

# The distribution and sources of polycyclic aromatic hydrocarbons in Narragansett Bay surface sediments

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## Abstract

Polycyclic aromatic hydrocarbons (PAHs) were measured in 41 surface sediments from Narragansett Bay, RI. All the analytes generally decrease down bay from the Seekonk, Providence and Taunton Rivers at the head of the bay. Total PAHs ranged from 0.569 to 216  $\mu\text{g/g}$  with 27% exceeding the effects range median (ERM) of 44.8  $\mu\text{g/g}$  and 73% of the stations exceeding the effects range low (ERL) of 4.02  $\mu\text{g/g}$  (Long et al., 1995).

Based on principal component analysis, the major source of the contaminants in Narragansett Bay was the Providence River while the coves and Taunton River/Mt. Hope Bay appeared to only have a limited influence on the open bay. PAH source ratios indicate that creosote and/or coal may be the most significant contributor of pyrogenic high molecular weight PAHs in Narragansett Bay, along with significant contributions from diesel exhaust.

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**Keywords:** PAHs; Narragansett Bay; Principal component analysis; Source ratios

## 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous trace contaminants in the marine environment and they vary both in their acute toxicity and sublethal effects to aquatic organisms (Neff, 1979). They may also bioaccumulate in shellfish and other benthic organisms (Stegeman and Teal, 1973; Harris et al., 1977; Fucik and Neff, 1977), which gives them a pathway to humans. Some studies have shown that bioaccumulated petroleum hydrocarbons and PAHs are released rapidly, in days to weeks, when the animals are returned to clean water (Stegeman and Teal, 1973; Lee et al., 1972; Lee et al., 1977), while other investigators have demonstrated that chronically accumulated PAHs are persistent, even after long periods of depuration (e.g. several months; Blumer et al., 1970; Boehm and Quinn, 1977). Some of these PAHs, such as benzo[a]pyrene, 7,12-dimethylbenz[a]anthracene, 3-methylcholanthrene, and benz[a]phenanthrene, have been shown to be carcinogenic to mammals (Neff, 1979). Several studies summarized by Neff (1979),

indicated that bioaccumulation factors of PAHs were much higher from seawater than sediments and that bioaccumulation from sediments could actually be attributed to PAHs that have desorbed from sediment particles into the interstitial water. In other words, PAHs in the dissolved phase were the ones considered to be bioavailable. Determination of the pore water concentration ( $[\text{PAH}_{\text{pw}}] = ([\text{PAH}_{\text{sed}}]/(f_{\text{oc}} * K_{\text{oc}}))$ ) of PAHs for the EPA sediment quality criteria is currently based on the concentration of PAHs in the sediment, the fraction of organic carbon ( $f_{\text{oc}}$ ), and the organic carbon/water partition coefficient ( $K_{\text{oc}}$ ) (McGroddy et al., 1996).

There are two primary sources of PAHs to Narragansett Bay, pyrogenic hydrocarbons from combustion sources and petrogenic hydrocarbons from petroleum sources. Past studies have indicated that the pyrogenic PAHs dominate in the sediments (Lake et al., 1979; Pruell and Quinn, 1985) while another more recent study suggests that the pyrogenic PAHs are not easily distinguished from weathered petroleum PAHs, which are ubiquitous throughout the bay (Latimer and Quinn, 1996). The major sources for pyrogenic PAHs are expected to be coal burning power plants, exhaust from diesel engines and used crankcase oil. This oil will be a mixture of both pyrogenic and petrogenic PAHs (Pruell and Quinn, 1988; Zakaria et al., 2002), and it enters the

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bay through runoff, waste water treatment facilities (WWTFs), combined sewer overflows (CSOs), and atmospheric deposition (Lake et al., 1979; Latimer and Quinn, 1996). The petrogenic PAHs can come from oil/gas/diesel spills directly into the bay or in adjacent waters as well as WWTFs, CSOs, runoff and industrial effluents (Lake et al., 1979; Latimer and Quinn, 1996; Latimer and Quinn, 1998). From the time of formation to final deposition in the sediments, PAHs are subject to many biological, chemical and physical processes. PAHs from pyrogenic sources may be strongly associated with soot particles that eventually settle in the water or on land from which they can enter the water with runoff (Lake et al., 1979). In the atmosphere and in the photic zone of the water column, the PAHs are subject to photooxidation. In addition, PAHs in the water column and in sediment, are subject to microbial degradation and partitioning to the surrounding water. The type of PAH inputs to the environment and the reactions they undergo at various points in their transport to the sediment will have a significant effect on bioavailability to both pelagic and benthic organisms.

