means.

newest fleets with average fleet ages of around 5 years, whereas Denver and West LA (despite the large geographical difference within the LA basin between the West LA and Riverside sites, the fleet ages and emissions are comparable) have older fleets of 7 and 6 years old, respectively. These age differences are reflected the strongest in the differences in CO emissions. The Denver site has the steepest grade (4.6°) and, therefore, the highest driving loads, which is one factor that contributes to the larger observed NO emissions. Other fleet differences that do not markedly impact the emissions picture are that the Chicago fleet has the lowest percentage of light trucks and diesel vehicles. All of the western sites have a much higher percentage of light trucks, and Phoenix (5.4% diesels) and Denver (3.3% diesels) have the largest percentages of diesel vehicles.

Each site plotted in Figure 1 has experienced large, fuel-specific, fleet-averaged emission reductions for CO, HC, and NO emissions. Using the beginning and ending year means, CO emission decreases range between 56% (Denver) and 71% (Chicago), HC emission reductions range between 27% (Phoenix), and 63% (Denver) and NO emissions have dropped between 48% (West LA) and 68% (Chicago). During the decade measurement span, the fleet in Chicago has become newer by 0.7 years and the fleets in Denver, West LA, and Phoenix have become older by approximately 0.4 years. Reductions of similar magnitudes have been noted by other authors with other data sets and for other species, reflecting what we believe to be a nationwide trend (*3, 12–14*).

There are at least three common factors that can be supported with data from any of the four sites contributing to the large measured reductions in fuel-specific emissions. The first is that the reductions are observed across all vehicle age groups. Figure 2 uses data from Phoenix and plots emissions by vehicle age for the five data sets collected in Phoenix. The 10−12 year old vehicles are averaged together to have a sufficient number of vehicles. The lines are least-squares fits to the 10−12 year old vehicles and the 1−5 year old fleets. By keeping vehicle age constant, the vehicle model years are incrementally increasing during the seven year time span. For example, the 5 year old vehicles (1994 models) measured in 1999 are part of the 10−12 year old average in 2006.

In absolute terms, the 10−12 year old vehicles show the largest improvements, but the 1−5 year old vehicles show a similar reduction on a percentage basis. One way to observe this is to notice the spread between one year old vehicles in 1999 (1998 models) and five year old vehicles (1994 models) and then compare that spread with one year old vehicles measured in 2006 (2005 models) and five year old vehicles (2001 models). This spread has contracted significantly for all three pollutants during the seven years (*15*). It is important to point out that only by this process of annual or biennial sampling can the difference between different model year fleets and the effect of age on a given model year be separated.

The second common factor is the gradual reduction of the influence of engine load on fuel-specific emissions. Using the road grade, speed and acceleration measurements that we collect, and an equation proposed by Jimenez, we can calculate the instantaneous vehicle-specific power (VSP) of an on-road vehicle in kilowatts per metric ton (*16*). The West LA site is a traffic light controlled on-ramp, and Figure 3 plots emissions data that has valid speed and acceleration measurements against VSP for the four data sets collected. In 1999, the CO emissions versus VSP pattern was nearly bowl shaped with emissions roughly tripling between its minimum VSP at 15 (approximately 18 mph and a 1.7 mph/s acceleration rate) and 30 kW/ton (approximately 22 mph and a 4.4 mph/s acceleration rate). The emission difference between these two load points in 2005 is statistically

