Supporting Information for: Emissions per km of benzo[a]pyrene from US vehicles from 1961 to 2004 based on historical tunnel studies.

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List of abbreviations.

[Cat] and [No Cat] = "catalytic converter" and "no catalytic converter." Used as subscripts to delineate emission factors.

LDV = light-duty vehicle

HDV = Heavy-duty vehicle, including gasoline trucks. Excludes light trucks.

HDDV = Heavy-duty diesel

HD = Heavy duty

HDGV = Heavy-duty gasoline vehicle

MHDT = Medium heavy-duty truck

HHDT = Heavy heavy-duty truck

E_f = emission factor. $E_f^{LDV}(t)$ = emission factor for LDV's in year, t, etc.

Vp = ventilation parameter

g_1 = fraction of LDVs in fleet

g_2 = fraction of HDVs in fleet

k_1 = fraction of LDVs without catalytic converters

k_2 = fraction of LDVs with catalytic converters

f_1 = fraction of HD trucks that are diesels.

f_2 = fraction of HD trucks that are gasoline without catalytic converters

f_3 = fraction of HD trucks that are gasoline with catalytic converters

BAA = benz[a]anthracene

BaP = benzo[a]pyrene

BEP = benzo[e]pyrene

BGP = benzo[ghi]perylene

COR = coronene

FLT = fluoranthene

PER = perylene

PYR = pyrene

PAH = polycyclic aromatic hydrocarbon

Pb = lead

GIS = geographic information system

CO = carbon monoxide

CO_2 = carbon dioxide

C_{14} = carbon 14.

OC = organic carbon particulates

TSP= total suspended particulates

VOC = volatile organic compound

Results for other pollutants.

Other PAHs.

BaP and benzo[ghi]perylene (BGP) were measured in the earliest U.S. tunnel study carried out in 1961 in the Sumner Tunnel (*1*). In the second study in the Sumner Tunnel (*2*), carried out in 1963, additional PAHs were measured, namely coronene (COR), benzo[e]pyrene (BEP), perylene (PER), and pyrene (PYR). In the 1975 study in the Baltimore Harbor Tunnel (*3*), benzo[a]anthracene (BAA) and fluoranthene (FLT) were added to the list. The next tunnel study does not occur until the 1980s, when catalytic converters had already caused the dramatic reduction in BaP. Therefore, in what follows, we only focus on the individual PAHs measured by 1975.

Three PAHs measured in 1975 have no subsequent tunnel measurements of their concentration, so cannot be compared with any modern emissions data. These are dibenzo[a,e]pyrene (1,2,4,5-dibenzopyrene), dibenzo[a,I]pyrene (3,4,9,10 dibenzopyrene), and pentacene (2,3,6,7-dibenzanthracene)

Table S1. Particulate PAH[a)] emitted from tailpipes in tunnels, measured by 1975, relative to BaP.

Tunnel/Yr	BaP	BGP	COR	PER	BEP	CRY	BAA	PYR	FLT
Sumner, 61	1	2.49	0.80						
Sumner, 63	1	2.65	1.0	0.155	0.62			1.6	
Baltimore Harbor, 75	1	1.28			1.05	1.6	1.55	1.88	1.41
Lincoln/ Holland, 81	1	2.7	1.2		2.2		2.0		
Caldecott, 83	1	2.2		0.31	0.88	2.8		0.99	1.26
Baltimore Harbor, 86	1	1.38	0.810		0.86	2.1 b)	1.31	4.7	3.45
Caldecott, 89	1	4.15				0.72	0.57	1.63	1.68
Van Nuys, 93	1	5.5		0.21	1.19	1.30 b)	1.14	0.33	0.2
Caldecott, 96	1	2.21				2.03	2.54	10	7.3
Caldecott, 97	1	0.85				1.1	1.22	4.6	3.4
Allegheny, 99	1	0.049		0.27	0.2	0.36	0.33		0.45
Washburn, 00[c)]	1	2.1		0.58	2.56	3.8 b)	1.06	4.0	3.55
Caldecott, 04[d)]	1	1.7	0.88	0.16	0.96	CRY 0.88	0.93	1.38	0.84

a Abbreviations: BAA, benz[a]anthracene ; BEP, benzo[e]pyrene; BaP, benzo[a]pyrene ; BGP, benzo[ghi]perylene, COR, coronene; PYR, pyrene; PER, perylene; FLT, fluoranthene

b) Chrysene and triphenylene

c) Mixed tunnel fleet as listed by authors

d) 3.75% heavy-duty diesels in bore 1. ½ that percentage for both bores.

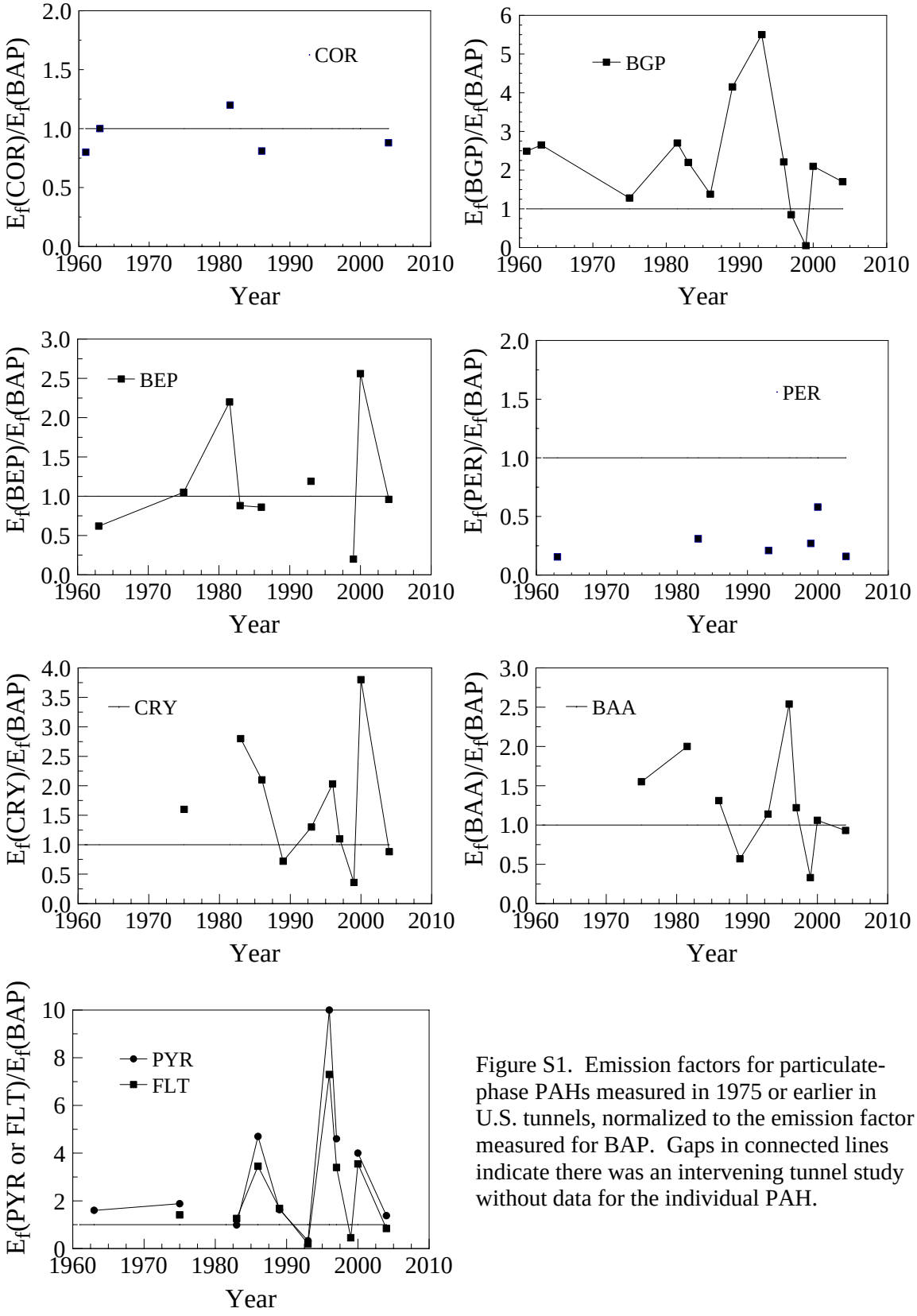
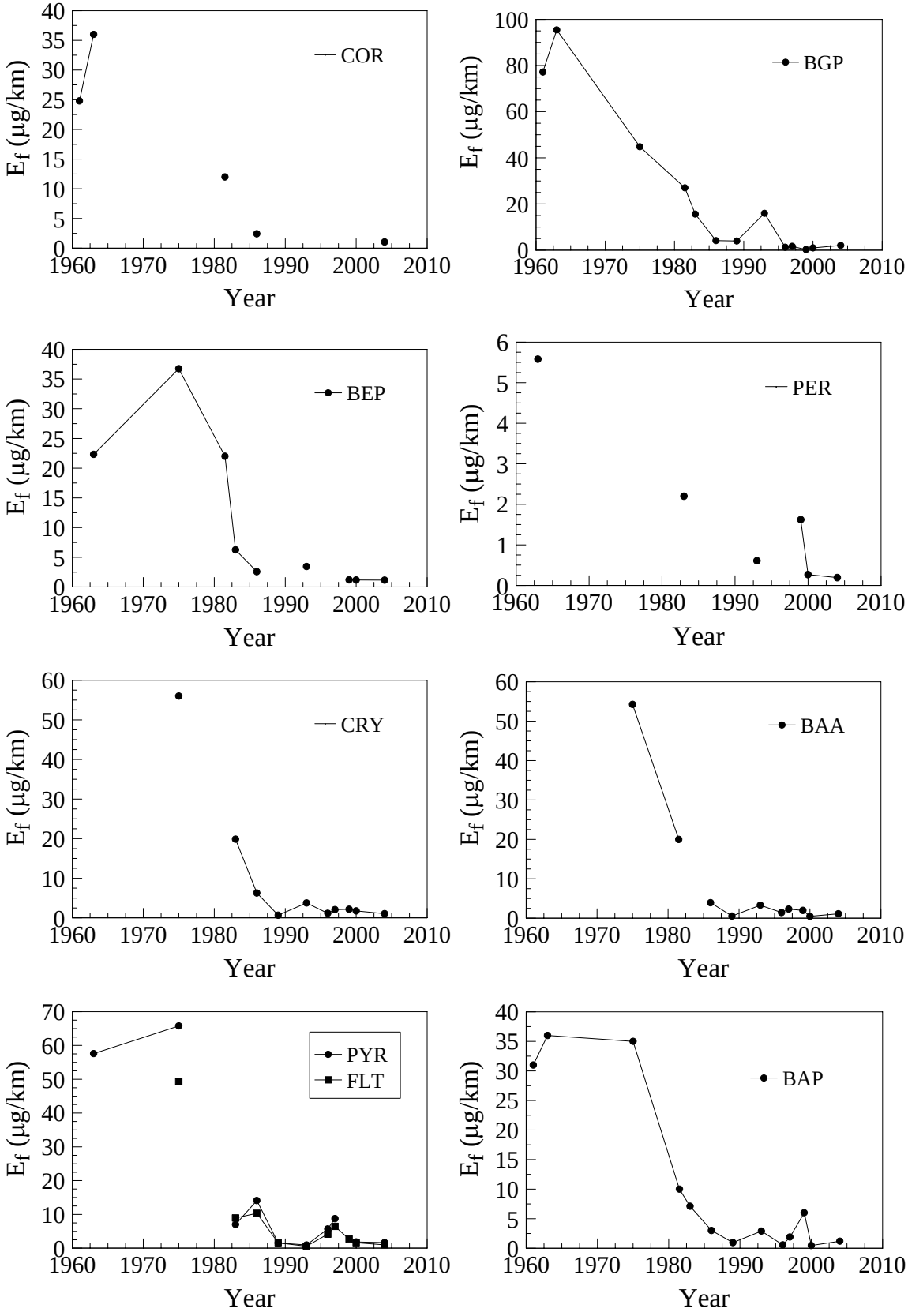


Figure S1. Emission factors for particulate-phase PAHs measured in 1975 or earlier in U.S. tunnels, normalized to the emission factor measured for BAP. Gaps in connected lines indicate there was an intervening tunnel study without data for the individual PAH.