The present study used 41 stations to evaluate the distribution of various PAH compounds in Narragansett Bay surface sediments. The relationship between PAHs in sediments and possible sources was explored statistically using principle component analysis (PCA). The groupings from the PCA were then evaluated relative to potential sources materials and geographic location.

## 2. Methods

### 2.1. Sampling

Surface sediment samples were taken in 1997 and 1998 at 41 stations (Fig. 1) throughout Narragansett Bay using a Smith-McIntyre grab sampler. Three grabs were taken from each location from which the top 2 cm were scooped into a solvent cleaned glass jar and the composite homogenized. The boat was moved up to 2–4 m between grabs so that the samples would be representative of the area from which they were taken. The samples were placed on ice in a cooler until they were returned to the laboratory where they were stored at –20 °C until analyzed.

### 2.2. PAH analysis

The extraction and analysis has previously been described (Hartmann, 2001; Hartmann et al., 2000). For the PAHs, one to six ml of four deuterated PAHs, naphthalene- d<sub>8</sub>, acenaphthene- d<sub>10</sub>, anthracene- d<sub>10</sub>, and benzo[e]pyrene- d<sub>12</sub>, each at ~2000 ng/ml in acetonitrile, were added as internal standards. Samples

were analyzed on Hewlett Packard HP5890 gas chromatograph (GC) fitted with a 5971A quadrupole mass spectrometer (MSD) using condition previously described (Hartmann, 2001; Hartmann et al., 2000) or with an Agilent 6890/5973 GC/MSD. Data was collected using selected ion monitoring (SIM), quantified on the primary ion and confirmed with secondary and tertiary ions. Twenty parent PAHs, five mono-, di-, or trimethylated PAHs and 20 PAH alkyl homologs were analyzed. A response like the parent compound was assumed except in cases where we had an authentic standard of the substituted PAHs. We used SRM 1944 from the National Institute of Standards and Technology (NIST) to confirm our alkyl homolog measurements. All values reported for the sediments are on a dry wt. basis.

### 2.3. Organic carbon analysis

Sediment OC analysis was done using a CHN analyzer. A sub-sample of the wet sediment was weighed into a tarred aluminum weighing pan, dried at 105 °C for >4 h and re-weighed. The sample was dried and re-weighed until a constant weigh was achieved. The dry weight minus the wet weight was then used to calculate the percent moisture of the sample. A 5–15 mg sub-sample was then weighed out into a tarred 5×8 mm silver capsule. Samples were then acidified with 25 µl of Milli-Q water to wet the sample followed by 25 µl of 1 N HCl. After at least one hour, 25 more µl of HCl was added and then the samples were dried at 60 °C for 8 h. The acidification and drying was done a total of three times in order to remove carbonate carbon. When this step was complete, the capsules were folded and compressed with tweezers for CHN analysis. A Carlo-Erba model 1108 CHN elemental analyzer was then used to analyze the samples.