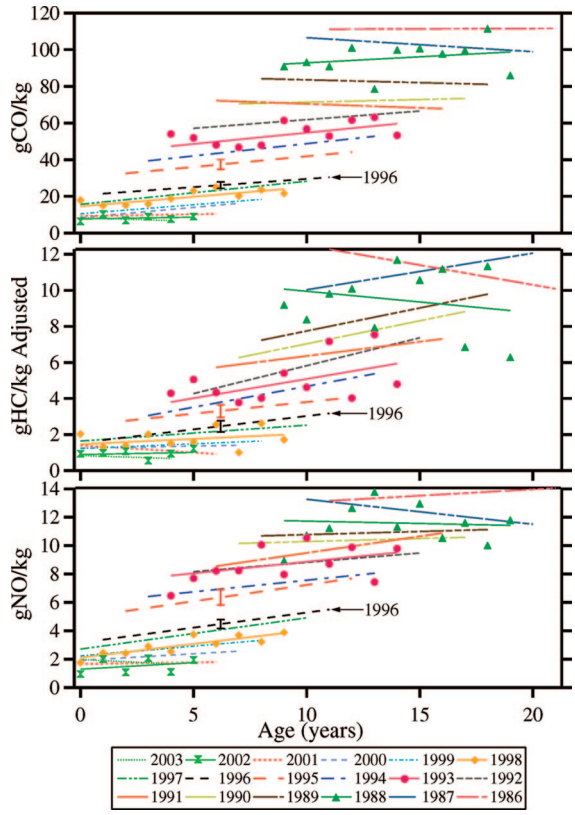

FIGURE 4. Linear least-squares fits to mean, fuel-specific vehicle emissions as a function of age from the combined data of Chicago, Denver, West LA, and Phoenix. The data are grouped by model year beginning with 2003, which is the first model with at least three measurements, and go back to the 1986 model year. The data used to construct the linear fits are displayed for model years 2002, 1998, 1993, and 1988. The error bars plotted for model years 1995 and 1996 are 95% confidence limits for the midpoints.

indistinguishable, meaning that far fewer vehicles are being forced into power enrichment during accelerations onto freeways. With lower CO emissions it would be reasonable to expect increases over time in NO emissions between these same load points. What we find is just the opposite, with NO emissions also declining and the emissions versus load curve flattening, as with CO, despite more vehicles operating near stoichiometric conditions.

Finally, the third factor observed is that on-road fleet-averaged emission deterioration is near zero for model years newer than 2001 and older than 1990. We have combined the emissions data from all four cities (to increase the representation of older model year vehicles) into a plot of fuel-specific emissions versus age that can be used to quantify the observed on-road emission deterioration rates. Vehicle age has been defined as the difference between the year of measurement and the vehicle model year. Assuming that vehicle emissions deterioration is a linear process, we can fit a straight line to each model year grouping, where the slope of that line is the grams of emissions per kg of fuel per year deterioration rate. Figure 4 is a plot of the linear emission trends for model years 2003 (the first year with at least three measurements) through 1986. The actual data points used to construct the linear fits are plotted for model years 2002, 1998, 1993, and 1988 to illustrate the noise in the data.

Two sources of noise important to mention are the sample size used to calculate the means and the instrument noise. Vehicle emissions are γ-distributed, and means sampled from a γ distribution are highly sensitive to the number of samples (*17*). In Figure 4, mean samples per measurement year are 2000 samples for model year 2002, 1800 for 1998, 950 for 1993, and 450 for 1988, and the plotted data points show how the noise increases proportionately. The HC channel has the poorest signal-to-noise ratio of all the species measured due to a combination of the low levels emitted by the vehicles and a smaller absorbance cross section. This contributes to about a threefold increase in the relative noise levels in the HC data for similar sample sizes when compared to the CO or NO channels (*7, 10*). We have also introduced some additional data variability to the NO data sets by combining data collected during alternating years from sites with differing vehicle load profiles.

Three notables in Figure 4 are the generally stair-step nature of emission differences between model years and an emissions gap between 1996 and 1995 models (significant at the 95% confidence level) that also serves as a demarcation point where the stair steps change step sizes (smaller before 1996 and larger after). Any possible explanation for the emissions gap has to account for the fact that its presence is universal, and we have shown upon analysis that it can be seen in all makes, models, and types of vehicles. Three of the more obvious possibilities are extended emission component warranties, the introduction of the second generation of on-board diagnostic systems (OBDII) and a reduction in new vehicle emission standards. The extended warranties began with 1995 models, and although OBDII was introduced with the 1996 models, it was not accompanied by any new legislated reductions in per-mile emissions and was generally viewed as being aimed at reducing long-term vehicle emissions deterioration, not reducing their initial emissions. However, as part of the changes that accompanied OBDII systems, one of the more notable was the requirement to monitor catalyst efficiency, which dictated the addition of two additional oxygen sensors, one before and one after the catalyst. These sensors have allowed manufacturers to correct for any long-term drift that might occur with the manifold oxygen sensor and, most importantly, to control the fuel trim on a cylinder-by-cylinder basis and produce lower emission levels of the size we have observed (*18, 19*).

