

Urban Sprawl Leaves Its PAH Signature

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The increasing vehicle traffic associated with urban sprawl in the United States is frequently linked to degradation of air quality, but its effect on aquatic sediment is less well-recognized. This study evaluates trends in PAHs, a group of contaminants with multiple urban sources, in sediment cores from 10 reservoirs and lakes in six U.S. metropolitan areas. The watersheds chosen represent a range in degree and age of urbanization. Concentrations of PAHs in all 10 reservoirs and lakes increased during the past 20–40 years. PAH contamination of the most recently deposited sediment at all sites exceeded sediment-quality guidelines established by Environment Canada, in some cases by several orders of magnitude. These results add a new chapter to the story told by previous coring studies that reported decreasing concentrations of PAHs after reaching highs in the 1950s. Concurrent with the increase in concentrations is a change in the assemblage of PAHs that indicates the increasing trends are driven by combustion sources. The increase in PAH concentrations tracks closely with increases in automobile use, even in watersheds that have not undergone substantial changes in urban land-use levels since the 1970s.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) represent the largest class of suspected carcinogens (1) and can present a threat to aquatic life (2). The presence and distribution of PAHs in the environment are largely a product of the incomplete combustion of petroleum, oil, coal, and wood (3). Anthropogenic sources such as vehicles, heating and power plants, industrial processes, and refuse and open burning are considered to be the principal sources to the environment (4). On the basis of 1989 data, vehicles produced 11% of PAH emissions in the United Kingdom, domestic coal burning produced 84%, and industrial processes produced 3% (5). Several studies in the 1970s and 1980s reported decreasing trends in PAH concentrations in the environment on a regional scale (United States and Europe) since their peak in the 1950s and 1960s (6–9), on the basis of data from sediment cores from remote and urban lakes and rivers. These reductions have been attributed to reduced use of coal for home heating, industrial emissions controls, and increased efficiency of power plants (7, 9–11).

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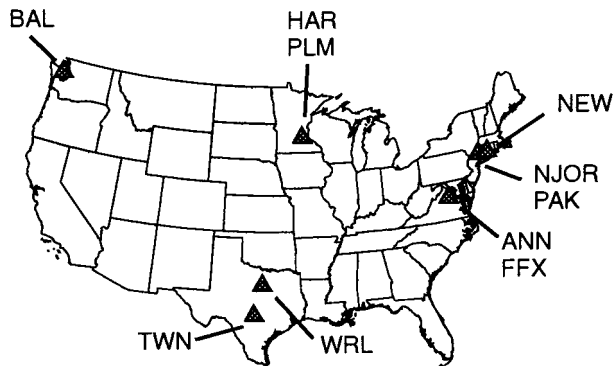


FIGURE 1. Locations of sampling sites. Abbreviations correspond to lake and reservoir names given in Table 1.

While loads of PAHs from some sources may have decreased, the changing face of the urban landscape has resulted in an increase in another source of PAHs: vehicle use. Growth in the use of land for residential and commercial purposes in the United States now far outstrips growth in population (12), a phenomenon termed urban sprawl. Increasing sprawl has resulted in decentralized employment and workplace facilities and greater dependence on vehicles, as reflected in number of miles traveled per capita and per vehicle (13, 14). What effect, if any, has this change had on urban water bodies?

As a part of the U.S. Geological Survey National Water Quality Assessment (NAWQA) Reconstructed Trends program, trends in PAHs were tracked over the last several decades to the mid-to-late 1990s in sediment cores from 10 lakes and reservoirs in six U.S. metropolitan areas. This study indicates that trends in PAH concentrations in developed watersheds over the last three decades, rather than decreasing, are increasing and that the increases may be linked to the increasing amount of urban sprawl and vehicle traffic in urban and suburban areas.