### 2.4. Principal component analysis

Principal component analysis (PCA) was used to try and distinguish between the samples to assess different sources of PAHs and how they are being deposited in the bay. Possible sources include automotive exhaust, used crankcase oil, wood combustion, coal-fired power plants, coal tar, creosote and petroleum oils spills. All but the petroleum spills are pyrogenic sources, which gives them a similar composition. The PCA was used to elucidate linear combinations of PAHs that may be useful to distinguish between these sources as has been demonstrated by other researchers (Simpson et al., 1998; Smirnov et al., 1998; Naes et al., 1998; Dickhut et al., 2000). The PCA was run with all the PAHs, fluorenone and anthraquinone. The data were first normalized to fluoranthene to remove concentration between samples as a variable and to accentuate differences in ratios be-

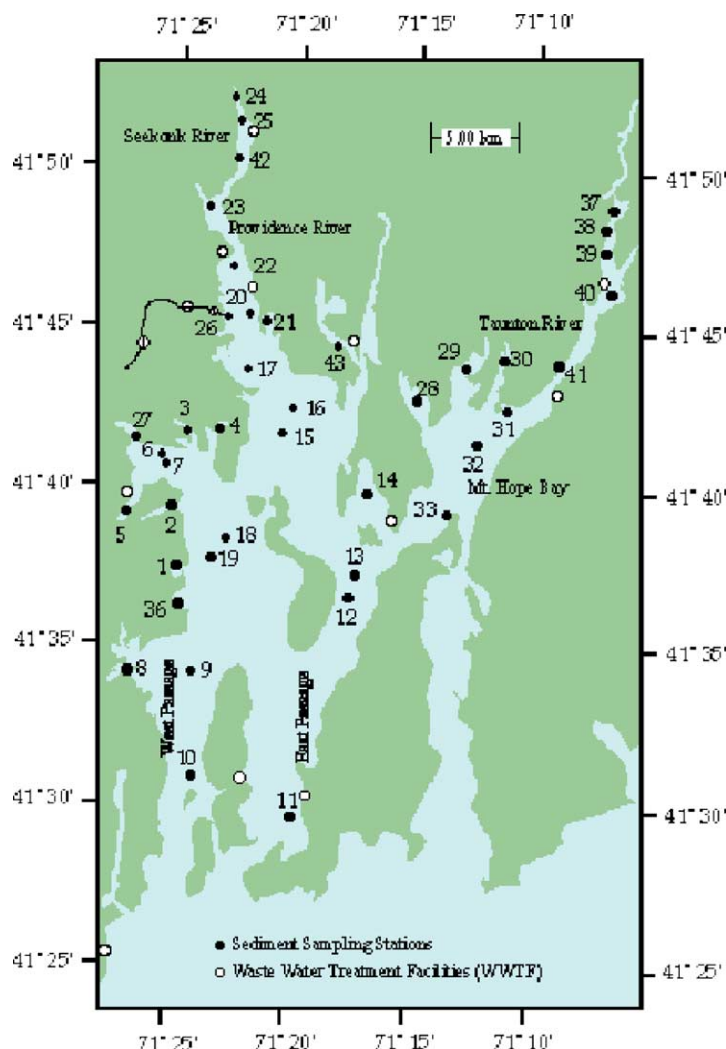


Fig. 1. Location of the 41 station locations for surface sediments in Narragansett Bay, RI taken in 1997–1998. There is no station 34 or 35.

tween analytes in the different samples. Data analysis including (PCA) was done on the correlation matrix using S-Plus 2000 from Math Soft, Seattle WA.

### 3. Results and discussion

#### 3.1. PAH concentrations

Thirty-one individual aromatic compounds including the 24 NOAA Status and Trends PAHs (O'Connor, 1998) were analyzed at 41 stations in this study (Fig. 1). In addition, twenty PAH alkyl homologs were measured. The total PAHs ( $\Sigma$  of 44 PAHs) ranged from 0.569  $\mu\text{g/g}$ , at station 2 in Potowamut Cove, to 216  $\mu\text{g/g}$ , at station 43 in the Barrington River (Fig. 2). The mean concentration of PAHs was 21.1  $\mu\text{g/g}$ , although a few very contaminated samples skew the data as the median was only 6.05  $\mu\text{g/g}$ . The later value is still above the (Long et al., 1995) effects range low (ERL) for PAHs of

4.02  $\mu\text{g/g}$ s. This is a sediment quality guideline below which the chemical concentration in the sediments would be expected to rarely have adverse biological effects. The ERL/ERM usually includes the 24 PAHs of the NOAA Status and Trends program plus any other PAHs measured. These 24 PAHs on average accounted for 70% of our total PAHs. Thirty, or 73%, of the 41 stations were above the ERL threshold and would be expected to have occasional adverse biological effects, or conversely, only 27% were below the ERL. Five of these, or 12% of the stations, were above the effects range median (ERM) of 44.8  $\mu\text{g/g}$  (Long et al., 1995), a level above which adverse biological effects occur frequently. Overall, the data indicate that sediment PAHs may have a significant impact on Narragansett Bay.

