

Size Distributions of Polycyclic Aromatic Hydrocarbons and Elemental Carbon. 1. Sampling, Measurement Methods, and Source Characterization

Chandra Venkataraman,[†] James M. Lyons,[‡] and Sheldon K. Friedlander*

Chemical Engineering Department, University of California, Los Angeles, California 90024

Ten polycyclic aromatic hydrocarbons (PAHs) were measured in size-segregated aerosol samples of 0.05–4 μm diameter collected by a low-pressure impactor sampler using a high-resolution analysis procedure based on HPLC and fluorescence detection. This method can be used to characterize both vehicular emissions and ambient aerosol, allowing meaningful intercomparison. A field study was carried out at two traffic tunnels to characterize the vehicular source. PAH and elemental carbon (EC) size distributions measured in the Caldecott Tunnel were unimodal with over 85% of the mass in particles smaller than 0.12 μm aerodynamic diameter. Ambient air intrusion into the relatively short Sepulveda Tunnel resulted in small secondary peaks in the PAH distributions in the 0.5–1.0- μm size range. These size distributions, when corrected for ambient air intrusion using a mass balance of PAH and CO, were unimodal. PAH profiles (PAH/Benzo[*k*]fluoranthene ratios) were developed for the vehicular source, which can be used in receptor modeling estimations for source allocation.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are products of incomplete combustion and, in urban and industrial atmospheres, are almost entirely anthropogenic in origin. Polycyclic organic matter (POM), which includes higher molecular weight PAHs, has been identified as a hazardous air pollutant in Title III of the Clean Air Act Amendments of 1990. Motor vehicles are thought to be a major source of atmospheric PAH (1). With the phase-out of organo-lead octane enhancers, lead is no longer a reliable tracer for automobile emissions, and PAHs have been suggested as alternative particulate tracers (2–4). Daisey et al. (2) suggested cyclopenta[*c,d*]pyrene and benzo[*b*]naphtho[2,1-*d*]thiophene as almost unique tracers for gasoline and diesel emissions, respectively.

While individual tracer PAH compounds for specific sources are not easy to identify, the potential use of PAH profiles to distinguish different combustion sources was suggested several decades ago (5). Characteristic ratios of PAH and PAH profiles have been used in qualitative and quantitative source apportionment estimations (3, 6, 7). Limitations to this method, when applied to vehicular emissions, arise from two main reasons: inherent variations in emission patterns which could result from variations in vehicle mix, driving mode, and altitude (8) and inaccuracies present in sampling and measurement techniques.

We wished to develop a sampling and measurement method for analysis of PAH and elemental carbon (EC) which could be used to characterize both source and ambient fine aerosol to facilitate meaningful intercom-

parison. Aerosol mass distributions measured in emissions from combustion sources show peaks at particle diameters of about 0.1 μm (9). A size distribution of elemental carbon in fresh vehicular emissions, which contributes about 35% of the fine mass (10), showed that over 85% of the mass was present in particles smaller than 0.5- μm diameter (11). It was therefore of interest to collect size-segregated samples of submicron aerosol particles. The Hering low-pressure impactor (12, 13), used in this study, segregates particles of 0.05–4.0 μm aerodynamic diameter into eight size cuts.

An additional advantage of impactor sampling is the minimization of losses that occur during high-volume sampling of semivolatile species on fiber filters (14). High-volume sampling on fiber filters, the most widely used particle sampling method, is prone to sampling losses (15, 16). "Blow-off" of volatile species and filter-catalyzed oxidation interfere with accurate estimations of particulate organic compounds (17).

Objectives of the study included the following: (i) Developing a measurement technique combining impactor sampling and a high-resolution analysis method based on HPLC and fluorescence detection to analyze particulate PAH. (ii) Measuring PAH and EC size distributions in fine aerosol emissions from the vehicular source using the sampling and analysis method applicable to ambient aerosol. (iii) Developing a characteristic source profile for the Los Angeles vehicle fleet.

The PAH species selected for analysis included the 5-ring and larger species (mol wt 252–278) that were proposed as potential particulate tracers for vehicular emissions (3) and 2–4-ring species (mol wt 202–228) that have been observed in automobile exhaust (6). These species, in addition to elemental carbon and CO, were measured as they originate primarily from the automobile source. The selected PAHs are among the 16 species identified by the U.S. Environmental Protection Agency as priority pollutants in the Clean Water Act because of their carcinogenicity. Species of molecular weight lower than fluoranthene (202 amu) were not analyzed in the particulate samples as they are present mainly in the gas phase under ambient atmospheric conditions.

Experimental Methods

A field study was conducted to collect size-fractionated aerosol from two traffic tunnels, Sepulveda Tunnel in Los Angeles (length, 0.5 km) and Caldecott Tunnel near Berkeley (length, 1.1 km). Aerosol samples were collected on April 22–24, 1989, in Sepulveda Tunnel and on August 9, 1989, at Caldecott Tunnel to coincide with an ambient field study conducted in 1989 (18). The samples were analyzed for elemental carbon and 10 PAHs as described in the following sections.

Sampling Instruments and Methods. Particles were collected using a Hering low-pressure impactor (LPI) (12, 13) connected to a CRIVAC S4A vacuum pump (Leybold

[†] Present address: CE-CERT, University of California, Riverside, CA 92521-0425.

[‡] Present address: Sierra Research, Inc., Sacramento, CA 95814.