Methods

Sediment cores from lakes and reservoirs can be used to reconstruct historical trends in water quality (15, 16). Trends are recorded for hydrophobic, persistent compounds, such as PAHs, that bind to sediment particles. PAHs sorb to particulates and are transported to receiving water bodies via atmospheric deposition (17), sewage effluent (18), and surface runoff (19). For this study, sediment cores were collected from seven reservoirs and three lakes across the United States (Figure 1). Land use in the watersheds of these reservoirs and lakes is largely mixed residential and commercial with percent urban land use ranging from 25 to nearly 100% (Table 1). Three sites are in watersheds experiencing rapid growth since the 1970s (58–122% increase in urban land use), three sites are in watersheds experiencing moderate growth (26–36%), and four sites are in watersheds with relatively stable levels of urban land use (0–5%). The onset of development ranges from the early 1900s (e.g., Lake Harriet and Newbridge Pond) to as recently as the 1970s (e.g., Palmer Lake).

Cores were collected from the deepest part of the lake or in the lower part of the reservoir and sectioned into vertically discrete subsamples for analysis of ¹³⁷Cs and PAHs. ²¹⁰Pb was analyzed for selected lakes. Samples were also analyzed for major and minor elements, chlorinated organic pesticides, and PCBs (not presented in this paper).

TABLE 1. Selected Characteristics of Lakes Sampled

reservoir or lake	sampling date	depth of water (m)	lake area (km ²)	watershed area (km ²)	% urban, 1970s ^a	% urban, 1990 ^b	% urban change, 1970s–1990 ^c
Lake Anne, VA (ANN)	Jun 1996	6.5	0.13	2.3	44.8	85.7	91
Lake Ballinger, WA (BAL)	Jun 1998	9.9	0.40	13.7	93.0	96.5	4
Lake Fairfax, VA (FFX)	Sep 1997	3.8	0.11	8.4	58.8	92.8	58
Lake Harriet, MN (HAR)	Jul 1997	21.3	1.2	6.1	79.7	79.7	0
Newbridge Pond, NY (NEW)	Sep 1997	2	0.042	7.9	99.6	99.6	0
Orange Reservoir, NJ (NJOR)	Sep 1997	nr ^d	0.35	11.7	77.1	97.1	26
Lake Packanack, NJ (PAK)	Sep 1997	3	0.33	4.8	73.2	93.2	27
Palmer Lake, MN (PLM)	Jul 1997	1	0.13	64.9	29.6	65.6	122
Town Lake, TX (TWN)	Aug 1998	8.5	0.73	404	23.8	25.1 ^e	5
White Rock Lake, TX (WRL)	Jun 1996	4.9	4.4	265	56.4	76.7	36

^a Based on ref 20. ^b Based on ref 21. ^c Sites grouped by percent change in urban land use: rapid (ANN, FFX, PLM), moderate (NJOR, PAK, WRL), and stable (BAL, HAR, NEW, TWN) urbanization. ^d nr, not recorded. ^e Based on ref 22.

TABLE 2. Sediment Accumulation Rates and Age Assignments in Core

lake	basis of age dating ^a	corroborating evidence for age dating ^a	approx time period of core	sediment thickness (cm)	av linear sedimentation rate (cm/yr)	av mass accumulation rate (g cm ⁻² yr ⁻¹)
Lake Anne	a, c, e, f	d	1968–1996	17	0.60	0.18
Lake Ballinger	c	d, e	1947–1998	23	0.45	0.11
Lake Fairfax	a, c	d	1952–1997	52	1.2	0.86
Lake Harriet	b, c	d, e	~1800–1997	16	0.60	0.07
Newbridge Pond	c	d, e	1952–1997	54	1.2	0.32
Orange Reservoir	d, e	e	1949–1997	32	0.66	0.24
Lake Packanack	a	d, f, Cu ^b	1932–1997	42	0.64	0.11
Palmer Lake	c, d	e	1949–1997	38	0.78	0.41
Town Lake	a, c	d, e	1959–1998	110	2.7	1.95
White Rock Lake	a, c	d, e	1913–1996	105	1.3	1.13

^a a, construction date of reservoir; b, ²¹⁰Pb; c, ¹³⁷Cs; d, DDT and(or) PCB profiles; e, total Pb peak; f, modeled exponential decrease in sedimentation rate following approach of ref 24. Sampling date used at top of all cores. ^b Dramatic trends in copper were matched to historical copper–sulfate treatments of the lake.