Table S1 and Figure S1 show the emission factors relative to BaP for the eight PAHs for which there exists an extended time series of tunnel data. The numerical values were calculated by scaling the PAH concentrations obtained from the studies. The ratio of concentrations equals the ratio of emission factors, because BaP is being used as a tracer measured in the same tunnel in the same year as the target compound. (See Equations 1 and 2 in the main text.) Uncertainty in the ratios can range as high as 50%, based on fluctuations in separate ratios calculated from measurements made at different times in a study.

Figure S2 shows the absolute emission factors for the 8-additional PAHs, computed by multiplying the relative emission factors given in Table S1 by the BaP emission factors given in Table 2 of the main text. For comparison purposes, the graph for BAP, already shown in the main text, is added here as the last of the series. Uncertainties in the emission factors are not shown, but implicitly include the uncertainty ranges in the BaP emission factors scaled from Table 2, along with the uncertainty in relative calculations, which can amount to a factor of two.

Figure S2. Emission factors for particulate-phase PAHs measured as early as 1975 in U.S. tunnels. Gaps in connected lines indicate there was an intervening tunnel study without data for the individual PAH.


COR
BGP
BEP
PER
CRY
BAA
PYR
FLT
BAP
E_f (µg/km)
Year

Through 1981, extraction of BaP and the other PAHs from filters was carried out in a Soxhlet apparatus using benzene or cyclohexane -- solvents which have been measured to have identical recovery rates, approaching 100% for airborne particulates (*4*). When dichhloromethane (DCM, synonym methylene chloride) was the solvent in a Soxhlet apparatus (*5*), long extraction times (18 to 24 hrs) were used to ensure high recoveries of PAH (*6*). After 1981, use of Soxhlet extraction gave way to sonication methods, primarily using DCM. Sonication methods have been shown to produce recovery rates of PAHs, between 95% and 98% (*4*). As a result of the high recovery rates expected for all of the extraction methods used, we conclude that no historical bias was introduced into PAH emission factors by the shift in extraction technology over time.

Table S2. Relative potency on individual PAHs compared with BaP according to different authors[a)]

PAH	Larsen and Larsen (1998)[b)]	Range
Coronene (COR)	0.01	No other assignments
Benzo[ghi]perylene (BGP)	0.02	0.01-0.03
Benzo[e]pyrene (BEP)	0.002	0.0-0.004
Perylene (PER)	No value[c)]	No value[c)]
Chrysene (CRY)	0.03	0.001-0.89
Benzo[a]anthracene (BAA)	0.005	0.0-0.145
Pyrene (PYR)	0.001	0.001-0.081
Fluoranthene (FLT)	0.05	0.001-0.06

a) From Table 13 of Boström et al. (*7*).
b) As cited in Boström et al. (*7*) and used by them for most assessments.. Values were determined based on carcinogenicity studies in experimental animals using, oral, pulmonary, and skin application of PAH. The value for coronene is taken directly from the authors' paper (*8*).
c) Not classifiable as carcinogenic under IARC assessments (*7*).

All eight of the additional PAHs shown in Table S1 are considered to have low carcinogenic potency relative to BAP on a mass basis (*7*). See Table S2. PYR and FLT, however, can be emitted in much larger quantities than BaP, so could still be important compared to BAP, particularly FLT (*7*). Unfortunately, the data presented here for PYR and FLT do not include the vapor-phase component, which was 99% of the total measured in the 1993 study (*9*). This ratio seems, at first glance, to be inconsistent with measurements in the 1986 tunnel study, however, where Benner found the percentage of PYR and FLT vapor to be close to 50% (*6*), and with a ratio consistent with the atmospheric partition data of Yamasaki et al. (*10*) at the 5-7 ^{o}C temperature at which the Benner measurements were taken. Relative to BaP, Benner finds about 15 times more PYR and FLT than does Fraser, suggesting greater net condensation in the colder temperatures. Still, there appears to be a discrepancy, which we cannot resolve.

We found no other measurement of the vapor phase of PYR and FLT in tunnels prior to 1986 with which to compare. As a result, we do not consider the PYR and FLT data in the Figures as definitive for assessing the time trend of total emissions of these more volatile PAHs. The only other PAH in the Figures with a significant vapor component in some tunnels is BAA, where the ratio to particulate phase was found by Fraser et al. in 1993 to be about 50/50 (*9*).

Nevertheless, the graphs for COR, BGP, BEP, PER and CRY all represent total emissions of these PAHs. No obvious change in ratio to BaP occurs over the 40+ year period, indicating that these PAHs are showing the same qualitative drop in emission factors after 1975 that was presented for BaP in the main text.

Given the increase in percentage of the heavy-duty truck fleet that has occurred since the 1980s (see Table S9), it might be expected that the amount of PYR and FLT in the tunnel data would increase relative to BAP. Indeed, there is a trend in the PYR data amounting to a 7% increase per year, but the p-value is only 0.4. The association is even weaker for FLT.

Some of the non-BaP, PAH data indicate a greater reduction in emissions per vehicle km of more than the factor of 15 estimated for BAP in the main text. The high value for BaP in 1999 keeps the average of the post 1995 measurements relatively high. Is the 1999 value for BaP anomalous? The fact that the ratio of BaP in 1999 to all other PAHs, with the exception of PER, is unusually high would suggest so. On the other hand, we do not know the underlying distribution of high-emitting vehicles and their chemistry, which may be unusual, making it difficult to brand any set of data as an outlier. Nevertheless, removing the 1999 data point for BaP would increase the historical BaP-reduction factor well beyond 15.

The 1999 measurement is particularly important, because it is the only modern study where ventilation rates were measured at the time data was collected. All other modern measurements were tracer measurements. Could it be the case that ventilation-measurement methods systematically over-predict emission factors or that tracer measurements systematically under-predict them? If so, the drop in BaP emission factors stated in the main text might be overstated, amounting to only a factor of 5 or 6, not 15. This seems unlikely, however, because of the fact that measurements of other PAHs are not unusually high in the 1999 study, indicating that the ventilation-measurement method does not produce results that systematically differ from those produced by tracer methods.

Carbon Monoxide.

CO was measured simultaneously in many of the tunnel studies discussed in this paper. In addition, there have been numerous other tunnel studies that measured CO, but not PAH, including one sophisticated study in the Holland Tunnel as early as 1927 (*11*), which measured CO emission factors by grade in the tunnel. Table S3, below, lists the total of measurements that we have collected. We do not claim the list to be exhaustive.

Table S3. Carbon Monoxide Emission Factors measured in US Tunnels.[a)]				
Year	Tunnel	Citation	CO (g/km)	CO, Caldecott adjusted (g/km)[b)]
1927	Holland	(*11*)	69	69
1961	Sumner	(*1*)	49	49
1963	Sumner	(*2*)	63	63
1976	Allegheny[c)]	As cited in (*12*)	16	16
1979	Allegheny[c)]	" "	10	10
1981.5	Allegheny[c)]	" "	8.9	8.9
1983	Caldecott	(*13*)	14	8
1987	Van Nuys[d)]	As cited in (*14*)	13	13
1992	Tuscarora & Fort McHenry average	As cited in (*12*)	3.8	3.8
1993	Van Nuys [d)]	(*9*)	21	21
1994	Caldecott	(*15*)	12	7
1995	Lincoln	(*14*)	4.6	4.6
1995	Deer park	(*14*)	4.0	4.0
1995	Callahan	(*14*)	3.4	3.4
1995	Van Nuys [d)]	(*14*)	8.7	8.7
1995	Sepulveda	(*14*)	6.4	6.4
1996	Caldecott	(*16*)	7.8	4.4
1997	Caldecott	(*17*)	6.5	3.7
1999	Tuscarora	(*18*)	1.9	1.9
2000	Washburn	(*19*), (*20*)	6.3	6.3
2001	Caldecott	(*21*)	7.1	4.0
2004	Caldecott	(*22*), (*23*)	5.2	2.9

a) Emission factors have been either 1) taken directly from the study, b) converted from g/kg or g/l of fuel using fuel economy values given in the study or national averages, or 3) converted from CO concentrations using ventilation parameters inferred from other tunnel studies.
b) Most of the Caldecott studies were undertaken while vehicles were traveling up a 4.2 degree grade. Multiplying the uphill CO emissions per km by 0.57 brings uphill data to the average as determined from Figure 1 of Kean et al. (*21*).
c) The fleet in this tunnel is younger than average (*24*).
d) There is a lot of acceleration/deceleration at this short tunnel due to traffic lights at the entrance and exit (*14, 25*).

In Figure S3, we graph the CO emission factors given in Table S3. In the graph, we have excluded 3 data points from the Van Nuys Tunnel, because of the acceleration/deceleration that takes place in response to the traffic lights at the entrance and exit of this short tunnel. These considerations have been called upon to explain the higher than average emission factors measured in this tunnel for carbon monoxide (*14, 25*). Cruise emissions are not being measured in the Van Nuys Tunnel.

The first of the three graphs shows the CO emission factors extractable from those PAH tunnel studies listed in Table 1 of the main text. The middle curve shows the CO data from the PAH studies augmented with other tunnel measurements of carbon monoxide. The bottom curve takes into account that many of the Caldecott Tunnel studies were carried out on traffic traveling uphill only, whereas the other tunnels included travel down into the tunnel and back up again, with an average slope of approximately zero. There is sufficient CO data for the Caldecott Tunnel to allow simulating an average emission factor for equal travel both up and downhill.

Specifically, multiplying the uphill emissions per km by 0.57 brings uphill data to the desired average, based on averaging the downhill and uphill components of measurements shown in Figure 1 of Kean et al.(*21*). Even with such adjustment, there is a major difference between the CO curve and the BaP curve, with the CO emission factor reaching low levels earlier than BaP. Some of this difference may be accounted for by the fact that the 1976 and 1979 studies were carried out in the Allegheny tunnel, which has a fleet traveling at relatively high speeds and a mix of vehicles (*24*) that was apparently newer than the average US fleet at the time. Consequently, standardizing to conditions that held in most of the other tunnels would probably increase the 1976 and 1979 values, bringing the CO curves into a better match with the shape of the BaP curve. With the existing curves, it appears that the CO drop occurred earlier than did the drop in PAH. On the other hand, this disparity serves as further evidence that a major uncertainty in the PAH tunnel data is the timing of the sharp drop in emission factors.

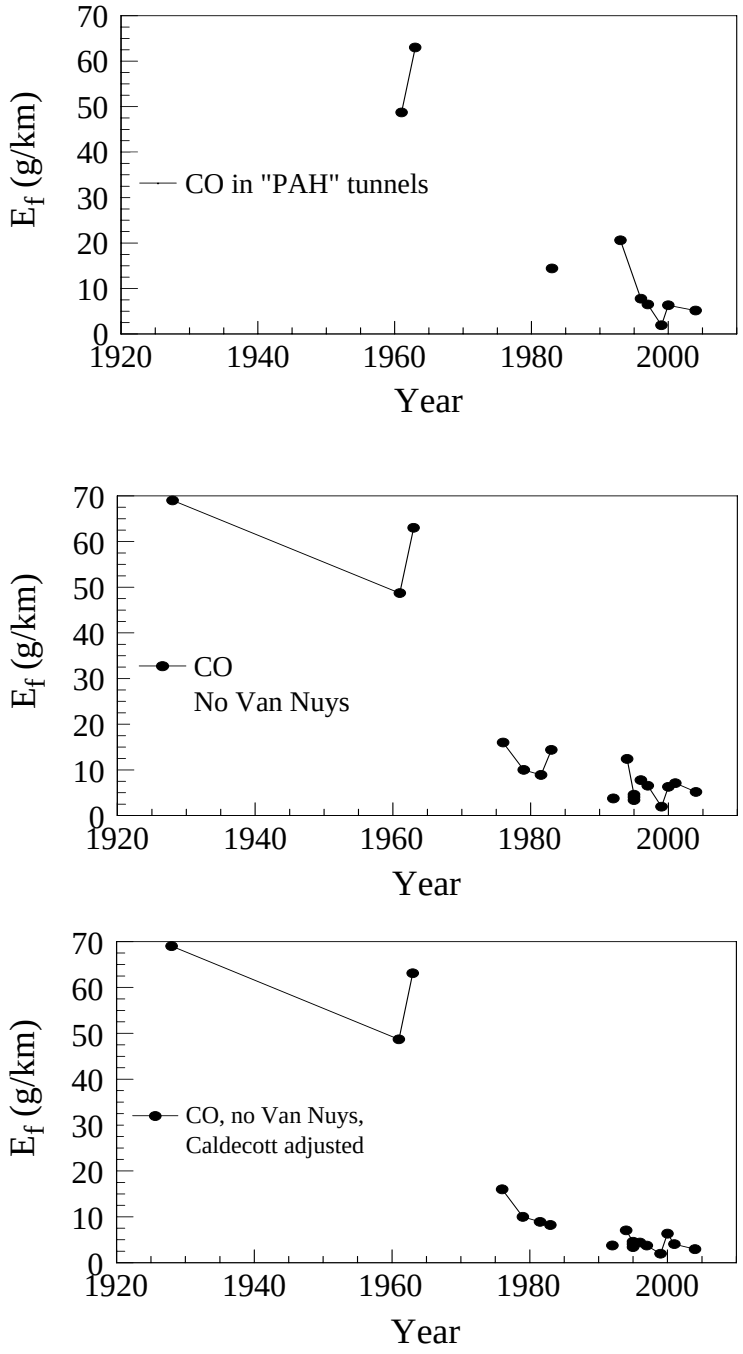


Figure S3. Emission factors for carbon monoxide as measured in tunnels. Top curve = values from tunnels in which PAH was also measured. Middle curve = data in top curve supplemented with other tunnel measurements. Bottom curve = Middle curve, but with Caldecott Tunnel points adjusted to compensate for the fact that data was collected only when vehicles were driving up-hill. Gaps in connected lines indicate there was an intervening tunnel study without CO data.