The most significant PAH contamination was at the head of the bay in the Seekonk and Providence River stations and the Taunton River stations (Fig. 2). Normalizing the data to OC did not change any of the trends. In the lower parts of the bay, PAH concentrations were

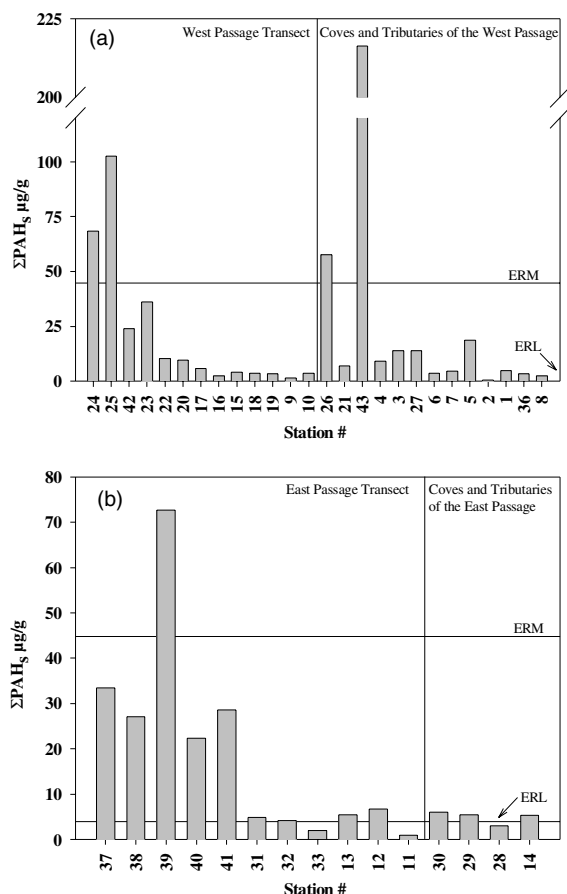


Fig. 2.  $\Sigma$ PAH surface sediment concentrations ( $\mu\text{g/g}$ ) arranged from north to south followed by the coves adjacent to the transect. The effects range median (ERM)=44.8  $\mu\text{g/g}$  and the effects range low (ERL)=4.02  $\mu\text{g/g}$  (1). (a) West Passage transect, inclusive of the Seekonk and Providence Rivers; (b) East Passage transect, inclusive of the Taunton River and Mt. Hope Bay.

generally higher in the coves than the open bay. This trend can be expected due to the coves having closer proximity to potential sources such as runoff, sewers, and marinas; they would also have less mixing and dilution, tend to be more anaerobic and have more OC, thus preserving the PAHs.

There are two notable stations in the West Passage transect which includes the Seekonk and Providence Rivers (Fig. 2a). Station 26 is in Pawtuxet Cove (at the mouth of the Pawtuxet River), which is adjacent to the lower Providence River. There are three waste water treatment facilities on the Pawtuxet River that may contribute PAHs to Pawtuxet Cove. This possibility is supported by the high concentration of LABs also reported at this site (Hartmann et al., 2000). The second notable station is #43 in the Barrington River, having a total PAH concentration of 216  $\mu\text{g/g}$ , which is the highest value found in this study. A possible source could be a landfill, closed in 1967, which is approximately 1000 m from the river and has a history of in-

dustrial disposal that includes petroleum waste from rubber manufacturing (NUS Corporation, 1991). This concentration is about 5 times the ERM for total PAHs. Two other stations (24–25) that are in an urban area are also above the ERM and 17 of 26 stations in the West passage are above the ERL guideline.