PAHs and alkyl-substituted PAHs (alkyl-PAHs) were extracted, isolated, and analyzed using a variation of the procedure of Furlong et al. (23). Briefly, wet bottom sediment was extracted overnight with dichloromethane in a Soxhlet apparatus. The extract was reduced and filtered. An aliquot of the extract was processed by automated gel-permeation chromatography (GPC) using a styrene–divinyl benzene column and a dichloromethane eluent to remove sulfur and partially isolate the PAHs from coextracted high molecular weight interferences such as humic substances. The PAH fraction was reduced in volume and solvent-exchanged to ethyl acetate in a micro-Snyder column. An aliquot of internal standard solution (a mixture of perdeuterated PAHs) was added to the final extract. PAHs were separated, identified, and quantified by capillary gas chromatography (GC) coupled to mass spectrometry (MS). The parent and alkyl-PAHs were resolved on a fused silica capillary GC column. Selected ion monitoring (SIM) was used to reduce chemical interferences and improve sensitivity. Parent PAHs were identified and quantified by comparison to authentic standards. Individual alkyl-PAHs were quantified when authentic alkyl-substituted standards were available. The multiple isomeric alkyl-PAHs were quantified from SIM mass chromatograms as the sum of all isomers at each alkylation level (C1-naphthalene, C2-naphthalene, etc.). When authentic alkyl-substituted standards were unavailable, a parent PAH was used for quantitation. Nineteen parent PAHs, 10 specific alkyl-PAHs, and the homologous series of alkyl-PAHs were determined for this study. Total PAHs (Σ PAH) was computed as the sum of these, excluding perylene.

Sediment ¹³⁷Cs activity profiles were measured in all 10 reservoirs and lakes (Table 2) by counting freeze-dried sediments in fixed geometry with a high-resolution, intrinsic germanium detector γ -spectrometer. The method is similar

to that reported by Callender and Robbins (24). Activity concentrations of ²¹⁰Pb were measured in two older reservoirs and two lakes (Table 2) by high-precision γ -ray spectrometry. A low-energy photon planar detector was used to detect γ -ray emissions between 40 and 400 keV (including ²¹⁰Pb), and a high-purity germanium coaxial detector was used for γ -ray emissions between 200 and 2000 keV (including ¹³⁷Cs).

Date of deposition for sediment intervals within each core was based on a variety of date–depth markers. Those indicators on which dates were based and those used to corroborate the dates assigned are listed in Table 2. The depth of the pre-reservoir surface in cores, for example, was matched with reservoir construction date in 5 of the 7 reservoirs. (In the other two, Newbridge Pond and Orange Reservoir, the age of the reservoirs and the presence of DDT, PCBs, and ¹³⁷Cs immediately above the pre-reservoir surface indicated a discontinuity.) The other major date–depth markers used were ¹³⁷Cs first occurrence (1953.0), ¹³⁷Cs peak concentration (1964.0), and sampling date. Additional and/or corroboratory date–depth markers included unsupported ²¹⁰Pb profile (Lake Harriet only), lead peak (mid-1970s (25)), and DDT and PCB profiles (first occurrence in the 1940s and peaks in the early and late 1960s, respectively (16)).

Cesium-137 profiles provided date markers in 8 of the 10 reservoirs and lakes. The core from White Rock Lake is a good example: ¹³⁷Cs is first detected at a depth of 58 cm, peaks sharply at 48 cm, and then exhibits a smooth exponential decrease to the top of the core. In addition to providing age control, this type of profile is strong evidence that sediments have not been disturbed by bioturbation or other post-depositional mixing. At one site, Lake Harriet, because the core penetrated sediment deposited long before the release of ¹³⁷Cs into the environment ²¹⁰Pb was used in addition to ¹³⁷Cs to provide date information for the lower