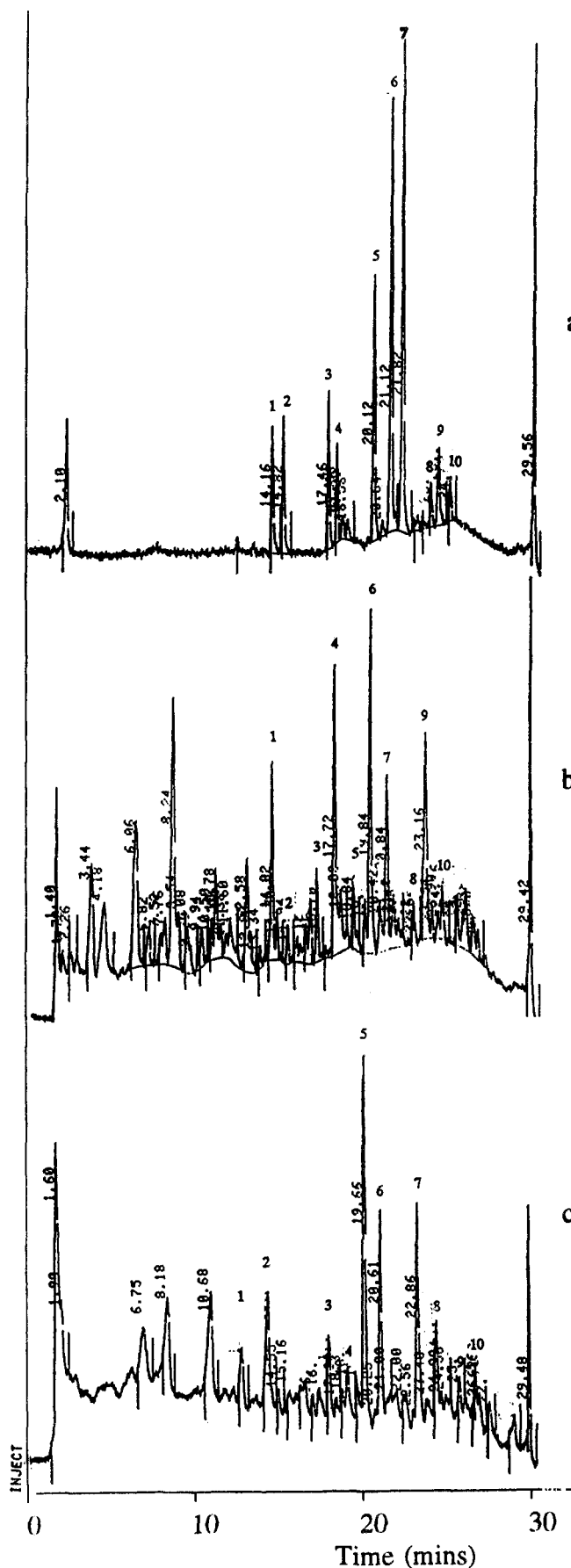


Figure 1. Typical chromatograms obtained for the NBS standard reference material (SRM 1647a) (a), tunnel aerosol sample (b), and ambient aerosol sample (c). Peak identification is as follows: (1) fluoranthene, FLT; (2) pyrene, PYR; (3) benz[a]anthracene, BAA; (4) chrysene, CHR; (5) benzo[b]fluoranthene, BBF; (6) benzo[k]fluoranthene, BKF; (7) benzo[a]pyrene, BAP; (8) dibenzanthracene, DBA; (9) benzo[ghi]perylene, BGP; and (10) indeno[1,2,3-cd]pyrene, INP.

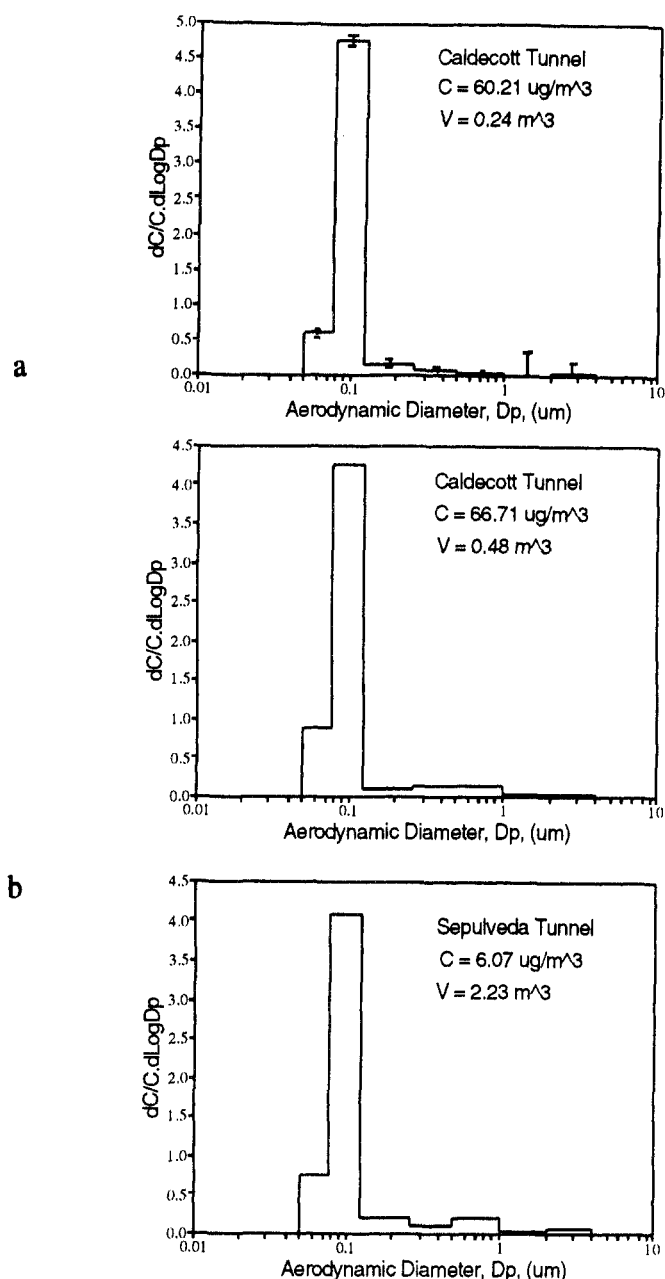


Figure 2. Size distributions of elemental carbon (EC) measured at the Caldecott and Sepulveda Tunnels. The strong unimodal character indicates that over 90% of the EC mass in fresh vehicular emissions is in particles smaller than about 0.1- μ m diameter.