The highest concentration of PAHs in the East Passage transect, inclusive of the Taunton River and Mt. Hope Bay, was at station 39 (73  $\mu\text{g/g}$ ) at Piercee Point in the Taunton River (Fig. 2b). There were no obvious sources to this site as the immediate area is surrounded by marshes and is relatively undeveloped. This was the only station in that transect above the ERM, although 12 of the 15 stations were above the ERL guideline. The rest of the Taunton River stations were in the range of 22.4–33.5  $\mu\text{g/g}$ . From the furthest south station in the Taunton River (station 41) to the next station in Mt. Hope Bay (station 31) the PAH concentration dropped by a factor >3, then remains fairly constant throughout the rest of the East Passage transect.

### 3.2. Principal component analysis

Principal component analysis, as mentioned earlier, was used to investigate combinations of PAHs that may have similar sources and modes of input. Principal component analysis of the major PAHs resulted in the first three components (38.1%, 20.4%, and 12.6%) accounting for 71.1% of the variance in the surface sediment data. The PCA distinguished three groups of PAHs. In the first principal component, there was a broad range of PAHs that contributed, although the high molecular weight PAHs and the two ring dibenzothiophene were the most significant. The second component was more strongly influenced by the two ring compounds along with the five ring indeno[1,2,3,c,d]pyrene and the third component was compromised of two, three and four ring species. Components one and three were plotted against each other for each station, because they represent higher molecular weight compounds, and the stations grouped based on their location in the bay, which helped ascertain potential differences in sources (Fig. 3). The Seekonk and Providence river samples fell in fairly narrow ranges and were distinctly separate, even though the Seekonk River flows into the Providence River. Three of the four Providence River samples also fell within the range of the open bay samples, possibly indicating a strong influence on the PAH assemblages in the open bay. Conversely, the coves on the west side of the bay had no overlap with the neighboring open Narragansett Bay stations indicating either minimal transport of PAHs out of the coves or that the PAH transport down bay from the Providence River overwhelms any signal that might be seen from the coves. Either way, the coves are not likely to be a significant source of PAHs to the

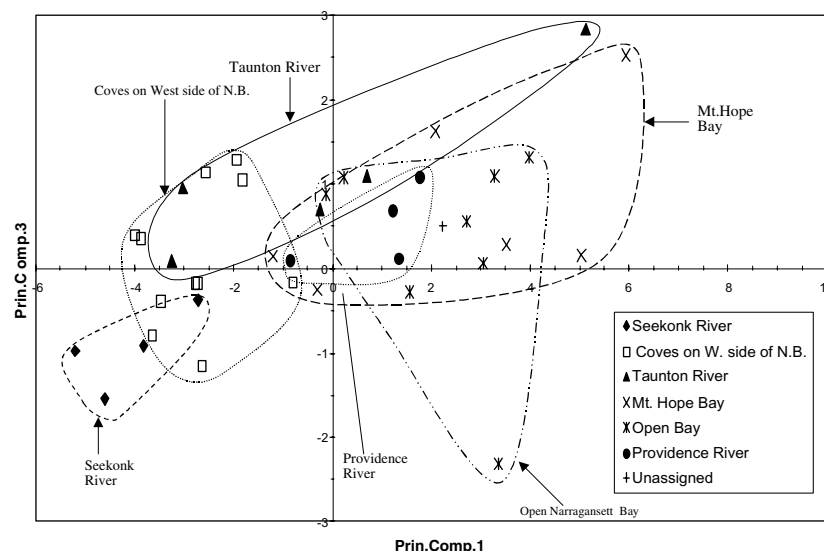


Fig. 3. Principal component analysis (PCA) of Narragansett Bay (NB) surface sediments normalized to fluoranthene. Principle component 1 accounts for 38.1% of the variance and principle component 3 accounts for 12.6%.

