Airborne Emissions from 1961 to 2004 of Benzo[*a*]pyrene from U.S. Vehicles per km of Travel Based on Tunnel Studies

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We identified 13 historical measurements of polycyclic aromatic hydrocarbons (PAHs) in U.S. vehicular traffic tunnels that were either directly presented as tailpipe emission factors in μg per vehicle-kilometer or convertible to such a form. Tunnel measurements capture fleet cruise emissions. Emission factors for benzo[*a*]pyrene (BaP) for a tunnel fleet operating under cruise conditions were highest prior to the 1980s and fell from more than 30-μg per vehicle-km to approximately 2-μg/km in the 1990s, an approximately 15-fold decline. Total annual U.S. (cruise) emissions of BaP dropped by a lesser factor, because total annual km driven increased by a factor of 2.7 during the period. Other PAH compounds measured in tunnels over the 40-year period (e.g., benzo[ghi]perylene, coronene) showed comparable reduction factors in emissions. PAH declines were comparable to those measured in tunnels for carbon monoxide, volatile organic compounds, and particulate organic carbon. The historical PAH "source terms" determined from the data are relevant to quantifying the benefits of emissions control technology and can be used in epidemiological studies evaluating the health effects of exposure, such as those undertaken with breast cancer in New York State.

Introduction

Polycyclic aromatic hydrocarbons (PAH), principally benzo[*a*]pyrene (BaP), have been associated with lung, bladder, and, possibly, breast cancer, as discussed by Boström et al. as part of an extensive review of individual PAH risks and sources (*1*). Traffic emissions are a major source of both indoor and outdoor exposures to PAH, and often the largest source in areas near cities, as has been confirmed in a number of experimental studies. (See Supporting Information for references.) We present here estimates of U.S. historical tailpipe emissions into the air of BaP per vehicle-km traveled throughout the period 1961−2004. We focused on BaP, because it is considered a good marker of overall PAH airborne exposure (*2*) and was measured in every PAH tunnel study located. Emissions for benzo[*ghi*]perylene (BGP), coronene (COR), benzo[*a*]anthracene (BAA), perylene (PER), pyrene (PYR), and fluoranthene (FLT) are presented in the Supporting Information for those studies that measured them by 1975. The resulting historical "source terms" are relevant to quantifying the benefits of emissions control technology. When the tailpipe emissions data are combined with estimates of yearly traffic flow and a meteorologic dispersion model (*3–6*), it is possible to estimate individualized ambient PAH exposures for use in retrospective epidemiologic studies (*6*), as is being done in studies of breast cancer (*7, 8*).

Tailpipe Emissions per km by Year Determined from Tunnel Measurements. Measurements in vehicular traffic tunnels provide a historical snapshot of the average performance of thousands of vehicles, including heavy-duty trucks (HDV), both gasoline (HDGV) and diesel (HDDV). We located 13 measurements of BaP-emissions or concentrations in eight different U.S. tunnels between 1961 and 2004. One study was unpublished. See Table 1.

Tunnel air was sampled in the different studies for $^1/_2$-h or longer during time periods lasting up to 2 weeks. Over the years, the filters that were used to collect BaP in the tunnels changed from glass and quartz fibers to Teflon coated fibers. Most authors found little difference in the collection efficiency of these filters (*14*), provided extractions were made relatively quickly (*24*) or stored in the dark (*25*). As a result of the high BaP recovery rates expected for all of the extraction methods used, we conclude that no historical bias was introduced into BaP emission factors by the shift in extraction technology over time (see the Supporting Information).

Potential competing sources of PAH in tunnels other than tailpipe emissions are thought to be small, particularly emissions from brake linings and tires (*26, 27*).

Of the 13 studies in Table 1, three of them gave measurements of BaP per vehicle-km directly (*9, 21, 28*) based on measurements of tunnel output concentrations and measured tunnel ventilation rates. For the other studies, measurements of copollutants in the tunnel with known tailpipe emission rates of their own obviated the need for tunnel ventilation measurements. To be useful, the excess concentration in the tunnel of these copollutants over outdoor background concentrations must be traceable to vehicle emissions and not some other source. As discussed below, under these conditions, when the copollutant's emission factor is known, the measured ratio of BaP to copollutant concentration can be used to extract the BaP emission factor. Following De Fré (*29*), we call these copollutants, "tracers". Five of the tracer studies used carbon in the form of CO_2 and CO as the tracer to measure BaP emitted per unit of fuel consumed (*17, 19, 20, 22, 30*). We converted results in units of fuel consumed to BaP per vehicle-km by dividing by national fuel economy rates for the relevant year as determined from Davis and Diegel (*31*), taking into account the percentages of truck traffic in each tunnel. The BaP emission factor was not explicitly given in one of the 5 studies (*22*), but its value could be extracted from data given in the paper and other publications by the authors (see the Supporting Information). We call the 8 studies discussed so far, "complete". The remaining five ("opportunistic") studies were not designed to estimate emission factors, but the