Heraeus). The LPI has eight stages with 50% efficiency cut points at aerodynamic diameters of 4.0, 2.0, 1.0, 0.50, 0.26, 0.12, 0.075, and 0.05 μ m, for stages 1–8, respectively. Glass disks of 25-mm diameter were used as the impaction surfaces. A total of 0.5 μ L of 2% vaseline in cyclohexane solution was applied at the center of each disk. Turner and Hering (19) showed that this surface collects polystyrene latex (PSL) particles with about 90% collection efficiency. In use with atmospheric aerosol, which is sooty and contains liquid-phase aqueous and organic components, higher collection efficiencies would be expected than those obtained for PSL particles.

The adsorption artifact observed during the sampling of semivolatile organic species on quartz fiber filters would not be of importance with the Hering impactor as it does not use a filter substrate for particle collection. Estimates of volatilization losses from the low-pressure stages of the impactor (18) show that species with vapor pressures less than 10^{-9} atm under ambient conditions would be collected

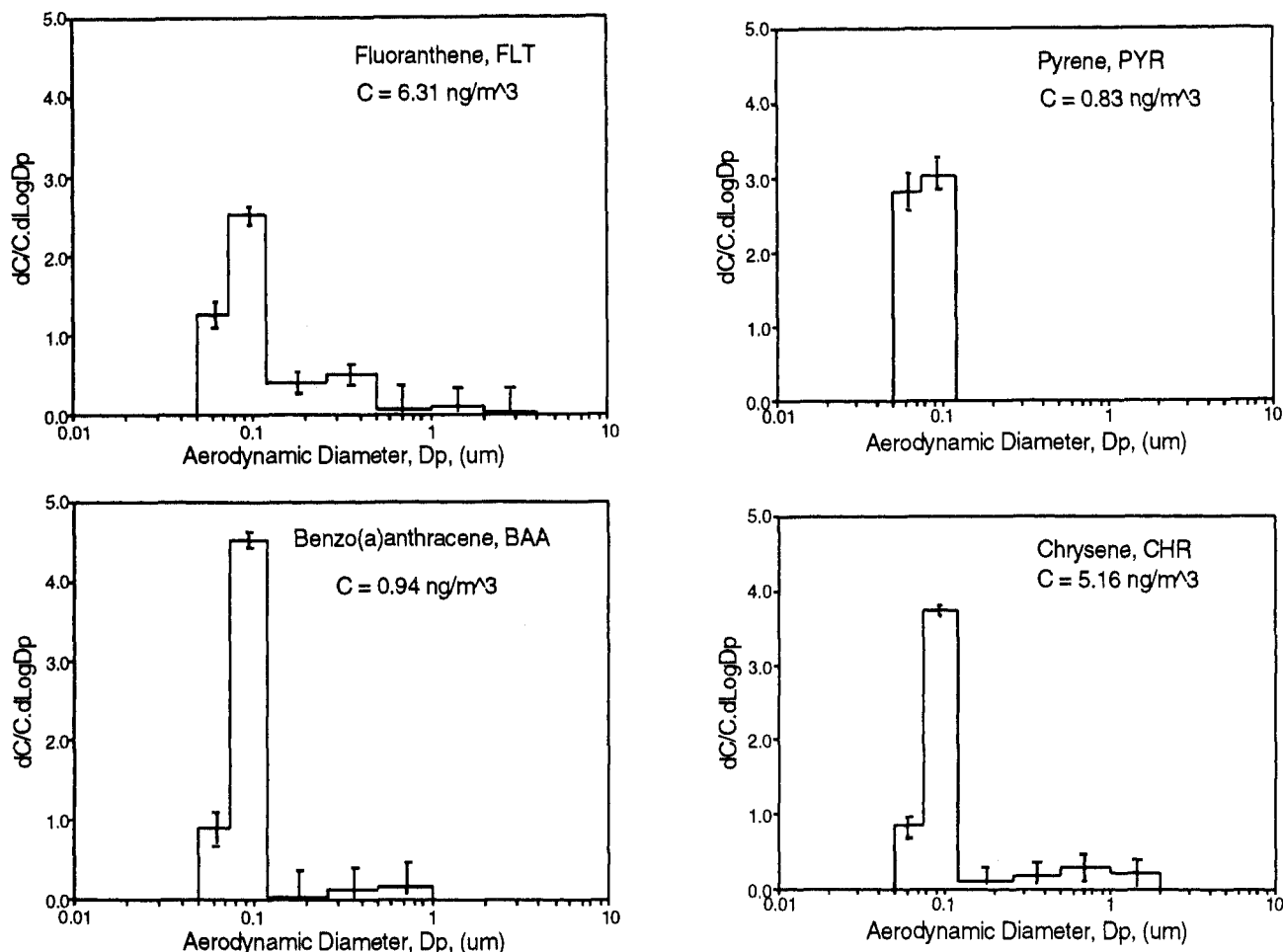


Figure 3. Size distributions of 4-ring PAHs (molecular weight, 202–228) measured at the Caldecott Tunnel. The unimodal character, similar to the EC size distributions, is consistent with the simultaneous formation of both pollutants during combustion.

with 98% or greater efficiency on all impactor stages. The pure component vapor pressures of the PAH analyzed in this study range from 10^{-9} to 10^{-13} atm at 298 K, and we believe that losses of these species during sampling would not be significant.

Samples were collected for elemental carbon analysis on a stainless steel strip (4 mm \times 25 mm) fixed along a diameter of the glass impaction disk. Particles were deposited in a spot of vaseline at the center of the strip applied as described above.

Carbon monoxide concentrations were measured using a CO monitor (Ecolyzer, Model 2106, Energetics Science, Inc.) and averaged over the entire sampling period.