West Passage. This is probably due to a combination of factors. The coves, with the exception of stations 26 and 43, have lower concentrations of PAHs relative to the Providence River to begin with and are relatively small and protected so that storms and tidal currents will not cause a great deal of sediment resuspension along with the associated remobilization of PAHs. The relatively tight grouping also indicates that the various sources to the Providence River have a very similar composition to one another. The Taunton River and Mt. Hope Bay (including the coves off Mt. Hope Bay) samples had a large range, yet the two groups did not have a lot of overlap as did the Providence River–open bay stations. This indicates that there may be several point sources for the Taunton River with different compositions rather than a single source type that contributes to the whole area as in the Providence River–open bay stations. This

also implies that the Taunton River has a limited influence on Mt. Hope Bay.

### 3.3. Source ratios

Benzo[ghi]perylene (B[ghi]P) and indeno[1,2,3,c,d]pyrene (IP) have been used by several researchers as a source identifier for aerosols from automobiles (Greenberg et al., 1981; Harkov et al., 1984; Simcik et al., 1999). The ratios of IP/BghiP in our study were  $0.96 \pm 0.05$  ( $n = 26$ ) and  $0.93 \pm 0.06$  ( $n = 15$ ) for the West and East passage transects respectively. These values are the average and standard deviations rather than from a linear regression because the data covers several orders of magnitude and is not evenly distributed. These ratios are considerably higher than the  $0.33 \pm 0.06$  observed for automobile emissions, but close to those of coal

Table 1

Source ratio of materials that may contribute high molecular weight PAHs to coastal marine sediments

Source	BaA/Chry	±	BaP/BeP	±	IP/BghiP	±
Automobiles <sup>a</sup>	0.53	0.06	0.88	0.13	0.33	0.06
Coal/Coke <sup>a</sup>	1.11	0.06	1.48	0.03	1.09	0.03
Wood <sup>a</sup>	0.79	0.13	1.52	0.19	0.28	0.05
Smelters <sup>a</sup>	0.60	0.06	0.81	0.04	1.03	0.15
SRM1650 <sup>b</sup>	0.30	0.07	0.14	0.04	0.78	0.06
SRM1597 <sup>b</sup>	1.59	0.16	1.83	0.18	0.96	0.10
BC	0.73	0.07	0.99	0.10	0.17	0.02
FP	0.74	0.07	1.17	0.12	0.92	0.09
Creosote	1.84	0.03	1.42	0.06	1.20	0.14
Narragansett Bay sediments	1.09	0.24	1.07	0.26	0.95	0.05

<sup>a</sup> Ratio from Dickhut et al. (2000).

<sup>b</sup> Ratio calculated from National Institute of Standards and Technology certificate of analysis. SRM1650 = Diesel Particulate Material, SRM1597 = Coal Tar B[ghi]P = benzo[ghi]perylene; IP = indeno[1,2,3,c,d]pyrene; BaA = benz[a]anthracene; Chry = chrysene; BaP = benzo[a]pyrene; BeP = benzo[e]pyrene.

( $1.09 \pm 0.03$ ) or aluminum smelters ( $1.03 \pm 0.03$ ) (Dickhut et al., 2000). Dickhut et al. (2000) made a similar observation for Chesapeake Bay sediments in which they reported a IP/BghiP ratio of  $1.21 \pm 0.04$ . The narrow range of the observed ratios in that study indicates these isomers have a similar source. Unlike the Chesapeake Bay area, coal is not widely used in Rhode Island for power generation for either utilities or industry (Energy Information Administration, 2000), although two power

plants on the Taunton River in Massachusetts use coal. The coal signature could come from earlier inputs of coal (Goldberg et al., 1977) that are still near the surface of the sediments or from atmospheric transport. In the upper reaches of the bay, most of the sediments are anoxic right to the surface of the sediment and support few benthic organisms that would contribute significantly to bioturbation. In the lower bay, a combination of bioturbation and slower sedimentation may increase the relative con-