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authors did report BaP-concentration data at various locations in their respective tunnels. We have converted this opportunistic, concentration data to emissions per km using tracer techniques, relying either on BaP itself, as measured by other researchers in the same tunnel in a different year, or relying on the copollutant lead, which was prominent in the early years. Tracer methods require knowledge not only of the concentration of the tracer but also its vehicle emission factor in one of the years of measurement.

Tracer methods, in effect, provide a way to infer tunnel ventilation parameters. For instance, when using BaP as a tracer, we, in effect, inferred effective ventilation parameters of a particular tunnel by computing the ratio of concentration to emission factors determined in "complete" studies in year, t = "1". Correction for ventilation from the movement of vehicles, the so-called, "piston effect," was made using a function of the traffic rate. This approach allowed us to convert to emission factors the concentration measurements given in the "opportunistic" studies carried out in the same tunnel in year, t = "2", using the following equation

$$Ef(2) = Ef(1)\frac{C(2)}{C(1)}\left(\frac{N(1)}{N(2)}\right)^{w} \quad (1)$$

where *Ef(t)* is an emission factor, *N(t)* is a traffic rate (vehicles/min), and *C(t)* is a BaP tunnel concentration.

The exponent, w, lies between 0.5 and 1 (as discussed in the Supporting Information). A numerical example computed by Schlaug and Carlin (Figure 5.4 of ref *32*) for a "composite" transversely ventilated tunnel produced an exponent of about two-thirds, which we take as the most likely value for, w. To account for the uncertainty in this exponent, we use a triangular distribution between 0.5 and 1, with the peak value set to 0.66.

The validity of eq 1 depends on the assumption that the concentration at both the tunnel entrance and the intake air vent can be neglected compared to the concentration in the tunnel at the exit portal. Generally, in the tunnel studies discussed in this paper, background concentrations at the intake air vent were 10% or so of the measured concentration, which means that neglecting background concentrations is a reasonable approximation. By relying on eq 1, it is also assumed that the fan capacity and operating protocols have not changed over time. This, for instance, was the case with the Caldecott tunnel in the East Bay area of California (Mailhot, R., California Department of Transportation, personal communication, 2007).

In the "complete" studies in the California Caldecott Tunnel, measurements were made 50 m from the traffic exit portal (*33*). There is some evidence that penetration of outside air reduces tunnel concentration at this distance, which could lead to an underestimate in our BaP-tracer method by as much as a factor of 2 for the emission factors inferred in the Caldecott tunnel for 1983 and 1989 (*34*). Comparison with emission factors obtained by the lead-tracer method allows us to test the reasonableness of ignoring the limitations of the inferred ventilation method using BaP as a tracer.

Another complication with the Caldecott studies is that they give us BaP concentrations in the tunnel proper, whereas the opportunistic studies measured the BaP concentration in the exhaust rooms above the tunnel. As discussed in the supporting material, the ratio between exhaust and tunnel concentrations, $C_{exhaust}/C_{tunnel}$, lies between 0.5 (when a tunnel is vented by piston effect only) and 1 (when a tunnel is vented by transverse ventilation only), on the assumption that there is no significant deposition or resuspension in the ventilation ducts. We have picked 0.75 as a midrange value for the ratio, assigning an uncertainty of ±0.25, which spans the full range allowed.

Deposition of fine particulates has been studied in tunnels (*27, 35, 36*), with the conclusion that it can be neglected, with the exception of particulates of sulfur dioxide (SO_2) and sulfate (SO_4). The assumption of negligible deposition in tunnels for fine particles is universally made in the kinds of studies analyzed here (*29*), as is the assumption of negligible resuspension of tunnel dust by moving vehicles. We assume the same low deposition/resuspension conditions hold for the parallel ventilation ducts, which, like the tunnel proper, are quite large.