Sample Extraction. The particle-loaded impactor disks were placed in 25-mL beakers, covered with about 5 mL of HPLC-grade methylene chloride (Aldrich), and extracted for 30 min in an ultrasonic bath (Bransonic Inc., Model 2200). Ultrasonication has been reported to give higher recoveries of PAH than Soxhlet extraction (20, 21) and has the added advantages of being rapid and using only small amounts of solvents. Extracts were filtered through a 5- μ m PTFE membrane MILLEX filter unit (Millipore Corp.) and evaporated to dryness under a gentle stream of nitrogen. A laminar flow of nitrogen was maintained to reduce sample loss through surface disturbances. The evaporated extract was taken up in 100 μ L of acetonitrile (Aldrich).

Sample Fractionation and PAH Quantification. Ten PAHs were analyzed using a Spectra Physics HPLC (Model SP 8700 XR) equipped with a Rheodyne injector (10, 20, or 50 μ L loop; Model 8750). A Supelcosil LC-PAH

C18, 5 μ m, 15 cm \times 4.6 mm i.d. analytical column (Supelco, Inc.) was used at room temperature. The analysis procedure was based on that used by Miguel and de Andrade (22) to analyze PAH in TSP samples collected on quartz fiber filters. HPLC-grade solvents were sparged with helium gas during the analysis and for 1 h preceding it to effectively remove dissolved gases. We used a solvent gradient of acetonitrile and water (40–100% acetonitrile in 20 min, held at 100% acetonitrile for 5 min) at a flow rate of 0.9 mL min $^{-1}$. Average retention times, in minutes, for the 10 PAHs analyzed were as follows: fluoranthene (FLT), 14.1; pyrene (PYR), 14.8; benz[a]anthracene (BAA), 17.5; chrysene (CHR), 17.8; benzo[b]fluoranthene (BBF), 20.1; benzo[k]fluoranthene (BKF), 21.1; benzo[a]pyrene (BAP), 21.8; dibenz[a,h]anthracene (DBA), 23.5; benzo[ghi]perylene (BGP), 23.9; and indeno[1,2,3-cd]pyrene (INP), 24.7. Elution times of PAH in the standard mixture did vary up to 10% between different days, based on the HPLC column used and the temperature in the laboratory, but there was very good agreement between standard and sample chromatograms obtained on a given day. A fluorescence detector (Spectra Physics, Model FS 970) set at 265 and 389 nm wavelengths, respectively, was used for excitation and emission. Fluorescence sensitivity was between 0.5 and 0.02 μ A.

The HPLC system was calibrated using an external standard. An NBS Standard Reference Material (No. 1647a, priority pollutant PAH mixture) was used at dilutions of 200, 400, 625, and 1000 to obtain calibration curves for each run. Linear fluorescence response was obtained for all PAH over the range of dilutions indicated.