Volatile organic compounds.

The factor of 5 decline in individual VOC concentrations measured in the Lincoln Tunnel from 1970 to 1982 (*26*) is comparable to the PAH declines in emission factors reported here for the same period. (Note that the authors estimate a factor of 4 decline for all NMHC together.) The 1970 measurement is the only early VOC tunnel data we were able to locate.

To equate the change in ratio of concentrations in different years to the corresponding change in ratio of emission factors, using Formula 1 in the main text, requires assuming that the number of vehicles traveling through the tunnel per minute did not change dramatically, which is the assumption made by the authors. Although the traffic volume number could well be different, the difference cannot account for a factor of five decline.

The authors did not estimate emission factors, which prevents a direct comparison with VOC emission factors measured in the same tunnel in 1995 by Gertler et al. (*14*).

We used two methods to convert the 1982 concentrations in the Lincoln Tunnel into emission factors. First, we used carbon monoxide as a tracer, because its concentration was listed in the 1982 concentration data. (See Table S4.) To get an absolute emission factor for CO in 1982, we read off the 1982 value interpolating from the (last) graph in Figure S3, obtaining 8.5 g/km. Absolute emission factors for the 1982 VOC data in the Lincoln Tunnel were then obtained by scaling 8.5 by the ratio of individual VOC concentration to CO concentration, on the assumption that the traffic rates were roughly the same in 1982 and 1995 in the Lincoln Tunnel at the time of the respective measurements. This appears to be the case based on information given in the respective write-ups. In both studies, measurements were carried out in the morning rush hour, with average traffic volumes within 10% of each other. The results for 14 VOC or VOC combinations indicate declines ranging from a factor of 3.2 to 17, with an average factor of 6, (standard deviation = 3.8).

Table S4. VOC emission factors in the Lincoln Tunnel measured and inferred, using CO as tracer for 1982 data.

	Concen-tration ratio[a)]		Inferred emission factor	Measured emission factor	Emission factor ratio	Emission factor ratio
VOC	Lonneman 1970/1982	Lonneman 1982 (Table V) (*26*)	Lonneman 1982	Gertler 1995 (*14*)	1982/ 1995	1970/ 1995
	Ratio	Ratio to acetylene	mg/km	mg/km	Ratio	Ratio
acetylene	6.4	1	85	19	4.5	29
isobutane	5.4	0.42	36	4.5	8.0	43
n-butane	5	1.2	103	18	5.6	28
isopentane	4.2	1.9	162	50	3.2	14
n-pentane	4.4	0.92	78	17	4.7	21
3-methylpentane	4.7	0.47	40	10	4.1	19
2,4-dimethylpentane	5.1	0.44	37	3.7	10	51
cyclohexane	1.3	0.28	24	1.4	17	22
benzene	5.8	1.3	112	16	6.9	40
toluene	4.2	1.9	161	37	4.3	18
p & m - xylene	5.7	0.31	88	24	3.7	21
o-xylene	3.9	0.46	39	8.9	4.4	17
p & m ethyltoluene	4.1	0.60	51	11	4.6	19
1,3,5-trimethylbenzene	4.9	0.17	14	4.6	3.1	15
carbon monoxide		100	8500[b)]			
Average	4.5			16	6.0	26
Standard deviation	1.1			14	3.8	12

a) We assume that the concentration ratio is the same as the emission factor ratio, which means we are assuming that the traffic flows were similar in 1970 and 1982.

b) Numbers in the previous column were scaled until carbon dioxide equaled 8500, which is the 1982 emission factor in mg/km that we used for CO as a tracer, based on Figure S3. Traffic flows in 1982 and 1995 during the experiments were similar.

Combining the decline from 1982 to 1995 with the decline from 1969 to 1982 gives a net average reduction factor of 25 $\pm$ 12.

The second method we used to infer emission factors for the VOCs measured in the Lincoln Tunnel was to use the ventilation parameters inferred from the PAH measurement in this Tunnel. (See Table S7 and preceding discussion) By this method of converting tunnel concentrations to emission factors, the reduction in VOC emission factors between 1970 and 1995 was even greater than a factor of 25, namely a factor of 35. Considering the uncertainty in these methods, this is reasonable agreement between the two assessments. We take a mid-range value of 30 to use as the reduction factor in the following discussion.

A factor of 30 is twice as high as the reduction factor estimated for the BaP emission factor, although comparable to the reduction factors found for other PAHs. A factor of 30 is also higher than the factor that can be extracted from EPA estimates of the decline in total traffic-related VOCs for the same period: The USEPA estimates a factor of 3.7 decline in total VOC emissions from highway vehicles from 1970 to 2002 (*27*), which amounts to a factor of ten on a per-km basis, after accounting for the factor of 2.7 increase in miles driven over the period (*27*). A factor of ten reduction is much less than the factor of 30 estimated from the tunnel data for the set of VOCs measured in the Lincoln Tunnel and somewhat less than the BAP declines presented in Table 2. However, EPA estimates of VOC emissions cover the full driving cycle.

Furthermore, the estimate presented here of historical decline in VOC as measured in tunnels is uncertain, depending as it does on an indirect estimate of the CO emission factor during the rush hour in the Lincoln Tunnel. Gertler et al. only had usable VOC data for 3 out of 11 runs in their 1995 study, which we averaged over. And, with only three tunnel measurements over the 25-year period, there is not the opportunity to average out fluctuations in trends, which we have seen can be an issue in tunnel measurements with more data points, namely the data set of PAH emission factors. As an example of a fluctuation in tunnel measurements of VOC emission

factors, we note that a measurement of the emission factor for tolulene in the Allegheny Tunnel in August/September 1979 is 5-fold lower than the value we estimate for the Lincoln tunnel in 1982. Part of the explanation for the difference may lie in the different tunnel characteristics, namely the fact that the average speed is higher in the Allegheny and its fleet tends to be newer (*24*). In addition, the authors note that the values are probably lower by a factor of 1.3 to 1.7 because of the downgrade of 0.5% and the tailwind, both of which improve fuel economy. Fortunately, the VOC series on which we rely were all measured in the same tunnel (Lincoln), which should tend to reduce fluctuations due to tunnel characteristics.

Particulate organic carbon

In the 1961 and 1963 tunnel studies, measurements were made of the concentration of total benzene-soluble organics, which provide a rough measure of the particulate organic carbon concentration. We turn the tunnel concentrations into emission factors using the approximate tracer techniques discussed in this paper. In short, we scale the BaP emission factor of Table 2 by the ratio of the benzene soluble concentration to the BaP concentration in the outlet air, neglecting the tunnel input concentrations. This gives an emission factor of 0.11 g/km for the 1961 Sumner Tunnel study and 0.26 g/km for the 1963 Sumner Tunnel study. The Sumner studies are the only early US ones that we could locate that provided data on both PAH and organic particulate carbon. Pierson et al. measured benzene soluble organic emission factors in early 1970 tunnel experiments reported in a 1976 paper (*28*), indicating that the 1961 emission factor measured by Larsen et al. was in the range they measured. We focus, therefore, on the Sumner studies.

Unfortunately, extraction with benzene can significantly underestimate total organic carbon, because it misses the polar component (*29*), (*30*). However, the underestimate in the early 1960s studies cannot be too severe, because the total particulate concentrations measured in 1961 were only 2.0 times more than the benzene-soluble values and the total particulate concentrations measured in 1963 were 2.5 times more than the benzene-soluble component.

Another limitation in relating the benzene-soluble component to particulate organic carbon is the possibility that volatiles condensed on, and organic carbon (OC) evaporated from, the filters (*31*). In what follows we ignore this potential sampling artifact. Finally, we note that the Sumner Tunnel is not physically high enough to allow 3-axle heavy diesels to enter, so the emissions from diesels is particularly low in this tunnel. Nevertheless, it is of interest to compare the results to other fleet measurements in tunnels.

In 1979, Pierson and Brachaczek measured a combined fleet value of approximately 0.05 g/km OC in the Allegheny Tunnel (*32*), using dichloromethane as a solvent. The percentage of

trucks, however, was considerably higher than in the Sumner or Caldecott Tunnels (approximately 10% heavy diesels).

Kirchstetter et al have measured emission factors for organic carbon during uphill driving in the Caldecott tunnel, (*31*) which allows us to make a rough estimate of the reduction in OC emission factors over the years. Using regression analysis applied to data collected with differing percentages of HDDV, the authors provide separate emission factors of 0.053 g/kg for LDV and 0.5 g/kg for heavy-duty diesel vehicles (HDDV), as measured in the summer of 1997. They implicitly assume, as is reasonable, that the OC contribution from heavy-duty gasoline vehicles (HDGV) is small compared to that from HDDV. Using the fuel economy and fuel densities assumed in their paper, their emission-factor units can be converted to emission rates per km, namely 0.0047 g/km for LDV and 0.20 g/km for HDDV. Assuming a typical fraction of HDDV for the Caldecott of 2.7%, the combined fleet emission factor is 0.01, which is 5 times lower than the value measured in 1979 and 11 to 25 times lower than the emission factor for the non-polar component of the organic fraction in the early 1960s. Geller et al. measured OC in the same tunnel in the summer of 2004 (*23*) obtaining values for OC that lead to a fleet average 8.5 times lower by our calculations than measured in 1997, but Geller et al. were not able to include the ultrafine contribution to the mass, which makes interpretation of their data difficult. In any case, there appears to have been a reduction in OC emissions per km during cruise emissions of a factor of 11 to 25 from 1961 to 1997 and possibly still greater reductions thereafter. Once again, we have tunnel studies indicating a post-1970 reduction for another class of pollutant, namely the organic carbon component of fine particulates, although it is more difficult to be quantitative with OC over the 40+ year period than with the other pollutants.

Accounting for the unmeasured polar fraction in the Sumner extractions would increase the reduction ratios even further, as would accounting for the absence of 3-axle diesels in the Sumner Tunnel. Note that all of these comparisons are approximate, for one reason, given the large role that the truck percentage may play in OC emission factors and the relatively small

number of tunnel studies in the sample considered. Nevertheless, the results for organic carbon, as well as for VOC and CO, do show that the large reductions in BaP, which are the prime focus of this paper, are consistent with large reductions in other pollutants brought on by pollution controls.

Issues related to data quality and interpretation.

In this section, we discuss details that were not covered in the main text about the relative collection efficiency of devices used in the different studies and other limitations. We also discuss the significance of resuspension in interpreting tunnel data, and we add some miscellaneous details about other data on which the main text relies or mentions.

Can differences in tunnel characteristics explain away the large apparent reduction in emission factors?

As discussed in the main text, multivariate regression (*33*) 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 ($p = 0.56$). The results for univariate analysis were, $p= 0.0001$, $p= 0.79$, and $p = 0.035$, respectively.

We obtained average daily ambient temperatures from NOAA archives (*34*) for each day that authors made measurements. If the researchers collected data for more than one day, we averaged the daily temperature for those days. Note that the temperature in a tunnel generally rises in the interior, as much as 15 ^{o}C at some locations in long tunnels with heavy traffic (*35*). If we add 7 degrees to the ambient temperature assigned to the 5 urban tunnel measurements, the association with temperature in the multivariate regressions becomes stronger, with $p = 0.03$ for temperature, $p = 0.0003$ for year, and $p = 0.36$ for speed.