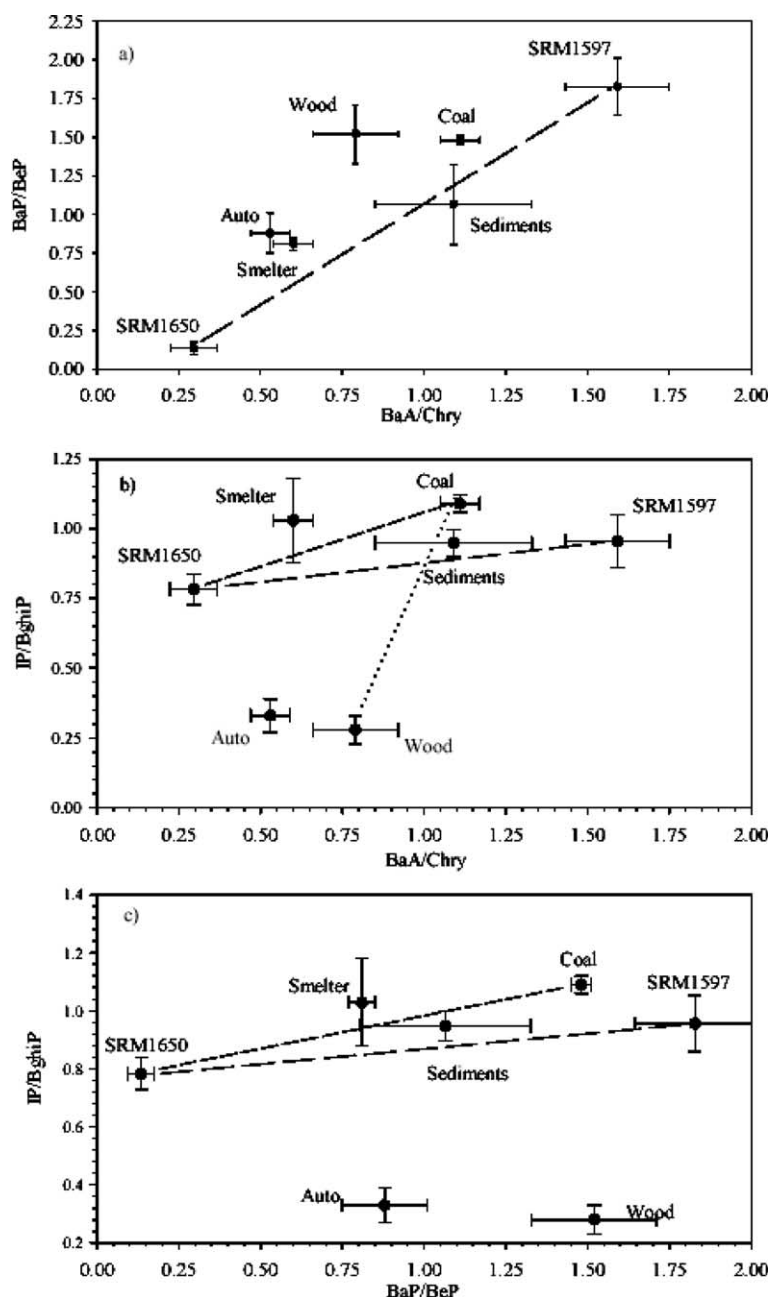


Fig. 4. Source ratios of selected reference material (wood soot, Auto exhaust, smelter emissions, coal (35), SRM1650 diesel particulate matter, and SRM1597 coal tar) relative to the average Narragansett Bay surface sediment ratios. (a) BaA/Chry vs BaP/BeP, (b) BaA/Chry vs IP/BghiP, (c) BaP/BeP vs IP/BghiP. B[ghi]P = benzo[ghi]perylene; IP = indeno[1,2,3,c,d]pyrene; BaA = benz[a]anthracene; Chry = chrysene; BaP = benzo[a]pyrene; BeP = benzo[e]pyrene.