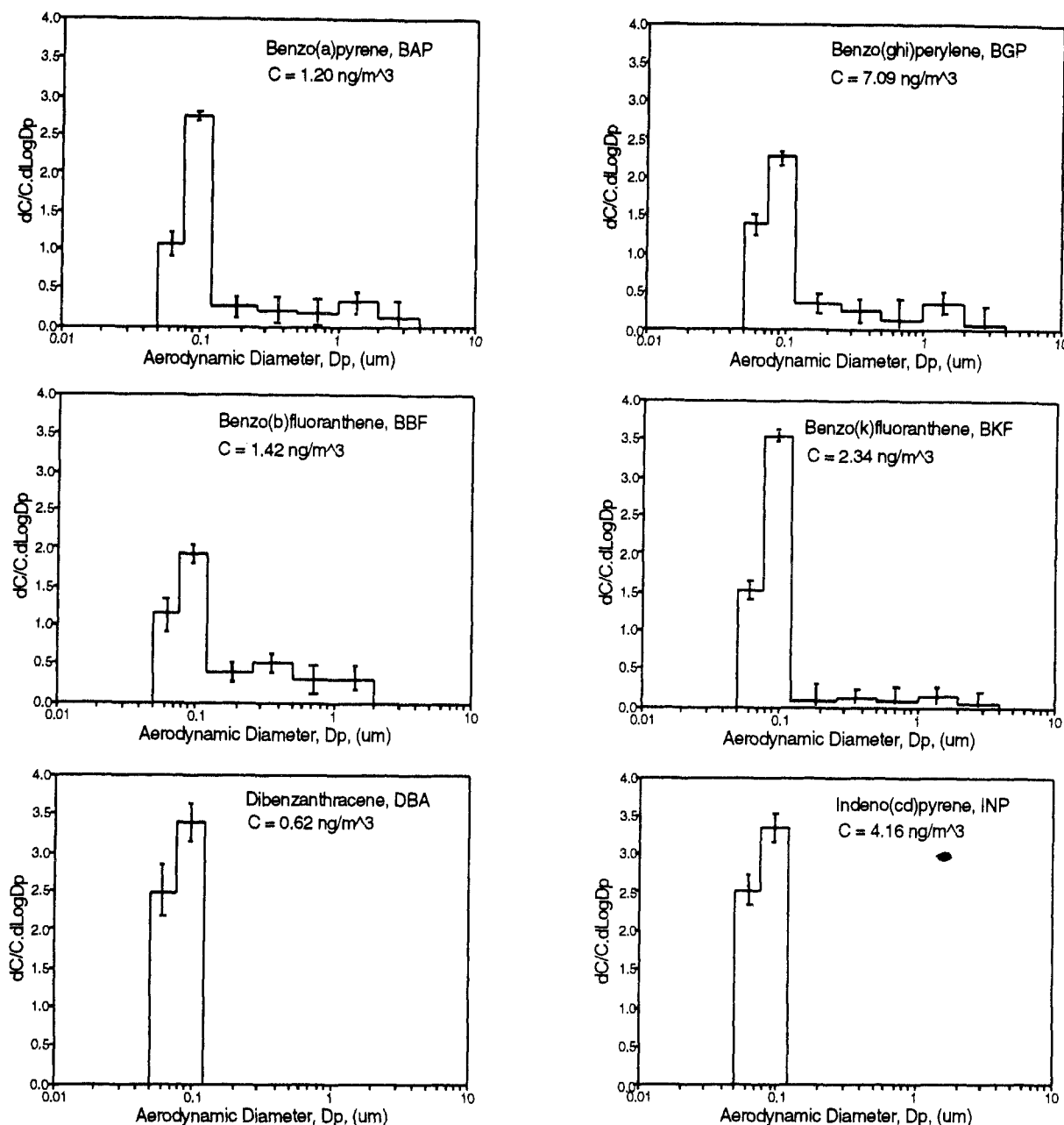


Figure 4. Size distributions of 5-ring and larger PAHs (molecular weight, 252–278) measured at the Caldecott Tunnel. The unimodal character, similar to EC size distributions, is consistent with their simultaneous formation during combustion.

Prior to actual analysis, three injections of the liquid standard were made to ensure stabilization of the column. After column conditioning, chromatograms were very reproducible. Detection limits obtained for the different PAHs were in the 2–40 pg range. Typical chromatograms obtained for the NBS standard and from the tunnel and ambient aerosol are shown in Figure 1. Collection disks with the vaseline substrate, for use as blanks for the PAH analysis, were prepared in a manner identical to those used in the impactors. The vaseline substrate did not interfere with the PAH analysis, and consistently low amounts of PAH were measured in blank samples.

Elemental Carbon Analysis. Elemental carbon was analyzed by AtmAA, Inc. (Chatsworth, CA) using a method developed by Fung (23). As the samples in this study were collected in a spot of vaseline, it was not possible to measure organic carbon. A Dohrman carbon analyzer, DC-52, was used for the analysis. The sample was placed in a platinum boat in contact with MnO_2 . Oxidation of the volatile carbon occurred at temperatures of 120–525 °C (not measured quantitatively here), and elemental

carbon was oxidized at 850 °C. The carbon dioxide resulting from the oxidation was reduced to methane and quantified by a flame ionization detector. The vaseline substrate did not interfere with the EC measurements. Vaseline blanks gave consistently low EC readings. Detection limits of 1 ng were obtained.

Results and Discussion

Elemental Carbon in Tunnel Aerosol. Elemental carbon (EC) constitutes about 35% of the fine particle mass (<1.3- μm diameter) in fresh vehicular emissions based on tunnel measurements (10). Pollutant concentrations in the tunnel reflect a dynamic balance between emission rates and removal rates. Table 1 shows the average EC and CO concentrations in both tunnels. The Sepulveda Tunnel, being short, has greater intrusion of outside air and shows lower concentrations of both pollutants.

The CO:EC ratio is significantly different in the two tunnels. The low CO:EC ratio in the Caldecott Tunnel

Table 1. Concentrations of Elemental Carbon and Carbon Monoxide at Caldecott and Sepulveda Tunnels

site/source	EC ($\mu\text{g m}^{-3}$)	CO (ppm)	CO/EC ($\mu\text{g m}^{-3}/(\mu\text{g m}^{-3})$)
Caldecott	63.5	26.7	541
Sepulveda	6.1	10.6	2462
gasoline vehicles ^a			2516
diesel vehicles ^a			7

^a A comparison of catalyst-equipped gasoline vehicles and diesel vehicles, based on dynamometer testing (24).

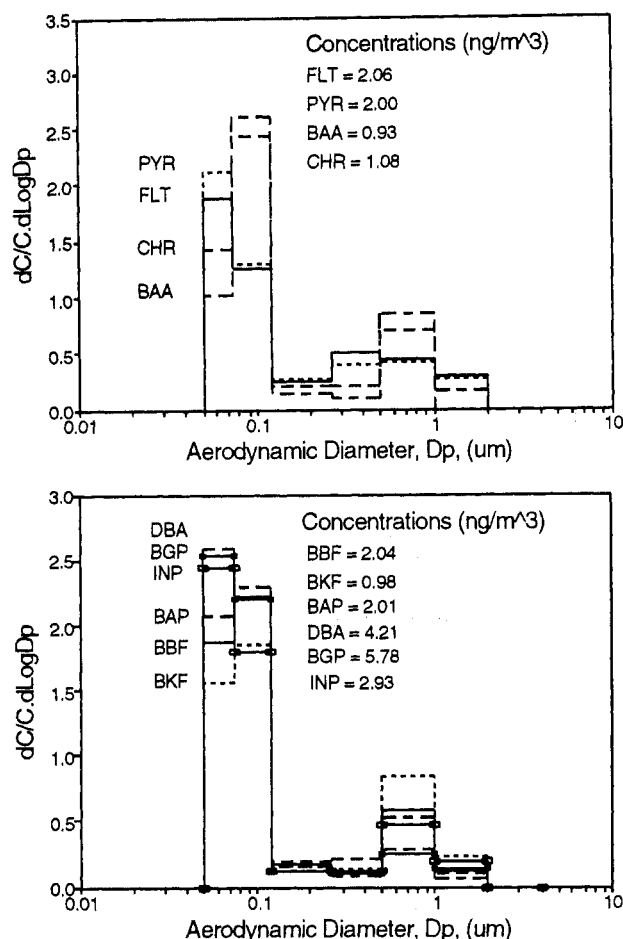


Figure 5. Size distributions of PAHs measured at the Sepulveda Tunnel. The mode in the 0.5–1- μm size range occurs because ambient air incursion into this short tunnel (length, 0.5 km) brings in aged, polydisperse ambient aerosol.

could result from a greater fraction of diesel vehicles, as this tunnel is on a freeway route. The Sepulveda Tunnel, in contrast, is on a surface street where the fleet is dominated by light-duty gasoline-powered vehicles. Dynamometer studies of the profile of pollutants from diesel and gasoline vehicles show that CO is emitted primarily from gasoline-powered vehicles while the diesel profile is dominated by black or elemental carbon (24).