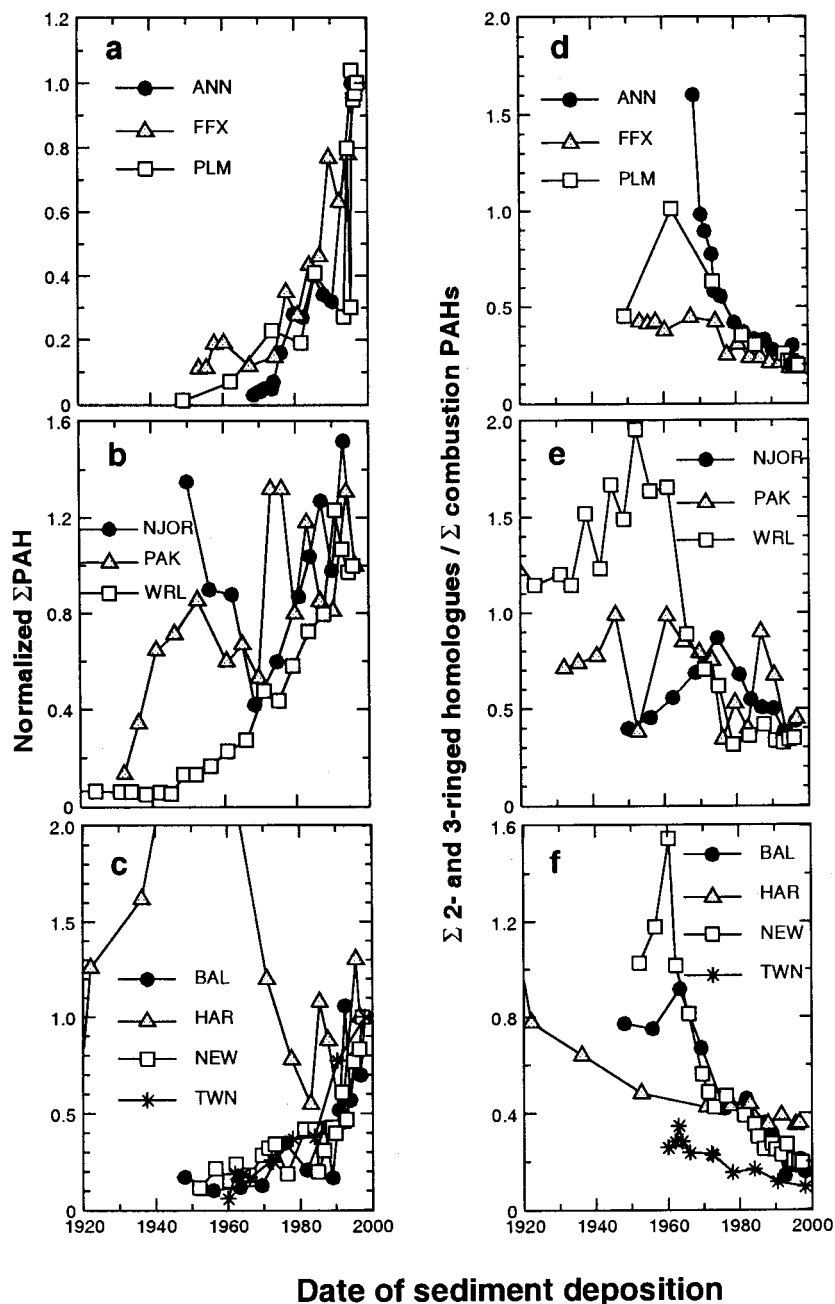


FIGURE 2. Normalized Σ PAH and ratios of PAH assemblages in sediment cores. Trends in Σ PAH among sites are compared by normalizing concentrations in samples from each core to the most recently deposited sample, thus showing trends relative to current conditions. Normalized Σ PAH are shown for watersheds with recent rapid increases (a), moderate increases (b), and stable levels (c) of urbanization. Ratios of PAH assemblages as indicators of PAH source are shown for recent rapid increases (d), moderate increases (e), and stable levels (f) of urbanization. Abbreviations for lake and reservoir sites are as given in Table 1.

portion of the core, following the constant delivery rate model (26).

A date–depth marker other than ^{137}Cs was used for 2 of the 10 lakes (NJOR and PAK) because the ^{137}Cs peak was not sufficiently defined. For NJOR, a well-defined DDT peak was evidence of undisturbed sediments, and a date of 1964 was assigned to that depth. For PAK, dates were assigned based on occurrence of Cu: treatment of the lake with CuSO_4 began in about 1953, which we correlated to the sharp increase in Cu in the core at 18 cm. For all cores, other date markers such as the DDT peak in the early to mid-1960s and the lead peak in the mid-1970s were checked to see if their depths within the cores were consistent with the dates assigned.