Tunnel grade is not likely to explain the BaP decline in emission factors. The grades are similar in the first set of studies in the urban East-coast studies, as discussed in the main text. Furthermore, from 1983 to 2004, we have a set of 4 measurements in the uphill direction of the Caldecott Tunnel, which show a decline in emission factor of 83% by the end of the period. Thus, the evidence is strong that there are two stages of major declines in fleet emission factors that cannot be explained by differences in tunnel grade.

Truck percentages were higher in three of the 12 studies listed in Table 2 of the main text. Arguments were made there as to why the contribution of HDV to the 1975 fleet average was relatively small. It is also worth noting that as long as the percentage drop in HDV emissions is less than the percentage drop in LDV emissions, which is reasonable from all of the available evidence, it follows that the percentage reduction in LDV emissions must be greater than the average reduction in fleet emissions, which was 57% from 1975 to 1981 and 91% from 1975 to 1986. It also follows that reducing the truck percentage, as in standardization to 4% trucks, would increase the reduction in emission factors and would not change any conclusion drawn in this paper.

Improvements in pollution controls (*36*), including recycling of crankcase emissions, improved efficiency of combustion, and catalytic converters are the most likely explanation for the decline in emission factors measured in tunnels over time, not differences in tunnel characteristics. Although tunnel characteristics are not likely to explain away the large drop in emission factors over time, the exact timing of the rapid decline in emissions is sensitive to the age distributions of vehicles in the various tunnel fleets during the time of rapid change of vehicle pollution controls. Fleet age distributions are not given in most of the tunnel studies, most significantly not in the studies carried out from 1975 through 1986.

Impact of particle size cutoff on historical collection efficiency of BaP. Research groups collected samples of BaP on particles with different size cutoffs. In this section, we provide additional backup for our conclusion that these differences in sampling technique did not significantly affect measurement of emission factors. Through 1982, tunnel data were collected with high volume samplers without cyclone or other cutoffs on particle size, so by definition the researchers were collecting total solid particulates (TSP). The data collected in 1983 had a 1.3-micron cutoff, but the 1986 data were again collected without size separation. By 1989 all of the BaP tunnel measurements excluded particles above PM2.5. The question arises as to whether or

not the high emission factors in the 1961-1975 tunnel measurements could be due to a difference in particle size cutoffs used in the different studies. Most importantly, could the high emissions in the early period consist of particles other than fine particulates that would not be relevant to health effects? This seems highly unlikely for the following reasons. While ambient samples in the early 1970s showed as much as two-thirds of BaP mass above 2 microns at two of eight sites studied in Toronto (*37*), measurements in 1977 in the Tsuburano Tunnel in Japan showed only 3.5% BaP mass above 2.1 microns (*38*). Similarly, ambient measurements carried out in 1976 and 1977 in Pasadena, a location heavily influenced by automotive emissions, revealed a unimodal distribution for PAH centered on very small sizes, with only 25% of the BaP mass above 0.26 microns (*39*). The most convincing evidence, however, that differences in particle-size cutoffs are not important comes from the tunnel emission factor results themselves. Comparison of the Table-2 results for 1981 and 1983 show very similar values for emission factors (10 μg/km vs. 7.1 μg/km), despite the fact that the 1983 study had installed a cyclone with a cutoff of 1.3 microns (*40*), whereas the 1981 study had no such equipment installed. The 1986 measurement (*5*) found a still lower emission factor (3.0 μg/km), even when collecting TSP, which suggests that there couldn't have been very much material above 1.3 microns to be excluded by cyclone cutoffs. The emission factor in the Baltimore Harbor Tunnel dropped from 35 μg/km in 1975 to 3.0 μg/km in 1986. Because both authors collected TSP, the decline cannot be due to differentially cutting out particles beyond 1.3 microns in size. Therefore, based on the behavior of emission factors in the 1980s, we can conclude that size cutoffs did not affect results in the period from 1975 onward. And, given the fact that the magnitude of the 1975 emission factor is comparable to the 1961 and 1963 values, there is nothing anomalous about the 1961 and 1963 data that would suggest the presence of any significant component of non-respirable particles.

Differences in filter types.

As stated in the main text, over the years, the filters that were used to collect PAH changed from glass and quartz fibers to Teflon coated fibers. To what extent might this have affected the relative efficiencies of BaP collection? Benner found no statistically difference in side-by-side samples of BaP collected on Teflon and glass filters in a Baltimore Harbor tunnel exhaust duct (*6*). Lee et al. found recoveries to be within 10% of each other on a variety of filter types, provided the extractions of the spiked C_{14}-BaP were made within one day (*41*). Grosjean found no differences, if the filters were left in the dark (*42*). However, if measurements were made 8 to 14 days after collection, relative efficiencies of glass and quartz could drop by a factor of 2 relative to Teflon filters, based on another lab study by Grosjean (*43*). Because most authors did not find any major difference in collection efficiency between these filter types and because pre-extraction storage time was not given in the tunnel sources, we decided not to apply correction factors derivable from Grosjean et al.

However, it is of interest to explore the sensitivity of our results to the possibility of differences between filter types, using Grosjean's results. For instance, the emission factor measurements made by Fraser et al. in September of 1993 in Los Angeles using quartz filters during daytime should be scaled by a factor of 1.4, based on averaging Grosjean's data over summer and winter to estimate the correction for fall. As for applying Grosjean's figures to other locations, it is not clear how generalizable results in the Los Angeles climate are to other climatic regimes, but we do so as part of the sensitivity exercise. For a number of studies using glass filters, there are no corrections to be made, because the measurements were carried out in daytime, and Grosjean's results for aged samples show differences only for nighttime samples on glass filters. However, researchers in Boston did make 24-hour measurements with glass fibers in September of 1961 and probably with glass in April of 1963 as well, although the type was not specified for the 1963 measurements. Averaging Grosjean's correction factors for glass over day

and night would imply a factor of 1.3 underestimate compared to what would be expected with Teflon filters.

For the concentrations measured in 1983 in the Northern California Caldecott Tunnel using quartz filters, we have scaled the winter data by 1.6 and the summer data by 1.2 to standardize to Teflon filters, in accordance with Grosjean's results for quartz filters. This adjustment does bring the summer emission factor closer to the winter emission factor (results not shown). For 1989 in the Caldecott Tunnel, we scale by 1.2 to match Grosjean's results for quartz-fiber filters used on a summer day. Similarly, we use a factor of 1.2 for the 2000 measurement in the Washburn Tuunel and the 2004 measurement in the Caldecott Tunnel, both of which involved quartz-fiber filters used in the summertime.

In summary, all of the potential correction factors are rather modest, when compared to the huge drop that occurred from the 1960s to the late 1980s, so no conclusion made in the main text would be changed, were the Grosjean corrections to be applied.

Resuspension in tunnels.

Researchers who study emission factors in tunnels generally assume that resuspension of a fine-particle pollutant can be neglected, if significant quantities are emitted from vehicle tailpipes. In contrast, Ondov considered the possibility that resuspension affected concentration of lead (Pb) in the Baltimore Harbor Tunnel during two-way traffic (*44*), because he found different results from measurements made during one- and two-way traffic. He focused on the fact that resuspension is higher during two-way traffic, because of the increased turbulence that occurs as vehicles pass each other in different directions. However, the specific non-linearity he found with traffic modes could have been caused by another effect of two-way traffic, namely the reduction of the extra ventilation caused by one-way movement of vehicles. Because we have measurements in the 1960s in the Boston Sumner Tunnel only two years apart, with and without two-way traffic, and because we have accounted for the piston effect, our results for those two years is an independent test of the magnitude of the resuspension contribution during two-way traffic collected in high-volume samplers. As shown in Table 2 of the main text, the two estimates of BaP emission factors for these years fell within 20% of each other, suggesting that, as is generally assumed in PAH tunnel studies, resuspension is not an important issue for BaP.

Miscellaneous Data issues:

There are two studies that we located that are not listed in Table 1 of the main text. First, a study in Chicago (*45*) was not used, because measurements were taken atop the tunnel for safety reasons, not inside (Khalili, N., Illinois Institute of Technology, personal communication, 1998). In any case, a lack of ancillary data would have prevented extraction of emission factors. There were neither accompanying ventilation measurements, nor simultaneous measurement of carbon monoxide (CO), carbon dioxide (CO_2), or lead (Pb) that would allow use of tracer methods.

The other study we did not use reported measurement of BaP in one of the California tunnels (*14*), but the sensitivity was so low that we did not include its zero finding in our main analysis. The error band includes other results we have for the same time period.

There is a misprint in the 1961 study in the Sumner Tunnel: the authors present BaP emissions as 491 μg per vehicle mile (*1*), which we reduce by a factor of ten to 49.1 ug/mi, based on the correction noted in subsequent papers (*46*), (*47*) and convert to metric units, for a net value of 31 plus or minus 12 μg/km.

Some of the 1983 BaP concentration values for the Caldecott tunnel have been published (*48*), along with measurement details (*40, 48*); the remaining concentration values that we have used were taken from an unpublished report by the same group (*13*) provided to us by the late Joan Daisey (personal communication, 1998) and discussed with Antonio Miguel (personal communication, 1998, 2006).

Tunnel studies make the assumption that that there is complete mixing of air within the vertical plane of the tunnel. Studies by Pierson and Brachaczek suggest that this is a reasonable assumption (*28*).

In some cases, traffic rates were needed by us to make emission factor estimates, but were not measured in the study. To analyze the 1975 data in the Baltimore Harbor Tunnel (*3*), we used average traffic flows for the tunnel for that year obtained from the tunnel authority (Lisa Grant, Maryland Department of Transportation, personal communication, 2007). For the 1989 study in the Caldecott Tunnel we interpolated between traffic data given in the studies carried out in 1983 and 1996.

The study by Chellam et al. in the Washburn Tunnel in Texas (*19*) listed emission factors for a number of PAHs as a function of the percentage of heavy-duty diesels (HDDV), but no emission factor data for BaP was presented. The authors did provide concentration measurements for BaP and the other PAHs, which was enough information to allow extraction of the average

BaP fleet emission factor by scaling the emission factor for another PAH, e.g., Fluoranthene (FLA), by the ratio of BaP to FLA concentrations. See Equation 2 in the main text. The results of this scaling using FLA agreed with similar calculations for two other PAHs, Pyrene and (C23) n-tricosane. The average of the three results was presented in Table 2 of the main text. An alternate approach to obtain the BaP emission factor, which gives the same answer, is to directly use the mass-balance equation required for the carbon-tracer method (*20*), obtaining the required values for excess tunnel concentrations of carbon monoxide and carbon dioxide from data given in an earlier publication (*20*).

For BaP, the intercept at zero HDDV turns out to be negative, indicating a problem with the HDDV-regression method. Nevertheless, the average of the BaP emission factors still gives us the fleet average. The same numerical value we compute is presented in a summary Table in a later study (*22*),

Emissions during cold starts and at intersections.

Depending on the sophistication of the modeling effort, emission values determined for cruise conditions can be adjusted to account for non-cruise operating conditions, such as cold-engine operation or acceleration and deceleration at traffic intersections. We discuss one such approach below, derived from our review of relevant vehicular studies, including chassis-dynamometer studies.

Total emissions from a roadway segment of length, L, are given by the product of the number of vehicles and the emissions per vehicle-km times L. Total emissions have a characteristic time profile, both for weekdays and weekends (*49*), which shows up in measurements of BaP and PAH air concentrations near roadways (*50-52*). Some of this variation is due to differences in traffic rate over the course of a day, but not all of it. Tailpipe emissions also vary with time of day, increasing during cold starts, which tend to occur more frequently in the morning.

Cold start emissions increase because a richer air/fuel mixture must be employed to ensure the presence of sufficient fuel vapor for combustion (*53*). The variation in BaP emissions has been studied as a function of the richness of the air/fuel mixture and can serve as an indicator of how emissions will change under cold start conditions. For instance, Hoffman found that emissions increased ten-fold when engines were running rich with a surplus of fuel compared to running lean (*54*). Pedersen found a three-fold increase in BaP emissions with the engine operating on a rich mixture (*55*). There is another effect to consider. Colder temperatures lead to longer warm-up times and a longer duration of running rich.