The most complicated derivation we had to make of inferred ventilation parameters was in the Sumner Tunnel in Boston. The 1961 (complete) measurements, which included direct measurement of the ventilation rate, took place with no additional ventilation from the piston effect, because traffic moved in two directions. This was not the case in 1963, when the traffic was changed to one-way, producing a piston effect. To mathematically correct for the 1963 piston effect so that the 1963-BaP data could be compared to the 1961 data, we relied on measurements of benzene-soluble particulates made in the 1963 study as a function of distance into the tunnel. As shown in the Supporting Information, fitting this data to the steady-state tunnel equation for transversely ventilated tunnels (*37*) allowed us determine that the concentrations would have been a factor of 2.3 higher in the absence of the piston effect, i.e. had the tunnel been two-way in 1963.

Although not previously used to estimate BaP-emission factors, lead in gasoline has been used as a tracer in tunnels to measure emission factors for total suspended particulates, SO_2, SO_4, carbon, sulfur, and barium (*35, 38*). The methodology is suitable for any copollutant, as described in ref *35*. In some cases, the Pb-tracer method allowed us to obtain a second, independent estimate of BaP per vehicle-km for the same year in the same tunnel, increasing confidence in the combined results, which we obtained by computing the geometric mean.

For lead, the tracer equation (eq 1) simplifies, because year 2, is the same as year 1, so the factors involving traffic rate cancel, leaving

$$Ef_{BaP} = (Ef_{Pb})\frac{C_{Bap}}{C_{Pb}} \quad (2)$$

where *Ef* is an emission factor and *C* is a tunnel concentration. To use the Pb-tracer, ratio method, it is necessary to have an estimate for the Pb-emission factor, namely the amount of small particles of lead emitted per km by the fleet of vehicles using the tunnel. We obtained these estimates for the tunnel study years by substituting information obtained from the literature in the following equation

$$Ef_{pb}(t) = \frac{S(t)\,C_{pb}(t)\,F}{m(t)} \quad (3)$$

where $Ef_{Pb}(t)$ is the Pb-emission factor in g/km for the year, t, of the study; $S(t)$ is the fraction of the fleet consuming leaded gasoline at the time; C_{pb}(t) is the number of grams of lead per liter of gasoline in the year of the study; F is the fraction of lead in a liter of gasoline that is emitted as particles with significant residence time in tunnels; and $m(t)$ is the fuel economy in km/L for the year of the study.

We obtained values for, $S(t)$, $C_{pb}(t)$, and $m(t)$ from the literature as referenced in the Supporting Information. We compared measurements in U.S. tunnels of the fraction, F, of lead in gasoline emitted as small particles and found them to be consistent over a 20+ year period (*9, 36, 39*), with an average value of 0.23 ± 0.04. This number is quite close to the average of 0.25 ± 0.04 obtained in 3 tunnels in Belgium during the period 1989−1991 by De Fré and colleagues (*29*).

After 1995, researchers reported emission factors separately for light-duty vehicles (LDV) and heavy-duty diesels (HDDV), having separated out the contribution of diesel

TABLE 1. Summary of PAH Studies in U.S. Tunnels, along with Methods Used To Estimate BaP Emissions per Vehicle-km