Lakes Anne and Packanack were the only two lakes for which the assumption of constant sedimentation rate seemed

questionable. In these two lakes the initial dating scheme placed the lead peak in the 1980s, unrealistically late. This indicated that mass sedimentation rates must have decreased substantially between the occurrence of the older date markers (^{137}Cs for Anne and Cu for Packanack) and the top of the core. We therefore assumed that a gradual change in sedimentation rate occurred, possibly caused by gradual completion of urban construction in the watersheds resulting in a corresponding reduction in erosion. We modeled this change using an exponential function following the approach of ref 24, where an exponential decrease in mass accumulation rate was demonstrated in many reservoirs.

Although confidence intervals cannot be assigned to the dates corresponding to individual sample intervals in the cores, the consistency of multiple date markers suggests that

TABLE 3. Concentrations of Selected PAHs in Cores

lake ^b	surficial (top of core) concn				range in concn in core ^a			
	benzo[a]pyrene	fluoranthene	pyrene	total PAH	benzo[a]pyrene	fluoranthene	pyrene	total PAH
White Rock Lake	154	262	219	2,790	e3-154	e9-317	e8-262	135-3440
Town Lake	585	1320	1100	11,400	33-582	57-1,320	66-1,100	670-11 400
Lake Packanack	528	815	754	12,100	41-744	147-960	118-1,000	1610-15 900
Orange Reservoir	913	1480	1260	21 600	314-1500	522-2660	524-2370	13 000-29 100
Lake Anne	1020	4850	3410	30 300	20-1020	62-4850	54-3410	1030-30 300
Lake Fairfax	1540	3500	2680	30 800	124-1540	346-4170	278-2690	3410-30 800
Lake Harriet	1960	4210	3160	35 900	e8-3430	39-16 800	26-12 700	430-48 300
Palmer Lake	2110	5480	4380	44 000	19-2110	65-5930	46-4600	518-45 700
Lake Ballinger	2350	4120	4340	46 300	72-2910	185-7420	263-7430	4810-49 000
Newbridge Pond	10 800	27 200	22 100	224 000	597-10 800	1450-27 200	1,590-22 100	26 300-224 000
Canadian SQGs ^c	31.9/762	111/2355	53.0/875					

^a Estimated values are made with 'e'. ^b Lakes are ordered by total PAH concentrations in surficial samples. ^c Canadian sediment-quality guidelines; interim sediment-quality guideline/probable effect level (27).

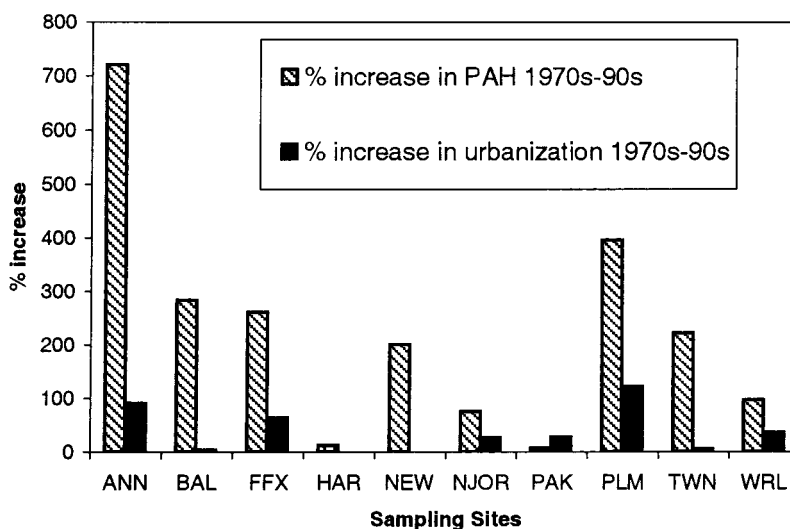


FIGURE 3. Comparison of percent increase in Σ PAH to percent increase in urban land use for each watershed (based on refs 27 and 22) from 1975 to 1995.

most dates are probably within a few years of the actual deposition dates. Pronounced peaks and systematic variations in chemical profiles indicate that contaminant trends have not been obscured by post-depositional mixing. We conclude therefore that these cores are recording multi-year to decadal trends in the contaminants of interest.