Table S5. Ratio of BaP emissions during the cold and warm cycles of the USEPA's urban driving test.[a)]

Study	Year of measurement	Ratio of cold-cycle emissions to warm-cycle emissions
Hoffman (*54*)	1971	30[b)]
Williams (*56*)	1979	4 to 6[c)]
Lang (*57*)	1981	6[d)]
Stenberg (*58*)	1983	2 to 5.4[e)]
Joumard (*59*)	1996	12.5 to 17[f)]
Ahlvik (*60*)	1997	8[g)]

a) With the exception of the 1971 measurement, all researchers used the 1975 version of the Federal Test Procedure (FTP) (*53*).
b) These data were obtained with a U.S. vehicle with a 283 CID, V-8 engine equipped with air injection, however, similar results were obtained with a vehicle equipped with an engine modification emission control system.
c) The 4-ratio is the average of 4 U.S. vehicles with catalysts; the 6-ratio corresponds to average results for 7 vehicles without catalysts
d) Based on 4 U.S. vehicles, apparently with catalysts
e) The authors studied 3 of the most popular vehicles sold in Sweden, with approximately 12 vehicles in each class. Average ratios were 2.0, 3.5, and 5.4, respectively
f) The 12.5-ratio corresponds to the average of 10 non-catalyst vehicles in the French fleet. The 17-ratio corresponds to the average of 10 vehicles with three-way catalysts.
g) 1 vehicle from the Swedish fleet with a three-way catalyst. Mileage = 90,000 km.

To make this type of information useful for estimating emissions, it is helpful to quantify the ratio of BaP emissions during cold starts compared to the normal driving cycle. To obtain such a ratio, we collected data from the literature (as shown in Table S5). By inspection, the ratios fall in the range of 8 plus or minus 4.

Knowing the ratio of emissions during cold starts is not useful for modeling without additional information about the numbers that occur. To obtain information on the distribution of light-duty vehicle (LDV) cold starts over the course of a day for use in exposure modeling, it is

possible to consult traffic diaries, which can be queried to give the number of trip starts by hour of day. For instance, trip-start data for the US can be obtained on a regional basis from the travel-diary database of the National Personal Transportation Survey (NPTS) (*61*). In geographic-information-system (GIS) models of exposure, morning cold start emissions can be restricted to residential neighborhoods, further improving spatial accuracy. We did not identify data useful for modeling cold-starts of heavy-duty trucks, however.

Another relevant modeling parameter is the extent of the cold-start period, Ahlvik et al. measured the distance to be 1-km (*60*). However, data in Joumard et al. suggests it might be longer (*59*). Stenberg et al. state that the effect lasts for 3-4 km (*58*). Modelers who account for cold-start variations will need to run sensitivity tests on this parameter. In the Table, we simply presented the range of values we found in the literature, namely 1-4 km.

Cold-start emissions may also vary by season, because increased use of the choke can be expected to increase in winter. The literature does not give much quantitative information about winter vs. summer tailpipe emissions of BaP. Handa et al. found agreement for BaP with calculated values in summer and winter without needing a seasonal separate emission factor (*62*), while others have reported BaP concentrations to be about 5-times higher in winter than summer (*63*, *64*).

Possible explanations, other than increased winter-time auto emissions, include greater heating emissions in winter, less chemical removal, and less atmospheric mixing (*63*). By itself, reduced atmospheric mixing in winter accounts for about a factor of two in the winter/summer ratio, based on our own meteorological calculations in the New York Metropolitan Area using the emissions estimates discussed here and a meteorological model developed for our work with PAH traffic exposure (*65*). Chemical reactions in the mid-climate region of New Jersey during summer were estimated to account for a 26% difference in BaP air from winter (*63*). The contribution of BaP from winter heating is difficult to disentangle because the data are inconsistent by climate region. The winter/summer ratio in Ontario was only about a factor of

two (*66*), much less that in warm climates such as Los Angeles (*67*) where summertime chemical reactions may dominate.

Based on these considerations, we doubt winter-time BaP-emissions from vehicles will exceed summertime emissions by more than a factor of two in mid-climate regions of the United States. A similar analysis would hold for other climates: In colder climates similar to Ontario's, the net winter/summer ratio there was only a factor of two to begin with. And for warm climates, the winter engines wouldn't be very cold in the first place.

Tailpipe emissions also vary with driving conditions: emissions of both PAH and carbon monoxide (CO) near intersections increase (Sheu et al. 1996a; Sheu et al. 1996b), (Sculley 1989), because of the increased amount of starting and stopping. Begeman and Colucci studied the emission of BaP during acceleration and deceleration on a chassis dynamometer (*68*). Based on their paper, it is appropriate to scale PAH-emissions near intersections upward by a factor of five, when traffic lights or stop signs force deceleration and acceleration. Our own work fitting traffic emission models to BaP soil data suggest increases at intersections by more than a factor of ten (*69*), but we were unable to tease out separate values for the scale factor and distance from the intersection over which increases hold.

We were unable to find any other quantitative data to support the numerical value of a specific finding for intersections, which means that sensitivity tests should be performed on results obtained with an intersection scale factor.

Because the literature base is limited, it will be difficult to use previous data to adjust for cold-engine operation or acceleration and deceleration at traffic intersections. A modeler may want to optimize model parameters on some set of field measurements (69). Possibly, some information can be gleaned from chassis dynamometer studies, which are usually programmed to replicate standard urban driving cycles. However, dynamometer studies have their own limitations, for instance, they may not include the right mix of poorly performing vehicles.

Details of Calculations

In this section, we fill in the details of a number of calculations referred to in the main text.

Fits to concentration data collected in 1963 along the Sumner Tunnel.

The 1963 study in the Boston Sumner Tunnel did not include any measurements of ventilation rates, unlike the 1961 study, which meant that the 1963 researchers could not calculate an emission factor using the 1961-methodology. However, the researchers did publish concentration data as a function of distance into the tunnel that allowed us to estimate an emission factor. The details of the calculation are presented here.

The 1963-researchers collected concentration data for benzene-soluble particulates, which we assume is proportional to the BaP concentration. We fit the data for benzene-soluble particulates to the expected function of distance given in Chang and Rudy (*70*), with the deposition velocity put to zero:

$$C_t(X) = A\left(1 - e^{-B(X-D)}\right).$$

Eq. S-1.

C_t is the concentration in the tunnel, and, X, is distance. In the notation of Chang and Rudy,

$$A = \frac{q}{\alpha} + C_{ti}, \qquad B = \frac{\alpha}{u}, \qquad BD = \ln\left(1 - \frac{C_t(0)}{A}\right),$$

Eq. S-2.

$$q = Ef\left(\frac{N}{Area}\right), \quad \alpha = \frac{R}{Area \times Length},$$

Eq. S-3.

where, u, is the exit velocity caused by the piston effect and natural winds, $C_{t,i}$ is the outside concentration, $C_t(0)$ is the concentration at the tunnel entrance, *Ef* is the emission factor, *N* is traffic rate (e.g., vehicles/minute), *Area* is the area of the ventilated tunnel tube, *Length* is the length of the tunnel, and *R* is the ventilation rate in m^3/s. The relations in Eq. S-2 hold for zero deposition velocity. As stated in the main text, deposition of fine particulates has been studied in tunnels (*28, 44, 71*), with the conclusion that it can be neglected for fine particles, with the exception of SO_2 and SO_4

To obtain the parameters, A, B, and D, we used a packaged routine for fitting this "monomolecular equation" (*33*). 4 data points are required to produce a fit to this equation. To get a 4^{th} point, we inferred the entrance concentration, choosing an initial value of 24.5 ug/m^3, which lay half way between the measured concentration at the intake air vent (8.3 ug/m^3) and the toll both (40.7 ug/m^3). Because the precise value was uncertain, we varied $C_t(0)$ until the fit was self-consistent, which means in this case that the fitted parameter *D* should equal the value predicted from the tunnel equation, namely, $1 - C_t(0)/A$. The resulting estimate for $C_t(0)$ of 21.5 ug/m^3 was within 20% of the initial, interpolated value.

Dividing the asymptote, *A*, by the measured concentration in the exhaust duct, gave us the scale factor (numerical value = 2.3) that we needed to convert exhaust data collected in one-way traffic to the value it would have had under two-way conditions in the Sumner Tunnel. Once the adjustment is made to remove the effect of the extra piston ventilation, the exponent in equation 1 of the main text becomes unity.

We could also have computed the scale factor using, not the actual measured exhaust concentration, but the value predicted by the tunnel equation. This increases the scale factor by 20%. We interpreted this 20% to be an indicator of the uncertainty in the scale factor.

Ratio of exhaust-tunnel concentration to traffic-tunnel concentration at the same location in a transversely ventilated tunnel.

There were two "opportunistic" studies in the Northern California Caldecott tunnel for which we converted concentration measurements to emission factors using an inferred ventilation parameter that was computed from the average of three, later complete studies. To make this conversion, we first had to take into account the fact that data were collected in the exhaust tunnel for the opportunistic studies, but in the traffic tunnel in the complete studies. Thus, to use the ventilation parameters derivable from the complete measurements, we had to have a formula for converting from traffic tunnel concentrations to the concentration overhead in the exhaust tunnel. We also needed this formula to complete the assignment of an error rate to the 1975 opportunistic data collected in the Baltimore Harbor Tunnel (BHT). For that study, we did not know the location of collection, whether in the exhaust duct or in the tunnel itself.

The measurements in the Caldecott and BHT tunnel (like all the other tunnels considered with the exception of the Tuscarora on the Pennsylvania Turnpike and the Van Nuys in Los Angeles) were made while intake and exhaust fans provided "transverse" ventilation through periodic openings in the tunnel walls and ceiling. Additional ventilation was provided by the one-way movement of the vehicles (so-called piston effect), which carries a percentage of the pollution generated in the tunnel out with the traffic, rather than out with the exhaust-duct air.

In the absence of resuspension and deposition in an exhaust duct in a transversely ventilated tunnel, the concentration in the duct at a particular distance equals the average of the tunnel concentration up to the same distance (*44*). This is simply a consequence of conservation of mass. As a result, the concentration in the exhaust duct cannot exceed the concentration in the tunnel. Integrating the tunnel equation (Eq. S-1) out to a distance X and dividing by X to get the average concentration in the tunnel, which equals the exhaust concentration, gives

$$C_e(X) = A\left(1 + \frac{e^{-B(X-D)} - e^{BD}}{BX}\right).$$

Eq. S-4

Where C_e = the exhaust concentration and L is the length of the ventilated section. The ratio of tunnel exhaust to tunnel concentration, C_e/C_t, is then

$$\frac{C_e(X)}{C_t(X)} = \frac{1 + \frac{e^{-B(X-D)} - e^{BD}}{BX}}{1 - e^{-B(X-D)}}$$

Eq S-5

For the Caldecott Tunnel measurements, we are interested in the concentrations near the end of the tunnel, where X ~ L. Taking limits in that case in Eq. S-5, which requires an expansion to second order, it follows that the ratio, Ce/Ct, has a minimum of 0.5 and a maximum of 1. The lower limit corresponds to minimal transverse ventilation and a linearly increase in pollution concentration along the tunnel, the solution to the tunnel differential equation when the piston effect is the only ventilation (*70*).

Given the bounds on the ratio of 0.5 to 1, we have picked 0.75 as a mid-range value for it, assigning an uncertainty range of $\pm$ 0.25, which spans the full range allowed.

Derivation of Equation 1 of main text.

We wish to derive Eq. 1 of the main text:

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

Eq. 1 (of main text)

which was used to convert opportunistic concentration measurements into emission factors. To obtain our central estimate of an equation to use in making the conversions, we have relied on numerical solutions to fluid-dynamic equations graphed by Schlaug and Carlin (*35*), recognizing that the calculations by Schlaug and Carlin were for a composite tunnel. The dependency on traffic plotted in Figure 5-4 of Schlaug and Carlin has pollutant concentration varying approximately as the two-thirds power of traffic rate over the range of traffic rates of interest in our study. This implies an exponent, w, in the above equation of 0.66 for the composite tunnel.