study	date(s)	sampler	particle filter fibers	extraction method	author method[a] (see text)	our method(s)[a]
Larsen, Boston (*9*)	Sept, 1961, 24 h	Hi-Vol, inlet/outlet of vent. bldg	glass	benzene in Soxhlet apparatus	direct	Pb-tracer
Conlee, Boston (*10*)	April, 1963, 24 h	Hi-Vol (vent and tunnel)	not specified	benzene in Soxhlet	none	Pb-tracer
Fox, Baltimore (*11*)	May, 1975, day	Hi-Vol, location not indicated	glass	benzene in Soxhlet	none	BaP-tracer
Kebbekus, NYC/NJ (*12*)	Nov, 1981, day	Hi-Vol sampler in moving truck	glass	cyclohexane in Soxhlet	none	Pb-tracer
Hering, Berkeley (*13*)	Feb, July, 1983, day	Hi-Vol in air duct, HLPI[b] set at PM1.3	quartz	see footnote *c*	none	(1) BaP-tracer, (2) Pb-tracer
Benner, Baltimore (*14*)	Sept–Nov, 1985, Dec, 1986, 24 h	Hi-Vol in exhaust rooms	Teflon	DCM[b] in Soxhlet for >18 h	direct	Pb-tracer
Venkataraman, Berkeley (*15, 16*)	Aug, 1989, day	HLPI[b] in exhaust duct of bore 3, cyclone set at PM4	quartz	DCM,[b] ultrasonication	none	BaP-tracer
Fraser, Los Angeles (*17*)	Sept, 1993, day	Hi-Vol DVI[b] in tunnel, PM2.5 cutoff	quartz (*18*)	hexane, benzene, propanol with sonication (*18*)	carbon tracer	convert from ng/gal to ng/km
Miguel, Berkeley (*19*)	Aug, 1996, day	AIHL[b] cyclone set at PM1.3. Inside tunnel and entrance of vent air in bores 1 and 2.	Teflon-coated glass	DCM[b] with ultrasonication	carbon tracer	convert from ng/gal to ng/km
Marr, Berkeley (*20*)	summer, 1997, day	AIHL[b] cyclone set at PM2.5. Inside tunnel and entrance of vent air in bores 1 and 2	Teflon-coated glass	DCM[b] with ultrasonication	carbon tracer	convert from ng/gal to ng/km
Gertler, Pennsylvania Turnpike (*21*)	May, 1999, day, night	cyclone set at PM2.5, inside tunnel	Teflon-impregnated glass	DCM[b] + sonification, methanol + sonification	direct	
Chellam, Houston (*22*)	Aug, 29-Sept 1, 2000, day	Hi-Vol inside tunnel, with impactor set at PM2.5	quartz	hexane, benzene, isopropyl alcohol mixture, mild ultrasonic agitation	carbon tracer	see footnote *d*. Convert from ng/L to ng/km
Phuleria, Berkeley (*23*)	Aug, 2004, day	Hi-Vol, inside tunnel in bores 1 and 2, impactor at PM2.5	quartz	DCM,[b] methanol, sonication	carbon tracer	sum over sizes, convert from ng/gal to ng/km

[a] See text. The direct method involves measurement of ventilation rates, rather than inference from tracer measurements. The BaP-tracer here is BaP found in the same tunnel in a different year in a different study. [b] HLPI = Hering Low Pressue Impactor. DCM = dichloromethane (methylene chloride). DVI = dichotomous virtual impactor. AIHL = Air and Industrial Hygiene Laboratory. [c] Extraction of February samples: cyclohexane using ultrasonic cleaner; July data: sequential extraction with cyclohexane, dichoromethane, and acetone in Soxhlet apparatus. [d] The BaP emission factor was not directly reported in the paper but can be derived from data in the paper (see the Supporting Information). The same value is given in a study summary table in ref *23*.

emissions by regressing against the percentage of HDDV in the fleet (*19–21, 30*). Averaging over the 4 HDDV emission factors measured from 1996 to 2004 gives a value for BaP for heavy-duty diesels of 16.8 μg/km, as discussed in the Supporting Information. The average of LDV emissions of BaP per μg/km over the comparable period was ten times lower. To obtain fleet emission factors for the post-1995 studies, we have combined the separately measured light-duty and heavy-duty emission factors, using a value of 4% trucks to match the most common percentage in earlier studies. See the Supporting Information for details of this standardization.

As discussed in the Supporting Information, error rates were propagated throughout the calculations using Monte Carlo techniques.

Results and Discussion

Table 2 shows the estimated BaP-emission factors. The last column gives the geometric mean of estimates derived using different methods in the same tunnel. The rates fell from more than 30-μg per vehicle-km to approximately 2-μg /km averaged over the 1990s. The geometric means are plotted in Figure 1.

The emission factors are reasonably consistent with tunnel data collected in Europe, if comparisons are made to emissions from fleets, not in the same calendar year, but in years when the percentage of catalytic converters are the same (see the Supporting Information). Results for those PAHs other than BaP that were measured in tunnels as early as 1975 are shown in Table S1, Figure S1, and Figure S2. The reductions over time were comparable to those found for BAP.

As discussed in the Supporting Information, declines for traffic pollutants other than PAH measured in tunnels are consistent with what we find for PAH. For instance, 14 individual VOC concentrations measured in the Lincoln Tunnel (*43*), and converted by us to emission factors, declined by a factor of 5 from 1970 to 1982 and by at least another factor of 5 by 1995, when they were measured in the same tunnel by Gertler et al. (*41*). Declines in emissions per km of particulate organic carbo. were also large, although uncertainties are significant. Carbon monoxide emission factors measured in tunnels also dropped markedly, as shown in Table S3 and Figure S3. Note that pollutant emissions during noncruise parts of the driving cycle may not have declined as much as cruise emissions.

In addition to emission factors, Table 2 also lists the tunnel grades, vehicle speeds, fleet percentages (4% from 1996 onward to match standardized emission factors), and average daily ambient temperature taken from the nearest weather station's archived data (see the Supporting Information). Average vehicle age in the fleet was not available for the pre-1990 tunnel studies, so is not included in the table.