Results

Modest to dramatic increases in Σ PAH concentrations are seen in sediments deposited in all 10 watersheds over the last 20-40 years (Figure 2a-c). In all three rapidly urbanizing sites, Σ PAH has increased sharply from pre-development levels to the present (Figure 2a). Recent concentrations were 1-2 orders of magnitude above pre-development concentrations at these sites. Among the moderately urbanizing and stable sites, differences in trends reflect the age of the onset of urbanization (Figure 2b,c). In those reservoirs in which the onset of urbanization was largely post-1960, Σ PAH increases steadily from the date of reservoir construction to the present, similar to the trends seen in the rapid-urbanization watersheds. In the three watersheds that underwent pre-1960s urbanization and in which the lake or reservoir was in place to record water-quality changes (HAR, PAK, and NJOR), Σ PAH peaked in the 1950s, similar to trends reported at other sites by other investigators (6-9), and then decreased (Figure 2b,c). In each case, however, this decrease was followed by a subsequent increase beginning in the 1960s in PAK and NJOR and in the 1980s in HAR.

Although many characteristics of the temporal trends are similar at these sites, the magnitude of concentrations varies greatly (Table 3). Concentrations of PAHs are a function not only of source strength but also of sedimentation rate and sediment dilution and thus can vary widely from site to site. The smallest concentrations are in White Rock Lake in Dallas, TX, with a Σ PAH at the sediment surface of 2790 μ g/kg. The largest concentrations are in Newbridge Pond, a small reservoir in an older residential and commercial neighborhood on Long Island in New York, with a Σ PAH in recent sediments of 224 000 μ g/kg. Intermediate concentrations, ranging from about 10 000 to 50 000 μ g/kg in surficial sediments, occur at the other eight sites.

The assemblage of PAH compounds in the cores indicates a general shift in PAH source over the last 40 years from uncombusted to combusted fossil fuels, coincident with increased concentrations and urbanization. Uncombusted sources (e.g., oil seeps, petroleum spills) contain predominantly two- and three-ringed compounds, whereas combustion (e.g., vehicle exhaust, domestic heating with coal, forest fires) results in predominantly four- and five-ringed species (7, 18). Figure 2d-f shows the change over time in the ratio of two- and three-ringed PAH compounds plus homologues to the sum of the major "combustion" PAHs (28). The combustion PAHs are fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, and benzo[g,h,i]perylene. A decrease in this ratio indicates