To account for uncertainties in applying the equation to specific tunnels, we begin by bounding the results with limiting cases. We use the steady-state tunnel equation (see Eqs. S-1 to S-3) to derive the following equation, which holds under the approximations listed in the main text, namely neglect of the background concentration in intake air, both at the fans ($C_{t,i}$) and at the entrance to the tunnel ($C_t(0)$). With these approximations, the full equation is,

$$Ef(2) = Ef(1)\frac{C(2)}{C(1)}\left(\frac{N(1)}{N(2)}\right)\left(\frac{1-\exp\left(-\frac{R}{Area\times u(1)}\right)}{1-\exp\left(-\frac{R}{Area\times u(2)}\right)}\right)$$

Eq. S-6

where, R, is the mechanical ventilation rate, and, u, is the air velocity at the tunnel exit, caused by the piston effect of the traffic and any significant natural wind. "Area" is the area of the tunnel. (An alternative to neglecting, $C_t(0)$, is to define a new concentration variable that equals the original minus $C_t(0)$, which leads to a replacement of the concentration variables in equations like S-6 with the term, C-$C_t(0)$.)

The correctness of Eq. S-6 can be established by replacing the concentrations, C(1), and, C(2), with the appropriate formulae obtained from Equation S-1, i.e.,

$$C = A\left(1 - e^{-B(L-D)}\right)$$

"L" is the tunnel length. The constants A, B, and D need to be expanded in terms of the physical parameters given in Equations S-2 and S-3, with $C_{t,I}$ and $C_t(0)$ set to zero, which gives,

$$A(t) = Ef(t)\left(\frac{N(t)}{Area \times R}\right)L,$$

$$B(t)\,L = \frac{R}{Area \times u(t)}$$

and, D = 0.

Eq S-7

The resulting expression for the emission factor in year, 2, Equation S-6, depends on the exit air velocity, u, which is a complicated function of traffic rate, requiring numerical solutions of the relevant equations of fluid dynamics. However, there are two limiting situations where we can find the value of, u, and hence, w, analytically. For the case, $u \cong 0$, i.e., when (Area * u) is very small compared to the mechanical ventilation rate, R, we recover Eq. 1 of the main text, with the exponent, w, equal to 1.

On the other hand, when, u, is very large, i.e., the mechanical ventilation rate, R, is relatively low, we can expand the exponential in Eq S-6, obtaining:

$$Ef(2) = Ef(1)\frac{C(2)}{C(1)}\left(\frac{N(1)}{N(2)}\right)\left(\frac{u(2)}{u(1)}\right)$$

Eq S-8

Under this case, when mechanical ventilation is negligibly small, Schlaug and Carlin, in their Equation 5.1, present an analytic expression for the exit velocity due to the piston effect, assuming constant vehicle velocity:

$$\widetilde{C} + \left(\frac{C_D A_V}{A}\right)\left(\frac{(N \times L)}{V}\right)(V-u)^2 = \left(\xi_i + \xi_e + \left(\frac{\lambda_L}{D}\right)\right)u^2$$

Eq. S-9

The first term,

$$\widetilde{C}$$

represents wind, barometric and temperature effects, which we neglect compared to the piston effect. N is the traffic rate, V, is the traffic velocity, u, is the exit air velocity. All the other terms are independent of these values, so can be treated as constants for a specific tunnel. As long as the speed of the traffic is much greater than the exit wind velocity, which is going to be the case in tunnels of interest under the assumption of negligible natural wind ventilation, we can treat the term, $(V - u)^2$, as a constant. Then, we can solve for, u, obtaining a value that is proportional to the square root of the traffic rate. Substituting such a term into Equation S-8 produces Equation 1 of the main text with an exponent, w, of one-half.

We now have three cases for Equation 1 where raising traffic rate to a power, w, is the appropriate formula. As stated we take our, “best,” value to be w= 0.66, based on the full numerical calculations in a composite tunnel. To account for uncertainties in applying this equation to other tunnels, we adopt the w-parameterization for all tunnels, using a triangular probability distribution for, w, with a peak at 0.66 and bounds at 0.5 and 1. The mean of the w-distribution is 0.72.

Details of the Pb-tracer method.

We were able to use lead as a tracer gas, because the historical vehicle emission rates for lead have been established in separate tunnel studies, allowing us to use a simple concentration scaling approach to infer BaP-emission rates. With this method, where we assume the tunnel concentration at the measurement point is much greater than the tunnel input and ambient background, we used the following equation, which was given in the main text.

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

Eq. S-10

We obtained the fraction of the vehicle fleet, *S(t)*, that consumed leaded gasoline over the years from compilations (*72, 73*). To obtain the amount of lead in a gallon of leaded gasoline in a particular year, $C_{pb}(t)$, we used a variety of sources. For 1961, we used data collected by tunnel researchers themselves to obtain an average consumption figure, $S * C_{pb}$, of 0.49 g/l (*1*), which agreed with nationwide figures (*74*). For 1963, we interpolated between the 1961 content of leaded gasoline and 1965 values given by Newell et al. (*72*). The result was an average consumption, $S * C_{pb}$, of 0.52 g/l. For 1975 we were able to average over 3 estimates (*32, 72, 75*). The resulting average consumption was 0.45 g/l. For 1981 and 1982, we averaged over two estimates (*72, 75*), obtaining a value for 1981/1982 of 0.15 g/l, which we assign to November of 1981, which is when the Lincoln/Holland Tunnel study was made. California put limits on lead content in gasoline as early as 1976. We, therefore, obtained a separate estimate (0.091 g/l) for use with a 1983 tunnel study in California, using data from the California Energy Commission (*73*). We assumed a value of zero for Pb-emissions from diesel trucks (*76*).

To obtain the fleet fuel economy, *M(t)*, in a particular year, we relied on compilations or individual values from a number of authors (*1, 77, 78*). For the light duty vehicle fleet, we

combined separate figures given in Davis and Diegel for passenger cars and light trucks (*77*). For heavy-duty *gasoline* trucks, we used the corresponding figures in Davis and Diegel for heavy-duty, single truck units. To determine the percent of trucks that consumed gasoline we relied on data discussed later in the section on heavy-duty vehicles. See Table S-9 for the percentage of trucks by vehicle type in each tunnel fleet.

To obtain lead *concentrations* in the Baltimore Harbor Tunnel in 1975 and 1986, we extrapolated from data measured in 1973 (*44*), scaling for differences in traffic rates raised to the *w*-power and adjusting for the known decline in lead in gasoline over the period. The origin of the exponent, *w*, and its distribution, was discussed in the main text and in this document.

Uncertainty in estimates of emission factors.

In this section, we discuss the details of the calculations of standard deviations listed in Table 2 and displayed as error bars in Figure 1. Error rates were propagated throughout the calculations using Monte Carlo techniques available in the Crystal Ball add-in to the Excel spreadsheet (*79*). As much as possible uncertainties given explicitly in the studies were used, but especially in the earlier work it was necessary to infer error rates based on other contemporaneous studies. Lognormal distributions were assumed for unbounded variables; triangular or uniform distributions were assumed for bounded variables. The Monte Carlo output distributions were approximately lognormal.

For a general, qualitative review of PAH sampling techniques and issues relevant to much of the early time period, see Leinster and Evans (*80*). Most of the measurement difficulties were associated with the lighter PAHs, not BaP. To quantify BaP measurement uncertainties, we relied on work of Sawicki, assigning a 26% uncertainty to extractions using the Soxhlet aparatus (*66*). We took uncertainties in traffic rates as 20%, based on information provided in those studies that discussed them (*5, 6, 13, 44, 81, 82*). We assigned a 20% uncertainty to the amount of lead in gasoline based on the range of different estimates for similar years. The uncertainty in the fraction of lead emitted as fine particles was taken to be 20%, based on the range of estimates made in tunnels, as discussed in the main text. Uncertainties in measurement of airborne lead particulates were taken as 20%, based on the thesis work of Ondov (*81*). Uncertainty in fuel economy was taken to be 20% based on the range of on-road measurements that have appeared in the literature for the Caldecott (*16, 83, 84*) and other Tunnels (*18, 85-87*), as well as the national fuel economy numbers we generated from Davis and Diegel. A 15% uncertainty was assigned to ventilation rate measurements (*32*). There is also uncertainty to be assigned when we have a data point, but no information on the exact time of day when the measurement was taken. This occurred for the 1975 and 1989 data points. For the 1975 measurement in the Baltimore Harbor Tunnel, we relied on variations in BaP measured in the same tunnel over the course of the day in

1986 (*6*). We concluded that the 1975 measurement should be assigned a 10% uncertainty for this factor, because the variation measured by Benner was quite small. Another 20% was assigned, because we do not whether the measurement was carried out in the exhaust ducts or carried out in the tunnel itself.

For the 1989 measurement in the Caldecott Tunnel, where once again we did not know the time of day at which the measurement was taken, although we did know the duration of the measurement, namely 4 hours (*88*). To obtain an estimate of how much the concentration varied over an average day, we relied on the measurements carried out in 1983 in the same tunnel (*13*), which led us to assign a 30% uncertainty to this factor.

The treatment of uncertainty in the ratio of concentrations in the tunnel exhaust and traffic bore, as well as the uncertainty in the effect of traffic on the piston effect, was discussed in the main text. To work with uncertainties in the inferred emission factors, it is useful to define a ventilation parameter, Vp, by collecting terms in the equation for the emission factor introduced in the main text (Equation 1), which relates emission factors at different times, t = 1, and, t = 2:

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

Eq. S-11

Ef is the emission factor, C is the concentration, N is the traffic rate, e.g., vehicles per minute, and, *w*, is the exponent discussed in the main text and earlier in this paper. We define the ventilation parameter, Vp, as follows:

$$Vp = \frac{Ef(1)}{C(1)}N(1)^{w}.$$

Eq. S-12

As with all calculated uncertainties, we compute its numerical value using Monte Carlo techniques. Equation S-11 can now be approximated as,

$$Ef(2) = <Vp\ (1)> \frac{C(2)}{N(2)^{w}},$$

Eq. S-13

where $<Vp(1)>$ is a log-normal variable with median and geometric standard deviation obtained from Monte Carlo calculations using Eq. S-12. This allows a straightforward computation to be made of the overall uncertainty in Ef(2), once the uncertainty in Vp is computed and combined with the uncertainties in concentration, traffic rate and traffic-rate exponent. The same distribution for, *w*, is used in all calculations.

To determine, Vp, and its uncertainty for the Caldecott Tunnel, we averaged over three sets of data collected in "complete" studies in 1996, 1997, and 2004 in two separate tunnel bores, numbers 1 and 2. We matched tunnel-bore data to the conditions that held for the data collected in "incomplete" studies. In particular, when determining, Vp, for use with the (incomplete) 1989 study, where opportunistic data was available only in a bore in which heavy duty vehicles were allowed (Bore 3), we obtained, Vp, from the data we had for a bore that allowed heavy-duty vehicles, namely, Bore 1. To account for the fact that Bore 3 was downhill, we used a Bore-3 figure for fuel economy (*21*) to convert from BaP per kg of fuel to BaP per km, assuming that the emission rate per liter of fuel consumed was the same uphill or downhill. To infer the emission factor for the 1983 incomplete study, for which we had concentration data for both bores 1 and 2, we averaged over the comparable 1996-2004 bore data to get Vp.

Table S6 shows the values of Vp calculated for the 6 cases. The average listed in the Table is the Monte Carlo ensemble average of (Vp(1996) + Vp(1997) + Vp(2004))/3 as computed

in each Monte Carlo realization. Similarly, for the listed standard deviation. The standard error given in the Table is simply the standard deviation divided by the square root of two.

Table S6. Inferred ventilation parameter, Vp, for the Caldecott Tunnel		
	Bore 1	Bores 1 and 2
1996	7.1	5.6
1997	4.2	4.9
2004	8.1	9.3
Average	7.2	7.4
Standard deviation	3.3	3.6
Standard error	2.3	2.6

As discussed in the main text, we do not include an error term to account for changes in fan strength and operating protocol over the years, assuming that the effect of those factors cancel. We were not able to quantify the net uncertainty in that assumed cancellation. For completeness, we list in Table S7 the inferred ventilation parameters computed for the other transversely ventilated tunnels. Note that we add another 10% into the uncertainty factor when computing emission factors using the inferred-ventilation method to account for neglecting the initial and background concentrations of BaP.

Table S7. Summary of inferred ventilation parameters[a)]	
Tunnel	Vp
Sumner	13 ± 2.4
Baltimore Harbor Tunnel (Ondov data)[b]	13 ± 6.1
Baltimore Harbor Tunnel (Benner data)[c]	5.1 ± 3.5
Lincoln/Holland combined	7.9 ± 4.5
Caldecott	7.3 ± 2.5

a) Mean and standard deviation of distributions that are approximately log normal. Data given to two significant places does not imply accuracy to two significant figures.
b) Based on lead measurements in the Tunnel taken in the early 1970s (*44*)
c) Based on BaP measurements in the Tunnel, using the 5-minute tunnel clearance time assumed by Benner based on a measurement in the early 1970s. In effect, the emission factor given for Benner's 1986 study in Table 2 of the main text represents a calculation using the average of the two parameters listed here for the Baltimore Harbor Tunnel. See discussion in text below.