Multivariate regression of the BaP emission factor against tunnel speed, ambient temperature, and study-year found only study-year, to be statistically significant ($p = 0.0005$), with ambient temperature having a weak association ($p = 0.11$), and speed not significant in multivariate analysis ($p = 0.56$), in contrast to univariate analysis ($p = 0.035$).

Tunnel grade cannot explain the large drop in emission factors, either: We have 5 measurements in East-coast, urban-area tunnels out to 1986, with comparable tunnel grades, and dramatic drops in emission factor.

As indicated in Table 1, before 1983, there was no particle size cutoff applied to the collected particulates other than that inherent in a high-volume sampler. After 1986, all of the measurements had at least a PM2.5 cutoff. However, the large drop in emission factors post-1975 cannot be due to a difference in collection efficiency, because the low 1986

TABLE 2. BaP Fleet Emission Factors by Method, along with Tunnel Characteristics

	tunnel characteristics				measurement method (units in μg/km)[a]				
tunnel/yr	% HDV[c]	slope (deg)	speed (km/h)	temp (°C)[d]	using vent rate	BaP-tracer[e,f]	Pb-tracer[e]	C-tracer[g]	combined emission factor[b]
Sumner, 61	3.6	±3.5	48	17	31 (10)		33 (18)		31 (12)
Sumner, 63	3.6	±3.5	48	10			46 (20)		36 (11)
Baltimore Harbor, 75	12	±3.5 (*34*)	46 (*40*)	26		31 (19)	63 (50)		35 (14)
Lincoln/ Holland, 81	12	−3.8 to + 3.5 (*41*)	43 (*41*)	3		23 (14)	10 (5)		10 (5)
Caldecott, 83	4.5[h]	+4.2	60	18[i]		11 (3.5)	4.6 (1.6)		7.1 (1.8)
Baltimore Harbor, 86	11	±3.5 (*34*)	46[i] (*40*)	4	2.0 (1)		4.6 (3.6)		3.0 (1.7)
Caldecott, 89	6[k]	−4.2	60	17		0.95 (0.45)			0.95 (0.45)
Van Nuys, 93	3.9	−1.7 to +1.0	64	19				2.9 (0.8)	2.9 (0.8)
Caldecott, 96	4[l]	+4.2	66−78	17				0.57 (0.15)	0.57 (0.15)
Caldecott, 97	4[l]	+4.2	59−89 (*42*)	19				1.9 (1.2)	1.9 (1.2)
Tuscarora, 99	4[l] (43)[m]	+0.3	88	20	6.0 (3.4)				6.0 (3.4)
Washburn, 00	4[l]	±6.0	56−72	33				0.46 (0.13)	0.46 (0.13)
Caldecott, 04	4[l]	+4.2	59−89[n]	20				1.2 (0.58)	1.2 (0.58)

[a] Mean values. Standard deviations of the (approximately) log-normal distributions in parentheses. [b] Monte Carlo average of geometric mean of results from different methods. [c] Trucks with 6 wheels or at least 3 axles. See the Supporting Information for sources. [d] Average daily temperature at nearest location with archived data. [e] Our calculations, except for the Pb estimate for 1961. [f] The BaP-tracer is BaP found in the same tunnel in a different year. [g] Authors' calculations converted to emission per km using National fuel economy figures (see the Supporting Information). [h] Average of Bores 1 and 2 (with and without HD traffic). [i] Summer temperature to match other Caldecott temperatures. The 1983 emission factor does not change with and without February data included. [j] Traffic flow rates in 1986 and 1975 in the BHT were similar. [k] Bore 3 only. [l] Standardized value. See text. See Table S8 for the actual study value. [m] Actual % of trucks in study. [n] Taken to be the same as the 1997 values.

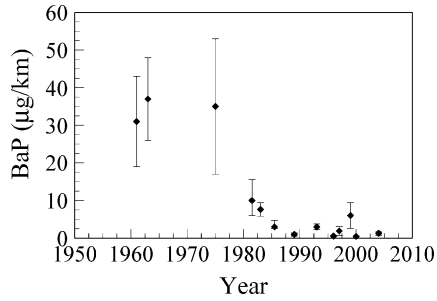

FIGURE 1. Emission of benzo(a)pyrene from U.S. vehicles per km of travel, based on data collected in tunnels.

measurement (3.0 μg/km) was made without any size separation. (See further discussion in the Supporting Information.)