Figure 5 compares model year emission deterioration rates (the slopes of the lines plotted in Figure 4) with deterioration rates predicted by the U.S. Environmental Protection Agencies vehicle emissions model MOBILE6.2 (*20*). The uncertainty bars plotted with the on-road data are the standard error of the slope of the least-squares fit. The model output has been created using an input file (see Supporting Information) that models a winter and summer on-road scenario with no refueling emissions where the vehicles are subject to the Denver, CO I/M program. The deterioration rates plotted are for a vehicle fleet composed of 50% light-duty gasoline vehicles and 25% each of light-duty gasoline trucks type 1 and 2. The modeled data have been converted to gram/kg data assuming a fuel density of 726 g/liter, and the modeled NO_2 data have been converted to NO for direct comparison with the on-road measurements.

Vehicles newer than model year 2001 and older than model year 1990 show little or no emissions deterioration, and only for the HC data are the magnitudes of these rates similar between the measurements and the model. The model predictions are much too high for 1996 and newer model year CO and NO emissions. The modeled CO emissions deterioration rates begin their large decrease with model year 1998 due to the removal of sulfur from gasoline that began in 2000 (*21*). Although there is a noticeable on-road emissions break between model years 1995 and 1996, there is not a significant difference between the measured on-road emissions deterioration rates accompanying OBDII systems. These systems were designed to reduce emission deterioration rates by finding emission system failures sooner

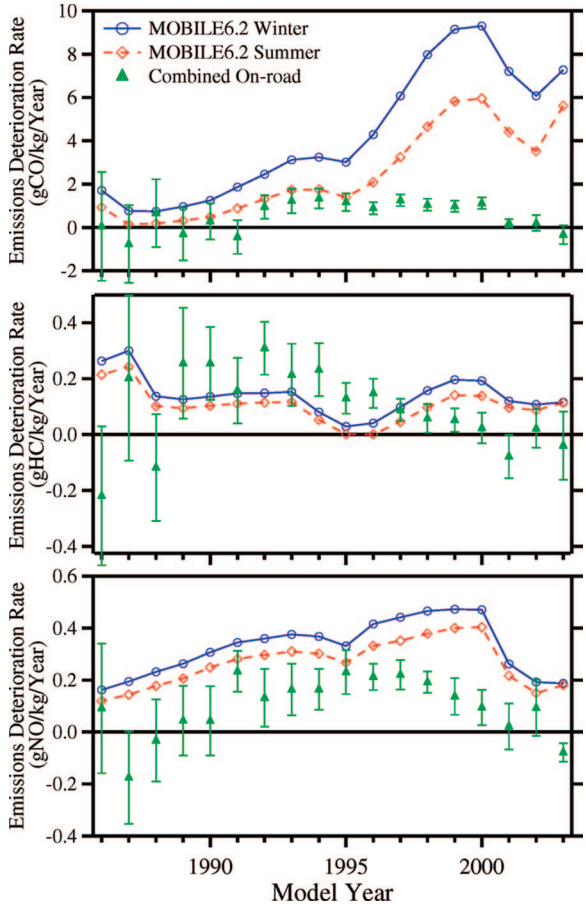

FIGURE 5. On-road emissions deterioration rates (see Figure 4) vs model year compared with emissions deterioration rates determined from the U.S. EPA's MOBILE6.2 vehicle emissions computer model. The uncertainty bars plotted with the on-road data are the standard error of the slope for the least-squares fit. The MOBILE6.2 g/gallon data have been converted to g/kg assuming a fuel density of 726 g/liter, and the MOBILE6.2 NO_2 emissions have been converted to NO equivalents for comparison with the on-road data.