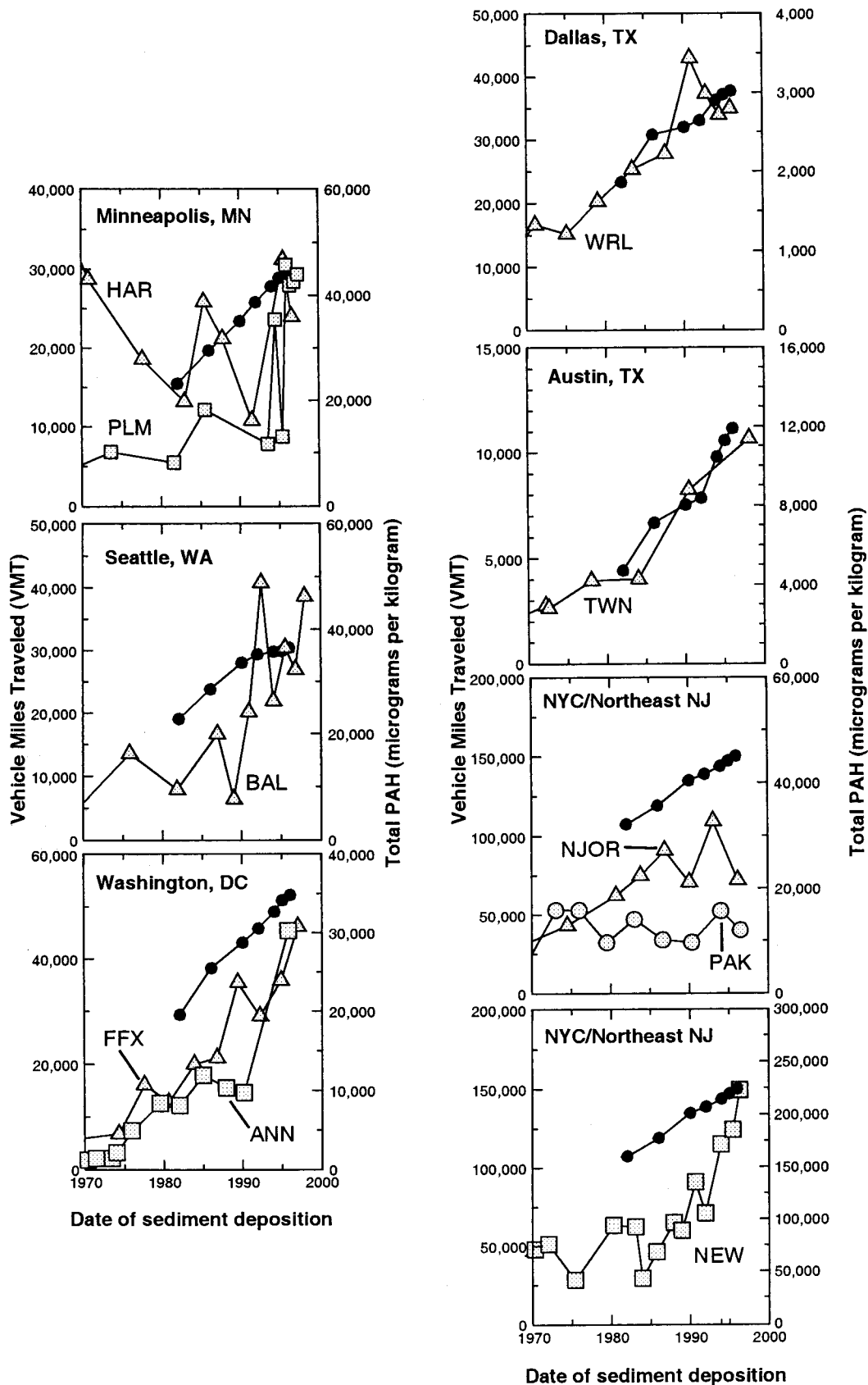


FIGURE 4. Comparison of increases in vehicle miles traveled (VMT) to changes in Σ PAH. Vehicle miles traveled per year over the entire metropolitan center (14) are shown by filled circles; Σ PAH for the lake or reservoir from the watershed within that area are shown by shaded symbols.

a shift from uncombusted to combusted fossil fuels as the PAH source. The proportion of noncombustion sources

relative to combustion sources increased during the early history of four watersheds (WRL, NEW, NJOR, and BAL),

peaking from the 1950s to the 1970s; all four sites had some degree of pre-1960s urban development. In contrast, over the last few decades, 9 of the 10 sites show an increase in the importance of combustion sources relative to noncombustion sources coincident with increasing concentrations. Only PAK shows a varying ratio with no trend.

The change in Σ PAH from the mid-1970s to the mid-1990s is compared to the change in the amount of urban land use for each watershed for the same period (Figure 3). In 9 out of 10 cases (PAK being the exception), increase in Σ PAH outpaces increase in urbanization. At some sites (BAL, NEW, and TWN), there was virtually no increase in the amount of urban land use during that period, yet Σ PAH more than doubled.

To investigate the possible effect of traffic on trends in Σ PAH, trends in vehicle use for the six metropolitan areas in this study were plotted versus Σ PAH (Figure 4). Trends in vehicle miles traveled (VMT) for each metropolitan area and Σ PAH for the 1970s–1990s show similar increases. This comparison is limited in that the VMT data are for entire metropolitan areas and are not specific to the sampled watersheds. In most cases the watersheds contributing runoff to the sampled lakes are much smaller than the urban area, and changes in traffic volume over time may not necessarily match those of the larger urban area. The possible correlation can be better investigated by looking at two reservoir watersheds that correspond well to the urban areas of the VMT data: Town Lake (TWN) in Austin and White Rock Lake (WRL) in Dallas, TX. Town Lake receives drainage from most of urban Austin; nonurban sediments from the Colorado River, which forms Town Lake, are trapped by a series of upstream reservoirs. The White Rock Lake Watershed covers 264 km² of diverse urban land use that is generally a similar mix of residential, commercial, industrial, and transportation land uses as greater Dallas (29). For these two sites, the slopes of PAH concentrations and VMT versus time are very similar (Figure 4), indicating that increases in PAHs are proportional to increases in vehicle traffic, at least for these two locations.