To compute the uncertainty in the emission factor computed from the concentration-vs-distance data collected in the Sumner Tunnel in 1963 during two-way traffic, we needed a term to account for the uncertainty in conversion of BaP concentration from two-way traffic to one-way traffic. For that purpose, we used variances obtained from the regression discussed in an earlier section.

To compute the uncertainty in the emission factor computed from the 1975 data collected in the Baltimore Harbor Tunnel, we used the uncertainty in the computed Vp for that tunnel when comparing BaP data across years, which is dominated by the uncertainty in the 1986 emission factor assigned by researcher, Benner. For the uncertainty in the 1975 estimate made using the Pb-tracer method, we included uncertainty in the extrapolated lead concentration from 1973 to 1975, as well as uncertainty in the time of day of the 1975 measurement. To estimate the latter variance, we inspected the variation over the day for the 1973 Pb-concentration in the tunnel, assuming that the 1975-measurement was carried out between 9 am and 6 pm. In addition, we took into account the other uncertainty terms that are involved in the Pb-tracer method, as discussed in an earlier section.

To assess the uncertainty in the 1981/82 measurement in the Lincoln and Holland Tunnels, we relied on the range of BaP- and Pb-concentrations published in the paper for different passes through the tunnel, using a bootstrap calculation to obtain a standard deviation. We used a bootstrap approach to get the variance, because the data were clearly non-Gaussian, showing a bi-modal distribution, with no obvious correlation to time-of-day and therefore no obvious correlation to traffic density.

As for the 1983 data in the Caldecott Tunnel, we computed the uncertainty in the Pb-tracer method as discussed in earlier sections. As for uncertainties in Pb and BaP concentrations, we used the range of values presented in the report for different measurements taken during the study period.

To estimate the uncertainty to be applied to the 1986 emission factor, we relied, first, on the researcher's published estimate of a 50% standard deviation on his direct measurement of the emission factor. For the uncertainty in the Pb-tracer estimate, we used the Vp random variable estimated from Ondov's 1973 Pb data as listed in Table S7.

For the 1993 data collected in the Van Nuys Tunnel, as well as the 1996, 1997, and 2004 data collected in the Caldecott Tunnel, we used the authors' estimate of uncertainty in emitted BaP per kg, along with our own estimate of a 20% uncertainty in the fuel economy figure needed to convert to BaP per km.

For the Tuscarora Tunnel, we obtained raw data from the author (Gertler, A.W., personal communication, 2006) and computed a bootstrap variance, because we were informed that the data was bimodal.

For the Washburn Tunnel, we used the internal variance evident in the BaP concentration data in the published Tables, along with our own estimate of uncertainty in fuel economy needed to convert from BaP per kg to BaP per km.

Post-1995 fleet emission factors.

The post-1995 papers do not include fleet emission factors, which are needed for comparison with results from earlier studies. To obtain emission factors for the post-1995 period, we have combined the separately measured light-duty and heavy-duty emission factors, standardizing to a truck percentage of 4% in order to match the percentage most common in earlier studies. As for the fraction of trucks that were heavy-duty diesel (HDDV), if a value was not specifically given in the study itself, we have taken the value from Table S9 appropriate for the year of the study. Multiplying this fraction by 4% trucks gives us the percentage of HDDV in the fleet, p_{HDDV}. The fleet emission factor, then, is

$$E_f^{fleet} = \left(1 - p_{HDDV}\right) E_f^{LDV} + \left(p_{HDDV}\right) E_f^{HDDV} .$$

Eq. S-14

The reason there is no term in the equation for heavy-duty gasoline vehicles (HDGV) is that all of the post-1995 authors implicitly assume that the contribution from such trucks is negligible compared to the contribution from HDDV. In fact, they generally define the term HDV to only cover HDDV. To the extent that there is a contribution from HDDG, it is included in Equation S-14 on the assumption that the percentage of HDGV trucks in the fleet is strongly correlated with the percentage of HDDV trucks. In other words, the contribution from HDDG would lead to a measured value for E_f^{HDDV} obtained through regression analysis on p_{HDDV} that is higher than the true value. We discuss the evidence for a strong correlation between HDGV and HDDV in the section on Heavy-Duty Trucks

Adjusting PAH emission factors to account for differences in tunnel grade

Because the measurements in the Caldecott Tunnel were made with vehicles on a fixed grade, unlike other studies where the vehicles descended and rose at exit, it would not be unreasonable to adjust the PAH Caldecott emission factors, perhaps using the value of 2 difference in CO emission factor per liter measured between uphill and downhill in this tunnel for CO (*21*). This would lead to a 40% increase in emission factor for the 1989 study and a 40% decrease for the other Caldecott studies.

Epidemiological issues.

In this section, we discuss some issues related to using the data presented in this paper in epidemiological studies.

Tailpipe emissions are not the only source of PAH. However, industrial emissions tend to be spatially distinct from the traffic network, which may allow an epidemiological study to proceed without accounting for industrial sources, e.g, in some suburban environments. In any case, the potential influence of industrial sources can be checked either through simulations or field measurements. Emissions from home heating will be correlated with the traffic network, albeit with a different density distribution along the road network and somewhat different dispersion patterns cause by different plume-rise characteristics. There would be no intersection effect. Emissions from residential heating would not have changed as abruptly over time as did traffic emissions, because there was no comparable regulation of residential burners.

Figure 1 in the main text revealed a dramatic drop in emission factors after 1975. Thus, exposure periods prior to 1980 may be the most fruitful to study for detecting associations between BaP and health effects. Yet, exactly when the decline reached its half-way point is uncertain. The data point we have in 1975 has a large standard deviation. For those potential users of the data, such as researchers supporting epidemiologists, who might need a precise value for the half-way point, it would be wise to study the sensitivity of the results to a range of values for the 1975 emission factor.

Although standardizing emission factors to 4% trucks would not change the historical pattern of a rapid fall after 1975 as discussed in the main text, truck emissions conceivably could be important after the mid-1980s, even when they make up only 4% of the fleet. Therefore, if exposures after the mid-1980s are of interest, it might be useful in an epidemiological study to look for data on the percentage of trucks on individual roads. If trucks contribute 50% of the emission factor at a penetration of 4%, the emission factor will almost double on a road with a 12%-truck fleet. Would neglect of truck-standardization in exposure modeling lead to exposure

misclassification in such a case sufficient to compromise an epidemiological study? Not likely, given the fact that high truck percentages are likely to be found on high trafficked roads in the first place, which means they will only tend to make a high exposure higher. Also, distance to roads and distance to intersections can have a much larger impact than a factor of two. Obviously, accounting for different truck percentages on different roads is desirable for calculating exposures, but it does not appear to be an essential step.

It is straightforward to adjust post-1995 data on roads for variations in truck percentages, because measurements were made in that period separately for LDV and HDV emission factors using regression techniques. Averaged over the 4-studies that gave explicit listings of BaP emission factors in the text, the emission factor for HDDV was 16.8 μg/km. The corresponding emission factor for LDVs averaged over the same 4-studies was 2.0 μg/km. As discussed in the section, "Miscellaneous data issues," the fifth tunnel study in the 1996-2004 period provided sufficient information in the text to allow extraction of the BaP emission factors. However, the results were somewhat problematic in that the intercept in the regression gave a negative emission factor for LDVs. Forcing the intercept to zero and including the results of this study in the average gave a 5-study average for E_f^{HDDV} of 26 μg/km and a 5-study average value for E_f^{LDV} of 1.6 μg/km. Additional details are given in the section on Heavy Duty Trucks.

Researchers in this period did not provide emission-factor estimates for heavy-duty gasoline vehicles (HDGV), presumably because HDGV emissions of most PAHs are negligible compared to HDDV. Unfortunately, this is not the case for the heavier PAHs, such as BaP. The question then arises, "Are the HDGV emissions counted in the assignment to LDV emission factors or in the assignment to HDDV emission factors?" The latter is more likely, given the high correlation found between HDDV and other trucks in the studies (see section on Heavy-Duty Trucks). Thus, it is appropriate to avoid explicit consideration of HDGV emissions, because they are already included with the heavy-duty diesel vehicles (HDDV). To include HDGV emissions

implicitly, when using the 16.8 μg/km figure, it is necessary to use the fraction of HDDV on the road, not the fraction of HDV. Tables S8 and S9 allow computation of the relative percentages of different types of trucks on the road at different study years. Once, a value for the percentage of HDDV in the fleet, p_{HDDV}, is obtained, the fleet emission factor in the post-1995 period, with the Washburn Tunnel results excluded from the average, is:

$$E_f^{fleet} = \left(1 - p_{HDDV}\right) 2.0 + \left(p_{HDDV}\right) 16.8 \cdot \qquad \text{Eq. S-15}$$

and, when the Washburn Tunnel results are included:

$$E_f^{fleet} = \left(1 - p_{HDDV}\right) 1.6 + \left(p_{HDDV}\right) 26 \cdot \qquad \text{Eq. S-16}$$

For a typical HDDV percentage of 2.75%, the fraction of emissions attributed to trucks is 19%, when the Washburn results are excluded, and 31%, when the Washburn results are included.

For years 1975 and earlier, it is probably safe to ignore correcting emission factors for varying truck percentages. For the period between 1975 and 1996, the situation is more complicated. One option would be to use information from chassis dynamometer data, such as that collected by Bailey et al. (*89*). These authors found a log-linear relationship between particulate emission factor and year, with a net decrease of 87% from 1950 to 1990. Although the information given in that paper as a function of fleet year is for HDDVs, it could be taken as a surrogate for HDV emissions, allowing back-calculation from the values obtained for the post-1995 emission factor. It would imply an emission factor of 130 μg/km for HDV in 1950.

To see the implication of going from a measured emission factor in a tunnel with a truck fraction, q, say 4%, to the emission factor corresponding to a truck fraction, p, which might be

used on a particular road segment, the values for the inferred HDV emission factor can be substituted in the following equation.

$$E_f^{fleet}(new) = \frac{\left(1-p\right) E_f^{fleet}(measured) + (p-q)\, E_f^{HDV}}{1-q},$$

Eq. S-17

BaP is not the only carcinogenic PAH, but it can serve as a surrogate for other PAHs. For instance, Schneider (*90*) found that the ratio of BaP to total PAH remained roughly constant over time and space for the inhalation pathway. Menichini (*64*) found that the average yearly ratio of atmospheric PAH to BaP was stable from 1993-1997 in a medium traffic site in Rome. The ratio was within a factor of $\pm$ 2 of the average going back to 1977. These yearly averages were found to be insensitive to the short-term (month-to-month) fluctuation Thus, BaP can be considered as a surrogate for all PAHs.

The curves shown in this paper for PAHs other than BaP support the idea of using BaP as a surrogate for other PAHs, given the fact that their historical decline in emission factors seems similar to those for BaP. One caveat: fluoranthene and possibly other light PAHs that are more likely to be emitted by diesels than gasoline vehicles may behave differently than BaP depending on the percentage of diesels in the fleet. Metabolites of fluroanthene have been found to be a mammary carcinogen in rats (*91*). Even though the toxic equivalency factor may be small on a mass basis for these light PAHs, the product of mass and TEQ might make them important compared to the heavier PAHs like BaP. Therefore, it might be important to pay attention in epidemiologic studies of PAH to the changes in diesel percentages from road to road. However, we have not found it an easy matter to obtain historical information on the percent of diesels on roads.

Because cancer risk estimates exist for PAH (*7*), the data presented in the Figures and in the Tables presented in the main text and the Supporting Information could be used as part of a calculation of the reduction in excess mortality that was gained from the introduction of catalytic converters. However, the full gain cannot be computed, until accounting is made for the reduction in PAH emitted in other parts of the driving cycle, namely cold starts and stop and go driving. We leave quantification of the historical decline in emissions from these other parts of the driving cycle to a subsequent paper, which will summarize historical measurements of BaP emission on chassis dynamometers programmed according to standard urban driving cycles.

Comparison of US results to measurements in other countries.