The elemental carbon size distributions in both tunnels (Figure 2) are unimodal with over 80% of the EC mass in particles smaller than 0.12- μm diameter. The error bars shown in the first distribution (measured at Caldecott Tunnel) reflect detection limits of the chemical analysis and are representative of uncertainties in the other EC size distributions as well. Total EC concentrations in particles of 0.05–4- μm size and the volume of air sampled are also shown. These data are consistent with a unimodal EC distribution reported previously from Caldecott Tunnel by Miguel and Friedlander (11) as well as with more recent

Table 2. Mass Median Diameters of Elemental Carbon and PAH Species Measured at the Caldecott and Sepulveda Tunnels (μm)

species	Caldecott Tunnel	Sepulveda Tunnel
EC	0.11	0.15
FLT	0.16	0.10
PYR	0.06	0.09
BAA	0.10	0.08
CHR	0.17	0.08
BBF	0.26	0.07
BKF	0.12	0.08
BAP	0.20	0.07
DBA	0.06	0.08
BGP	0.21	0.07
INP	0.13	0.07

measurements of aerosol mass distributions in emissions from combustion sources (9).

PAH Size Distributions in Traffic Tunnels. PAH size distributions in the aerosol collected in Caldecott Tunnel are unimodal as shown in Figures 3 and 4. Again, error bars reflect detection limits of the HPLC analysis. In the Caldecott Tunnel aerosol, over 80% of the PAH mass was in particles smaller than 0.12 μm aerodynamic diameter. The distributions measured in the Sepulveda Tunnel (Figure 5) showed a second mode in the 0.5–1- μm size range, which contains 10–15% of the PAH mass. This tunnel is only 0.5 km long, and we expect some ambient air intrusion to bring in aged, polydisperse, ambient aerosol.

In order to correct tunnel PAH concentrations for ambient air incursions, a mass balance was performed over the tunnel for a pollutant P and CO, which is a gaseous tracer for the vehicular emissions. A mass balance for CO around the tunnel can be written as follows:

$$Q[\text{CO}]_a + q[\text{CO}]_v = (Q + q)[\text{CO}]_t$$

where Q and q are the molar flow rates of ambient air and vehicular emissions, respectively, through the tunnel. $[\text{CO}]_{a,t,v}$ are the respective molar concentrations of CO in ambient air, tunnel volume, and vehicular emissions. As the amount of vehicular exhaust added to the tunnel is small compared to the ambient air flow rate, we approximate q/Q to be near zero. The mass balance reduces to the following:

$$[\text{CO}]_v \approx \frac{[\text{CO}]_t - [\text{CO}]_a}{q/Q}$$

A similar balance for a pollutant P gives the following expression:

$$P_v \approx \frac{P_t - P_a}{q/Q}$$

where $P_{a,t,v}$ are the respective pollutant concentrations (in this case a PAH species) in ambient air, tunnel volume, and vehicular emissions. Taking the ratio of P_v to $[\text{CO}]_v$ gives the following result:

$$\frac{P_v}{[\text{CO}]_v} \approx \frac{(P_t - P_a)}{[\text{CO}]_t - [\text{CO}]_a}$$

This represents the ratio of pollutants emitted by the original source. This relation was applied to correct the PAH concentrations measured at Sepulveda Tunnel in each size range, using ambient data from North Main Street (18) from a concurrent field study. This site is closest to the tunnel, and the data are the best available