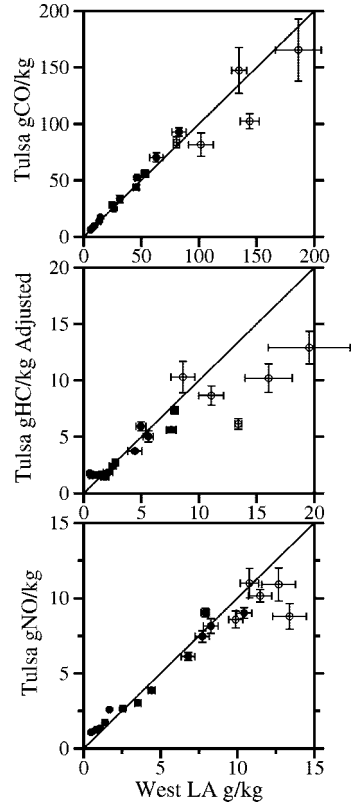

FIGURE 6. Fuel-specific emissions comparison between the 2003 data sets collected in Tulsa, OK and West LA, CA for model years 2003–1986. The uncertainty bars plotted are standard errors of the mean calculated from the daily means at each site. The 1:1 line is plotted as a reference. The 1990–1986 model years (open circles) with the most noticeable uncertainty bars constitute less than 12% of the West LA fleet and less than 6% of the Tulsa fleet.

and alerting the driver to the need for repair. The similar deterioration rates suggest that emission system failure and repair is not the dominant factor (vehicle retirement rates and initial fleet emissions are others) determining on-road deterioration rates.

We have argued in the past that the majority of the on-road emissions reductions that have been observed are likely the result of continued improvements in function (i.e., three oxygen sensors) and durability of vehicle emission control systems and not state I/M programs or fuel reformulations (*15*). To attempt and differentiate between emissions reductions from vehicle technology and state I/M programs and fuel mandates, we collected data in Omaha, NE and Tulsa, OK. Neither of these cities has ever been involved in any type of state I/M program nor have they required additional fuel reformulation. The uphill interchange ramp in Tulsa is the most comparable location to the other sites with a similar layout and driving mode.

We have previously shown that I/M programs export failing vehicles to outlying areas, thus producing artificial emission increases when those areas are compared to a neighboring I/M fleet (*22, 23*). Tulsa is geographically isolated in that it is buffered along all of the major commuting routes with at least one major metropolitan area without I/M between it and a city with an I/M program. For example, Oklahoma City is between Tulsa and Dallas to its south. These buffer cities, along with the increased distances from major I/M cities, should work to limit the number of emission failing vehicles being imported to the Tulsa area, allowing the collection of emission data from a true non-I/M control fleet.

Figure 6 is a series of comparison plots between data collected in Tulsa, OK in September 2003 and in West LA in October 2003 for the three measured pollutants. Because there is only one month between the two collection periods, changes in fleet age are minimized. The mean emissions for vehicle model years 2003 to 1986 are plotted, and the uncertainty bars are standard errors of the mean calculated using each sites' daily means. The line is a 1:1 line drawn for a visual reference. The agreement between the two data sets is apparent, not withstanding the increased noise in the older model years due to the shrinking sample size. This agreement suggests that the large emission reductions that we have observed at the West LA site have also occurred in Tulsa. It also suggests that any site-specific fuel effect on these three emission species are minor at best, although fuel reformulations have been clearly shown to reduce other species, such as benzene, that are not measured in this study (*14*).

However, we do not wish to overemphasize the absolute agreement because socioeconomics plays a role in absolute vehicle emission levels even when age is controlled for, as in this case (*24*). It is more important to compare the two site's emission versus age distributions and to take advantage of the fact that the first four model years (the first four model years are not required to be inspected in CA) at each site have not been subject to an I/M inspection. Conventional wisdom and the MOBILE computer model predicts that an I/M program will change the shape of this distribution (i.e.,

Figure 6 should not be a straight line) after the first four model years to the extent that the program works to retard the normal emissions deterioration and that these benefits should accumulate over time (*25, 26*). The fact that vehicle emissions versus age in Tulsa, OK have a similar distribution (the linear relationship continues after the first four model years) when compared to vehicles in Los Angeles suggests that vehicle emission inspections in California should not be considered a major factor in the large on-road emission reductions observed at the West LA site (*27*).

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Supporting Information Available

Listing of the MOBILE6.2 input file used to create the data used in generating Figure 5. This material is available free of charge via the Internet at http://pubs.acs.org.

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