The historical tunnel data shown in Figure 1 cannot be compared directly to tunnel measurements made in other countries, because the US was the first to put control measures on its passenger vehicle fleet. Instead, comparison must be made to different time periods, allowing sufficient time for the fleets to become comparable. For instance, the BaP emission rate was found to be 16 μg/km in tunnel measurements carried out in 1991 in Belgium (*92*), but that was only two years after catalytic converters were introduced there, when the percentage of three way catalysts was only 3%. A comparable period in US history would be the early 1980s. Figure 1 in the main text indicates a BaP emission rate of 8-15 μg/km in the US at that time, which is reasonably consistent with the Belgium result, given the uncertainties in the comparison.

A measurement in a Swedish tunnel (''Soderledstunnel'') obtained between Dec 1998 and February 1999 gave an emission factor of 4.6 μg/km (*93*), about a factor of three lower than the 1991 measurement in Belgium. Sixty percent of the light-duty vehicles (LDVs) had catalytic converters by this time, which is the same penetration reached in the US in the early 1980s, when the US BaP fleet emissions were measured to be 7.6 μgm/km. Given the fact that the LDVs without catalytic converters in Sweden in 1998 were probably much better controlled than comparable vehicles in the US in 1981, the 65% percent difference in emission factors is not out of line. In another tunnel measurement in Sweden a few years later in the Gothenburg tunnel (*94*), the fleet emission factor continued to fall, now reaching a value (0.57 ± 0.4). The percentage of LDVs at the time with catalytic converters in the Gothenburg Tunnel is not known, but the value presumably rose above the 60% measured a few years earlier. The range of 0.57 ± 0.4 μg/km overlaps with the range of emission factors measured in California in 1989, which was 0.95 ± 0.45 μg/km, at a time when the penetration of LDVs with catalytic converters was 80%, probably not too different from the value in the Gothenburg Tunnel. Based on these comparisons, we conclude that, within a factor of two, there is consistency between the European

and US measurements at a comparable stage of automotive pollution control. It is of interest that the Gothenburg-Tunnel study found no correlation between HDV percentage and the emission factor for carcinogenic PAHs, a result that is consistent with two of the five relevant studies carried out in the US (*16, 18*). The use of these regression techniques produces unstable results from study to study, which argues for averaging over many studies, as we have done in this work.

In 1958, before the introduction of pollution controls, Waller and colleagues measured BaP and Pb concentrations in the Blackwell Tunnel in the United Kingdom on a number on occasions during a 4-day period, along with the percentage of truck traffic present during each measurement (*95*). We have used the ratio of BaP concentrations to Pb concentrations to estimate emission factors for BaP. By regressing to the percentage of heavy-duty vehicles we were able to obtain estimates of emission factors for both light duty and heavy-duty traffic. As for the amount of lead in gasoline in the UK, the value in the UK in the late 1950's is thought to be between 0.6 and 0.8 g/liter (personal Communication, J.M. Pacyna, Norwegian Institute for Air Research, 2006.) We have taken a mid-point value of 0.7 g/liter. Following Waller et al. we assumed that 50% of the trucks were diesel and proceeded to calculate emission factors using the Pb-tracer method, as we did for US tunnels.

Emissions in the British Blackwell Tunnel should be high, because of the nature of the tunnel. "There are corners and gradients to be negotiated in each tunnel and driving conditions are similar to those in highly congested streets." (*95*). Average speeds were very low, ranging between 3 and 7 km/hour. As a result the calculated emission factors are of limited relevance to the US tunnel studies. However, they do serve to give some idea of how emission factors change under congested driving conditions, bearing in mind that the UK fleet may have differed from the US fleet. Assuming that gasoline trucks emit the same amount of lead per kg of fuel consumed (but not per km), gasoline-consuming trucks emit twice as much lead as passenger vehicles per km, based on the ratio of US fuel efficiencies at the time. Thus, the emission factors for automobiles in the Blackwell Tunnel determined from the regressions was 93 μg/km, which is

about 3 times higher than we find for the Sumner Tunnel in 1961. The ratio for BaP truck emissions is 800 μg/km, which is more than a factor of ten higher than an estimate for the Sumner Tunnel in 1961 made by scaling the fleet emission factor by the ratio of fuel efficiencies of LDVs and trucks. Possibly, emission rates increase faster for trucks under congested driving conditions than they do for passenger vehicles. In any case, the finding of relatively high BaP emissions under congested driving conditions is consistent with our own fits to BaP soil measurements near traffic intersections (*69*) and our number for emission increases at intersections given in the main text.

Heavy-duty truck data and disaggregation of emission factors by vehicle type.

Measurements of diesel emissions made on chassis dynamometers have indicated that, while emissions of the lighter PAHs are dominated by diesel engines, diesels have not historically been the major source in the US of the heavier PAHs, such as BaP (*7, 56, 68, 96-98*), which are the most carcinogenic on a mass basis. Nevertheless, even for the heavier PAHs, it may be useful to consider the truck contribution in standardizing emissions in epidemiological studies. In this section we consider heavy-duty truck percentages for the various study fleets, along with a discussion of those tunnel studies in recent years that have used regression techniques to separate out the heavy-duty vehicle (HDV) component of emissions from the light-duty vehicle (LDV) components.

Table S8. Fractions of traffic fleet by vehicle category

Year/ Tunnel/ City	Fraction of LDV vehicles in fleet (g_1)	Fraction of HDV vehicles in fleet [a] (g_2)	Fraction of LDVs without catalysts[b] (k_1)	Fraction of LDVs with catalysts[b] (k_2)
1961 (Sumner, Boston)	0.964	0.036 [c]	1	0
1963 (Sumner, Boston)	0.964	0.036 [c]	1	0
1975 (Baltimore Harbor Tunnel)	0.88	0.12[d]	0.965	0.035
1981.5 (Lincoln/Holland, NYC)	0.88	0.12 (*99*)	0.55	0.45
1983 (Caldecott, San Francisco area)	0.955	0.045	0.45	0.55
1986 (Baltimore Harbor Tunnel)	0.89	0.11	0.30	0.70
1989(Caldecott, San Francisco area)	0.96	0.04 [e]	0.15	0.85
1993 (Van Nuys, Los Angeles)	0.96	0.04	0.076	0.924
1996(Caldecott, San Francisco area)	0.97	0.03	0.052	0.948
1997(Caldecott, San Francisco area)	0.97	0.03	0.044	0.956
1999 (Tuscarora, Pennsylvania)	0.57	0.43	0.028	0.972
2000 (Washburn, Houston)	0.97	0.03	0.02	0.98
2004(Caldecott, San Francisco area)	0.974	0.026	0.01	0.99

a) Taken from the original study, unless otherwise indicated.
b) For the fraction of vehicles with and without catalytic converters, we have relied on data for Los Angeles, except for the Caldecott Tunnel, where data from San Francisco were used (*73*), (*100*), (*101*).
c) Based on 1977 total truck counts (*102*). Note that the Sumner Tunnel has a height restriction that keeps out 3-axle trucks.
d) Lisa Grant, Maryland Department of Transportation, personal communication re 1975 truck percentage, 2007.
e) Interpolated between the values for the 1983 and 1996 studies.

The makeup of the truck fleet can be described in terms of coefficients, f_i, that delineate the fractional share of the different truck types in each year. To obtain the weights, f_i, we relied on historical data from the California Energy Commission's emission model (*73, 100, 101*), including an estimate of the fraction of medium and heavy-duty (MHDT and HHDT) gasoline trucks that had catalytic converters. The numerical values of the f_i are given in Table S9. We found no comparable historical data for the East Coast, so used California data. Of the California data, truck traffic though (urban) Los Angeles was chosen as the most likely best match to truck traffic traveling through the Baltimore Harbor, Lincoln, and Holland tunnels.

Table S9. Fractional share of truck categories over time.[a)]

Year/ Tunnel/ City	Fraction of HDDV (f_1)	Fraction of $HDGV_{[no\ cat]}$ (f_2)	Fraction of $HDGV_{[cat]}$ (f_3)	
1961 (Sumner, Boston)	0.32	0.68	0	
1963 (Sumner, Boston)	0.32	0.68	0	
1975 (Baltimore Harbor Tunnel)	0.32	0.68	0	
1981.5 (Lincoln/Holland, NYC)	0.48	0.45	0.075	
1983 (Caldecott, San Francisco area)	0.3	0.6	0.1	
1986 (Baltimore Harbor Tunnel)	0.52	0.34	0.14	
1989(Caldecott, San Francisco area)	0.51	0.29	0.2	
1993 (Van Nuys, Los Angeles)	0.65	0.16	0.19	
1996(Caldecott, San Francisco area)	0.69	0.13	0.18	
1997(Caldecott, San Francisco	0.71	0.11	0.18	

area)				
1999 (Tuscarora, Pennsylvania)	0.74	0.075	0.19	
2000 (Washburn, Houston)	0.75	0.06	0.19	
2004(Caldecott, San Francisco area)	0.81	0.03	0.16	

a) For the fraction of vehicles with and without catalytic converters, we have relied on data for Los Angeles, except for the Caldecott Tunnel, where data from San Francisco were used (*73, 100, 101*).

b) Computed from Tables in Davis and Diegel (*27*), assuming that gasoline trucks fall into their category of heavy-duty, single-unit trucks.

Heavy-duty diesel emission factor

In this section we add more detail to our discussion of issues surrounding the assignment of an emission factor to heavy-duty diesels. As stated in earlier sections, there were 5 studies carried out from 1996-2004 in which the authors used regression techniques to separate out HDV emission factors from the fleet value measured at different times of the day or in different tunnel bores, when different percentages of HDVs were present. The researchers defined HDV to include only HDDV, in effect, assuming that the contribution from HDGV was small compared to the contribution from HDDV. Four of the studies presented results for BaP. The researchers implicitly make the assumption that neither the composition of the light-duty fleet nor the behavior of LDV drivers changes in a correlated way with the percentage of diesel vehicles. In other words, they assume there is no confounding. Nevertheless, we take the average of these tunnel values as the best estimate available of heavy-duty diesel truck emissions for the period 1995 to 2005. To compute the average of the four studies, we assigned zero to the heavy-duty diesel (HDDV) emission factor in the Caldecott Tunnel in 1996, based on the statement in the paper that the HDDV component was negligible. The resulting average value of the HD-slopes is

48 plus or minus 28 μg/kg per heavy-duty diesel truck, which is equivalent to 16.8 μg/km per heavy-duty diesel truck, when account is taken of truck fuel economy (*27*).

As a sensitivity test we consider other possibilities. First, Miguel et al. found more than a two-fold higher BaP concentrations in the low-truck bore (*16*), which meant that the concentration was higher in the low-truck bore, even when normalized to the traffic flow of the bore in which HDDV are allowed, suggesting the possibility that the HDDV slope might have been negative. We note that a measurement in a Swedish Tunnel in April of 2000 also found a negative slope with diesel percentage and large PAHs (*94*). Should the slope in fact have been negative, a case can be made that the negative value should be included in the 4-study average, thereby, reducing the average emission factor. On the other hand, it is also possible to work with the data given in Chellam et al. (*19*) and compute a HDDV slope. If we do this, forcing the intercept to zero, we obtain an emission factor higher than any of the other studies, namely 63 μg/km per HDDV vehicle. Adding this value into the 4-study average raises the average HDDV emission factor from 16.8 to 26 μg/km.

To the extent that heavy-duty gasoline vehicles make a significant contribution to the truck emissions for an individual PAH compared to HDDV, the HDGV contribution is implicitly included to some extent in the emission factor determined by the researchers, because of the method chosen for analysis. The researchers performed their regressions by choosing the number of HD-diesels as the independent variable, not the total number of trucks. Yet, the number of gasoline trucks is likely to be correlated with the number of diesels. For instance, based on data given in the supporting information for the 2004 study in the Caldecott Tunnel, the correlation coefficient between diesels and non-diesels was 0.77 in the bore carrying heavy-duty vehicles, while the corresponding correlation coefficient between diesels and all trucks was 0.98 (*103*). Furthermore, when the correlations were made using data for both bores, including the bore with limited truck traffic, which is the most appropriate approach to take when the goal is to interpret

regression outputs, both correlation coefficients turned out to be greater than 0.96. Therefore, BaP emissions from gasoline trucks are likely to be included in the 16.8 μg/km assigned to a diesel truck, as discussed in earlier sections. The same is true for medium-duty diesels.

Diesel passenger cars, as opposed to trucks, represent less than 1% of the in-use fleet (*104*). Therefore, we include them in the light-duty emissions category. Thus, in our results, LDV emission factors implicitly include passenger car diesel emissions.

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