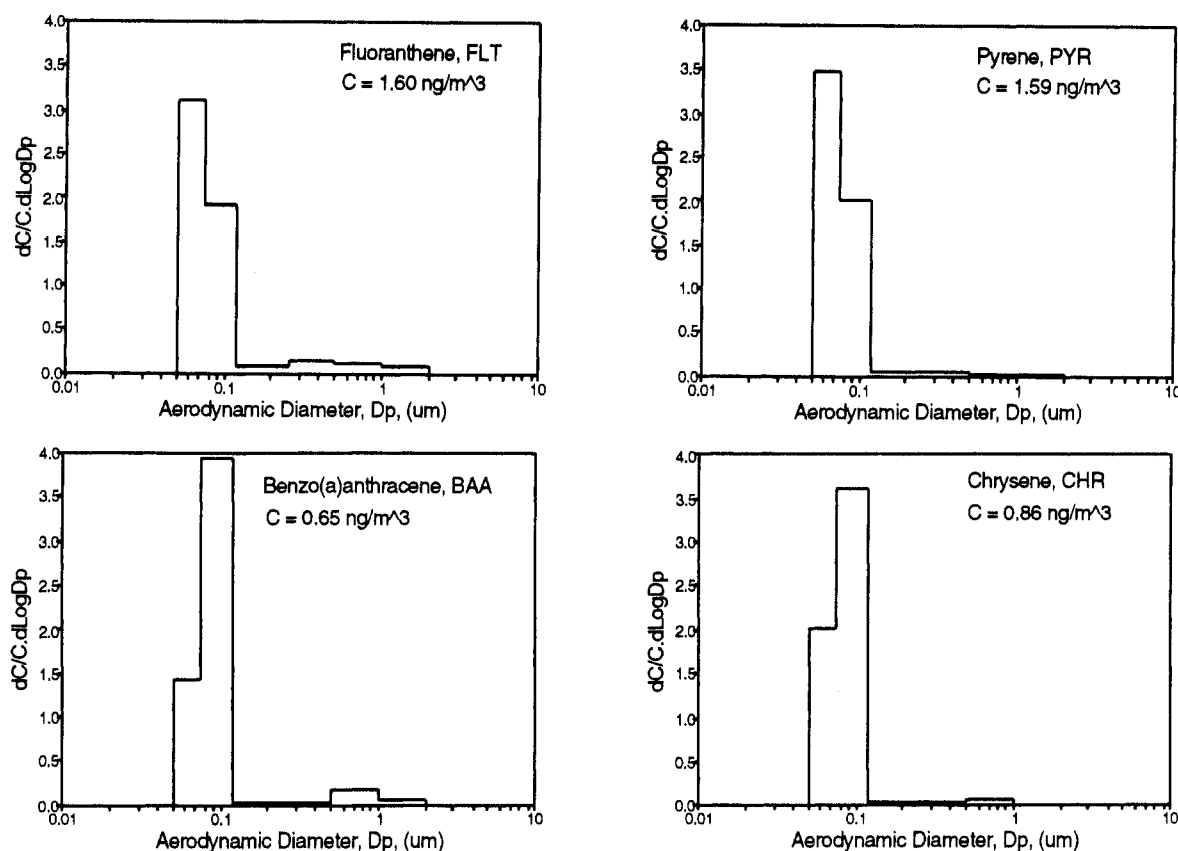


Figure 6. Estimated size distributions of 4-ring PAHs (molecular weight, 202–228) in vehicular emissions. The contribution from ambient air incursion was subtracted from the Sepulveda Tunnel measurements resulting in unimodal size distributions.

Table 3. Ratios of Concentrations of PAH to Those of Benzo[k]fluoranthene Measured in Traffic Tunnels

PAH	present study		Santa Barbara, Brazil, 1989	Carpark, Paris, 1986	Baltimore Harbor Tunnel, ^a 1989	Caldecott Tunnel		Lincoln & Holland Tunnel		Essen 1982	av
	Caldecott Tunnel, 1992	Sepulveda Tunnel, 1992				1986	1984	1983	1983		
FLT	3.9	1.9	2.5	5.2	1.1–2.7	1.1–1.4					2.4 ± 1.7
PYR	0.2	1.9	2.0	6.1	1.2–3.8	0.7–1.2					2.3 ± 2.0
BAA	0.3	1.0	1.0	2.4	0.6–0.8			2.1–5.7	0.3–1.9	0.7	1.4 ± 1.2
CHR	2.0	1.0	2.3	3.8	0.8–1.4	3.2–3.9				1.0	2.1 ± 1.2
BAP	0.5	2.5	2.6	4.1	0.4–0.6	0.9–1.4		0.8–1.4	0.3–1.3	0.7	1.6 ± 1.2
BGP	4.3	8.0	7.4	2.0	0.6–1.0	2.1–2.2	1.7	0.8–2.6	1.2–4.6	0.4	3.1 ± 2.6
INP	1.4	3.4	3.4	5.2	0.3–0.5	0.8–1.1	1.0	0.3–1.5	1.2–2.2	1.0	1.9 ± 1.5
BBF	0.4	2.2	2.3	2.0							1.7 ± 0.9
BKF	1.0	1.0	1.0	1.0	0.8–1.2	1.0	1.0	1.0	1.0	1.0	1.0

^a Ratios to benzo[b+j+k]fluoranthene shown in studies which did not speciate them. Compiled from refs 2–4, 10, and 31–33.

estimates of background concentrations near the tunnel. The corrected distributions did not show the secondary mode, leaving unimodal distributions shown in Figures 6 and 7.

The unimodal nature of the distribution is characteristic of all the PAH species in both the tunnels and is similar to the EC distributions. This is consistent with PAH and EC being formed simultaneously in combustion systems. It is known that monocyclic organic species formed in flames condense to polycyclic species (25), and there is some experimental evidence that PAHs are intermediates in soot formation (26, 27). The only PAH size distribution measured previously in vehicular exhaust aerosol (28) is from the Caldecott Tunnel for BAP, and it showed a similar unimodal structure with the mode in the 0.05–0.12- μ m size range.

The mass median diameters of EC and the different PAH species at the two tunnels are shown in Table 2 and fall in the narrow range 0.07–0.15 μ m for Sepulveda Tunnel

aerosol and between 0.06 and 0.26 μ m for Caldecott Tunnel aerosol. The size distribution of pollutants in emission aerosol is useful in identifying their sources in multimodal ambient size distributions and has been used in the case of trace inorganic species in previous studies (29).

PAH Profiles for Motor Vehicles. The relative concentrations of pollutants in the mix emitted from vehicles, as measured in a traffic tunnel, are used in developing a profile characteristic of the vehicular source. Such profiles have been used in receptor modeling to apportion the contribution of different sources to carbonaceous aerosols at a receptor site (6, 30).

Vehicular PAH profiles have been reported in previous studies (2, 4, 7). Profiles have generally been reported as ratios of PAH to benzo[e]pyrene (BEP), which is a nonvolatile and unreactive species. We did not analyze BEP in this study, as it is not included in the standard (SRM 1647a, NBS) used to calibrate the system. The profiles given in Table 3 are ratios to benzo[k]fluoranthene