Fleet emissions dropped 91% from 35 μg/km in 1975 in the BHT to 3 μg/km in 1986 in the same tunnel. Truck percentages were similar (~12%), so the drop cannot be attributed to a change in percentage of trucks. Nor can the bulk of the change be attributed to improvements in truck emissions. Unlike the LDV fleet, the HDV fleet was not subject to effective emissions regulation until after 1981 (*44*). Data on CO and particulates indicate that the improvement in truck emissions was relatively modest compared to the decline in BAP shown in Table 2. For instance, measurements in tunnels of CO emission factors for HDV showed virtually no change from 1979 to 1992 (*45*). Estimates of heavy-duty diesel fine-particulate emission factors based on chassis dynamometer measurements showed only a 43% decline over the 1975−1986 period, not a 91% reduction. Taking that 43% decline as the maximum reduction in the BaP emission factor for all HD trucks from 1975 to 1986 allows us to work backward from the 1986 data to put an upper limit on the HDV truck emission factor in 1975: Assume that all of the 3 ug/km fleet emission factor in 1986 was due to trucks, which made up a fraction (11%) of the fleet. Then, the maximum per truck-km emission rate would be 27 μg in 1986 and 47 μg in 1975, which would mean a modest 5.6-μg/km contribution to the 1975 *fleet* average (12% trucks).

The historical data given in Table 2 can be used to estimate yearly emissions from a road meteorological dispersion model to estimate individual exposures for use in an epidemiological study (*7, 8*) but only for cruise conditions and possibly after adjusting the numbers in Table 2 for the tunnel grade and percentage of heavy duty traffic, for instance, as described in the Supporting Information. Additional exposures occur during cold start and at intersections, where acceleration and deceleration take place. Some historical data are available on how these parameters change emissions as indicated in the Supporting Information, and it is possible to use field data to quantify their effect further. For this purpose, soil PAH measurements can be used (*7*). Airborne measurements of CO can also be used for calibration (*8*), because there is a reasonable correlation between CO and PAH (*7*).

For the 1983 and 1989 opportunistic measurements in the Caldecott tunnel, we have three complete measurements in the same tunnel that can be used as a consistency check of the inferred-ventilation-rate methodology. Despite the fact that the tunnel concentration varied 4-fold between 1996 and 1997, the ratio of tunnel concentration to computed emission factor differed only by 20% from the average, which suggests that the assumptions underlying the method are reasonable.

There are 4 tunnel measurements in Table 2 where the Pb-tracer methodology can be compared with the inferred-ventilation methodology. The results differ by a factor of 1.5 $\pm$ 0.4. Considering the limitations of these two methods, this difference is reasonable and can serve as a marker of the uncertainty of the overall methodology.

The emission factor for 1999, measured in a Pennsylvania turnpike tunnel, is noticeably higher than values measured back to 1986. There have been reports of increased PAH in urban areas in this period based on measurements in dated sediment cores from reservoirs and lakes (*46*). However, in subsequent papers, the authors of this finding have attributed the increase to sources other than deposition of atmospheric PAH, especially runoff from parking lot sealcoat (*47, 48*). Thus, the high estimate in Figure 1 for 1999 remains unexplained and may simply represent the variance in the tunnel methodology and/or variance in the vehicle fleet (see the Supporting Information).

The 2.9 μg /km emission factor reported in 1993 for the Van Nuys Tunnel in Los Angeles may be higher than its neighboring data points in Figure 1 because of the acceleration/deceleration that takes place at traffic lights entering and leaving this (short) tunnel. These considerations have been called upon to explain other higher than average emission factors measured in this tunnel, namely for carbon monoxide (*41, 49*).

The quantitative information in Figure 1 and Table 2 can help to track the changes in BaP tailpipe emissions over time back to 1960 for use in health studies. The data indicate that there was more than a 15-fold decline in BaP and other PAH cruise emission factors for the U.S. vehicle fleet following introduction of automotive pollution controls. Total yearly (cruise) emissions dropped by a lesser factor, because overall vehicle miles traveled in the U.S. increased by about a factor of 2.7 between 1970 and 2005 (*31*). In summary, there were additional benefits from the introduction of automotive emission controls that are not usually counted in cost-benefit studies of air pollution regulations.

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Supporting Information Available

Information on pollutant trends other than BaP, details of uncertainty analysis, derivation of equations, and disaggregation of HDV truck emissions. This material is available free of charge via the Internet at http://pubs.acs.org.

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