Discussion

The results of this study indicate a reversal in the decreasing trend in PAHs in older urban watersheds; a rapid increase in PAHs in all watersheds over the last two decades, resulting in severe degradation of sediment quality at most of these sites; and a possible relation between vehicle traffic and recent deterioration of sediment quality with respect to PAHs in receiving water bodies in urban areas of all ages.

The decreasing trends in PAHs reported in studies carried out in the 1970s and 1980s have been largely attributed to the transition from home heating with coal to the use of oil and natural gas and increased efficiency of centralized power plants (6–9). A possible reversal of the decreasing trend is suggested by a few data points in two earlier studies, one rural and one urban (7, 8). Cores from older urban lakes sampled in this study (HAR, PAK, and NJOR) record the presence of the 1950s peak and subsequent decreasing trend but also document the reversal of the decreasing trend with variable but increasing PAH concentration to the present (1990s). This suggests that at older urban sites the improvements to sediment quality caused by changes in home heating and power generation technology have since been overwhelmed by increases in other sources of PAHs.

The increasing trends in Σ PAH in all 10 water bodies over the last 20–40 years have resulted in high to extremely high concentrations in recently deposited sediments. To put the concentrations of PAHs analyzed in this study into an ecological perspective, the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life (27) for three of the most prevalent PAHs are listed at the bottom of Table 3. Concentrations in surficial sediments (top of core) for all

10 lakes exceed the interim freshwater sediment quality guidelines (ISQGs—the concentration below which adverse effects are unlikely to occur) for these three compounds and 6 or more of the 10 lakes exceed the probable effect levels (PELs—the concentration above which adverse biological effects are expected to occur) for these compounds. For the worst case, Newbridge Pond, concentrations of benzo[a]pyrene, fluoranthene, and pyrene are 14, 12, and 25 times the PELs, respectively. Clearly, increases in PAHs in urban water bodies have degraded sediment quality to the point that it is an ecological concern.

The increase in PAHs in new urban settings cannot be attributed solely to urbanization of the watersheds (Figure 3). This is most clearly illustrated by those watersheds in which urban levels are stable (Table 1; BAL, HAR, NEW, and TWN). The increase in PAH concentrations in these watersheds is, however, coincident with increases in automobile use (Figure 4). Among the sources of PAHs related to automobiles are tire wear, crankcase oil, roadway wear, and car soot and exhaust (19, 30–32). Trends in Σ PAH from the 1970s to the present compare well to trends in VMT on freeways and major arterial streets for the associated urban areas for all sites and ages of urban development (Figure 4).

One intriguing possibility suggested by the results of this study is that urban growth or sprawl outside the watershed may adversely affect water quality within the watershed. This is most evident when a watershed has undergone only a relatively minor change in degree of urbanization. For example, Austin, TX, is one of the most rapidly growing cities in the country, but the majority of the growth there has occurred around the fringes of the city and outside of the watershed of Town Lake. This growth has contributed to large increases in vehicle traffic in the Town Lake watershed; traffic on MoPac Expressway, most of which lies within the watershed and which crosses Town Lake, almost doubled between 1990 and 1997 (33). The large increases in traffic offer an explanation for why PAHs more than doubled in Town Lake from 1975 to 1990 while percent urban land use only increased by 5%. This suggests that urban sprawl in outlying areas may affect traffic patterns and water quality in the inner city.

The similar trends in VMT and PAHs in reservoir and lake sediments seen here indicate that, in the absence of reductions in vehicle-related PAH releases, Σ PAH will continue to increase at rates comparable to those projected for automobile use. Reduction of PAHs in the environment will not be easily achieved. Although fluxes of several contaminants (nitrogen oxides, airborne particulates <10 μ m, carbon monoxide, and volatile organic compounds) in transportation-related air emissions in the United States have been decreasing since the 1970s (34), PAHs in the environment, as shown here, are continuing to increase coincident with increasing VMT. This apparent contradiction may result from the fact that there are several sources of vehicle-related PAHs in addition to exhaust, including asphalt wear, tire wear, and leaks and spills of engine oil. The multiple vehicle-related sources, coupled with increasing dependence on the automobile, highlight the complexity of reducing PAH releases into the environment.

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