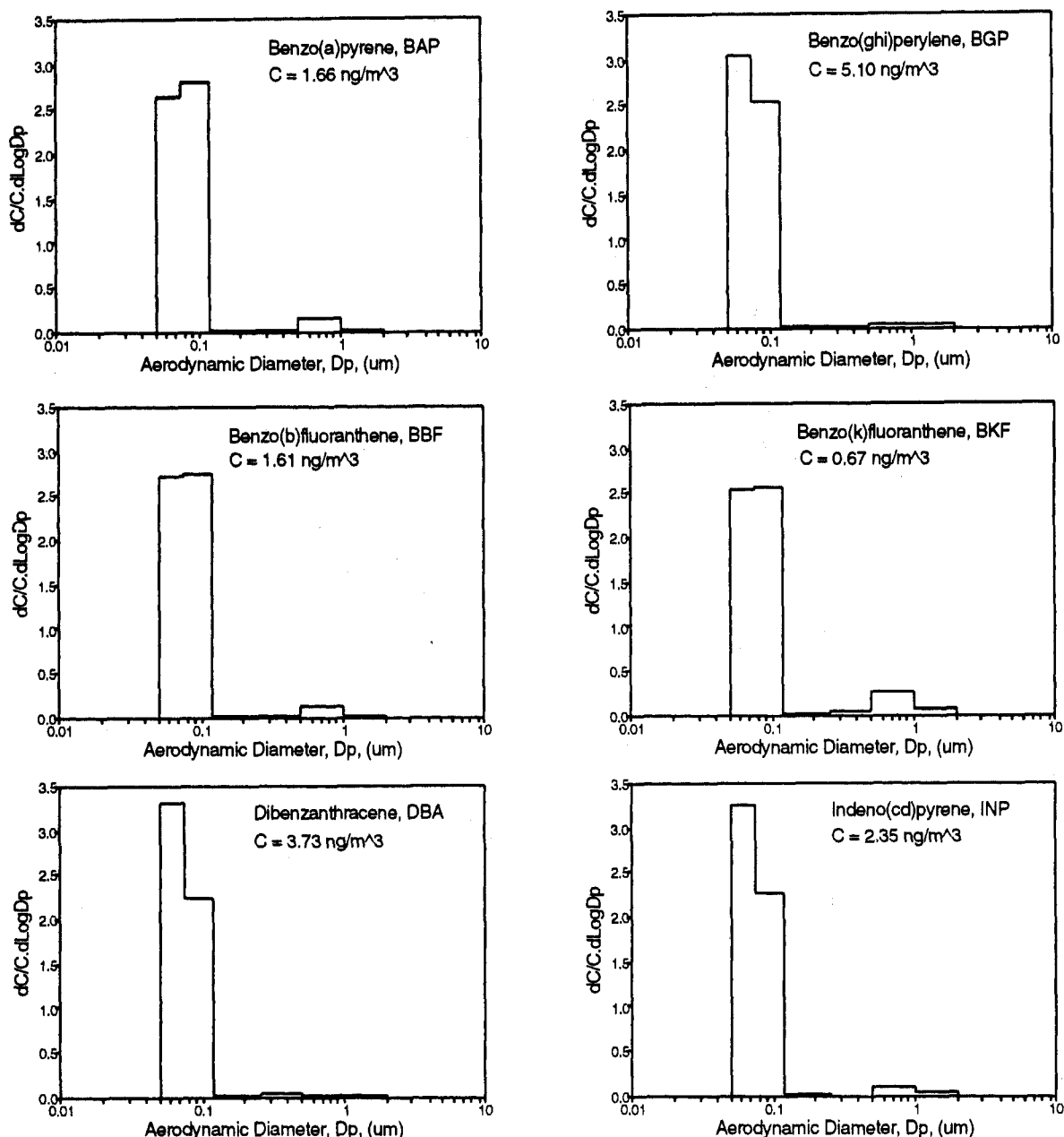


Figure 7. Estimated size distributions of 5-ring and larger PAHs (molecular weight, 252–278) in vehicular emissions. The contribution from ambient air incursion was subtracted from the Sepulveda Tunnel measurements resulting in unimodal size distributions.

(BKF), which is the least reactive, nonvolatile species measured here. For the Baltimore Harbor Tunnel (4), ratios to benzo[*b+j+k*]fluoranthene are reported as the individual isomers were not measured.

The profiles measured in the current study match well with those reported in the literature. Ratios of the nonvolatile PAHs (BAP, BGP, BBF, and INP) agree to within a factor of 3, with the exception of low BGP in Essen (31) and low INP in the Baltimore Harbor Tunnel (4). This falls within the range of agreement in previous studies (2) given the differences in sampling and analysis methods. The ratios of the lower molecular weight species (FLT, PYR, BAA, and CHR) show greater variation. These partition between particle and gas phase to different extents at different temperatures (17, 34) and their particulate concentrations vary with ambient conditions. While the motor vehicle PAH profiles from different investigations, summarized in Table 3, do show considerable variation, PAH profiles of different source classes are unique enough to be used in receptor modeling calculations (35).

The high CHR ratio and the low BGP ratio at the Caldecott Tunnel compared to Sepulveda Tunnel point to a higher fraction of diesel vehicles in the former. Other than CHR, the ratios of all other PAHs to BKF in the Caldecott Tunnel are lower than corresponding ones in the Sepulveda Tunnel, implying that Caldecott Tunnel aerosol is enriched in BKF. Previous studies have shown that diesel exhaust is enriched in FLT, CHR, and PYR (33), while gasoline exhaust has high BGP concentrations (2). Hering et al. (10) observed that the BKF/CO ratio correlated with increasing diesels in the vehicle fleet in the Caldecott Tunnel. These observations are also consistent with the low CO:EC ratio in the Caldecott Tunnel (Table 1), which points to a vehicle fleet with a larger fraction of diesels.

Conclusion

An accurate method for PAH measurement was developed combining impactor sampling with a high-resolution analysis procedure based on HPLC and fluorescence

detection. Impactor sampling minimizes losses that are significant in high-volume sampling of semivolatile organics on fiber filters. The analysis method allows the quantification of PAHs present in the aerosol with detection limits of a few picograms.

A field study was carried out at traffic tunnels to characterize the fine vehicular aerosol emissions for PAH and EC using the measurement method, which is applicable to ambient aerosol as well. PAH and EC size distributions measured in the Caldecott Tunnel are unimodal in vehicular emissions with over 85% of the mass in particles smaller than 0.12 μm aerodynamic diameter. Ambient air intrusion into the Sepulveda Tunnel, which is only 0.5 km long, resulted in small secondary peaks in the PAH distributions in the 0.5–1.0- μm size range. Estimated vehicular size distributions for these pollutants, using a mass balance over the tunnel, were unimodal.

PAH profiles were developed for the vehicular source, as ratios of concentrations of PAH to BKF measured in traffic tunnels. These showed good agreement with previously published profiles for the vehicular source. Differences in the PAH profiles measured at two traffic tunnels were attributed to different vehicle fleet compositions. The measured profiles can be used in receptor modeling estimations for source allocation.

Acknowledgments

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