tribution of coal derived PAHs. Alternatively, the IP/BghiP ratio is also close to the ratio calculated from reported values in SRM1650, Diesel Particulate Matter, of  $0.783 \pm 0.05$  (NIST, 1985). Our analysis of this sample gave a ratio of 0.784 ( $n = 1$ ). This suggests that diesel exhaust from trucks may also be a significant source of PAHs to Narragansett Bay. A third possibility is creosote used as a wood preservative on wood pilings and bulkheads. Creosote is a distillation product of coal tar in which the IP/B[ghi]P ratio is  $1.04 \pm 0.25$  (NIST, 1987). An analysis of coal tar standard SRM1597 gave a ratio of 0.96 ( $n = 2$ ) and a local creosote sample had a ratio of 1.20 ( $n = 1$ ), which is higher than the published average, but within the range of reported values. The SRM1597 was used in the following source apportionment as a proxy for creosote because it is a standard reference material.

Two other isomer ratios, benz[a]anthracene (BaA) to chrysene (Chry) and benzo[a]pyrene (BaP) to benzo[e]pyrene (BeP), were explored to try and distinguish between the possible sources of PAHs to the bay (Dickhut et al., 2000). The ratios of BaA/Chry were  $1.08 \pm 0.22$  ( $n = 15$ ) and  $1.10 \pm 0.26$  ( $n = 26$ ) in the East and West passages respectively. The BaP/BeP ratios were  $1.13 \pm 0.24$  ( $n = 15$ ) and  $0.97 \pm 0.20$  ( $n = 26$ ) respectively. These two ratios are not as tight as the IP/B[ghi]P ratio which could reflect differences in degradability between compounds and/or different sources. Overall, these are small variations relative to different possible source materials (Table 1). The various mixing ratios in several reference materials were plotted against one another to establish possible mixing lines (Fig. 4). The average sediment value for BaP/BeP vs BaA/Chry fall just to the right of any potential mixing lines (Fig. 4a). This may be due to enrichment of the BaA over Chry during atmospheric transport (Dickhut et al., 2000). The closest mixing line would be between SRM1650 and SRM1597. The IP/B[ghi]P vs BaA/Chry plot has the sediments falling along two mixing lines, one within the coal and wood mixing line, closer to the coal, and the other between SRM1650 and SRM1597 (Fig. 4b). A coal-wood mix is discounted because this is the only plot in which this mixing line works. Additionally, the spread in the data is along the SRM1597-SRM1650 mixing line, which is more suggestive of these components as sources. The IP/BghiP vs BaP/BeP plot shows the sediments fall in between several source materials (Fig. 4c). The closest mixing line, as in the first two plots, is for the SRM1650 to SRM1597, although the line is also close to the coal to SRM1650 line. Assuming aluminum smelters and wood are minor contributors in this area and applying linear mixing as described by Dickhut et al. (2000) between coal and SRM1650 along BaP/BeP (Fig. 4c), coal accounts for ~83% of the high molecular weight PAHs in Narragansett Bay.

$$\left( \frac{(R_{\text{sed}} - R_{1650})(R_{\text{coal}} + 1)}{(R_{\text{coal}} - R_{1650})(R_{\text{sed}} + 1)} \right) \times 100 = \text{Coal}\%, \quad (1)$$

where  $R$  is the ratio of BaP/BeP and the subscript is one of the components. Applying the linear mixing to the SRM1597 and SRM1650 contributions along BaP/BeP, coal tar would contribute ~75% of the high molecular weight PAHs and diesel exhaust 25%. Due to the mixing lines more consistently falling closer along the SRM1650 to SRM1597 line, coal tar (i.e. creosote) is probably a more significant contributor of high molecular weight PAHs than coal in most of Narragansett Bay. Overall, the source ratios suggest that creosote and diesel soot are the major contributors of high molecular weight PAHs with some contributions from other sources such as coal. An earlier study found that coal tar was locally important (Lake et al., 1979), but not to the extent found in this work. Lake et al. found that a combination of petroleum and combustion products contributed to the total PAH load in the Bay (Lake et al., 1979). A plot of the phenanthrene/anthracene vs fluoranthene/pyrene (not shown) indicates that there is a contribution from petroleum in about half the samples while the rest are strongly pyrogenic.

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