ANTHROPOGENIC TRACE ELEMENTS AND POLYCYCLIC AROMATIC HYDROCARBON LEVELS IN SEDIMENT CORES FROM TWO LAKES IN THE ADIRONDACK ACID LAKE REGION

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Abstract. Sediment cores were taken from the remote Sagamore and Woods Lakes in New York State's Adirondack acid lake region and analyzed for 3 to 7 ring polycyclic aromatic hydrocarbons (PAHs) and Ag, Al, As, Be, Cd, Cr, Cu, Hg, Ni, Pb, Se, Sn, Tl, V, and Zn. With the exception of perylene, all of the parental PAHs, e.g. those without sidechains, and several of the metals, Pb, As, and Cd, were found to be significantly increased in the sediments of both lakes compared to their natural integrated deposits (ng cm^{-2}) and their background concentrations (μg g^{-1} or ng g^{-1} dry wt). Although the concentrations were generally much higher in Woods Lake, the total anthropogenic integrated depositions were about the same in both lakes for most of the metals and the 3 to 4 ring PAHs. The prime source of most of the 3 to 7 ring PAHs and trace elements measured is ascribed to anthropogenic combustion. Anthropogenically derived materials decreased in concentration with depth to baseline levels in sediment layers estimated by ^{137}Cs analyses to be $\sim$30 yr old, while biogenic or crustal derived species remained constant or fluctuated with core depth.

1. Introduction

It is stated that as a result of the combustion of coal and oil the United States annually discharges $\sim 5 \times 10^7$ tonne of S and NO_x into the atmosphere. These pollutants may be transformed into deleterious compounds which are atmospherically transported and returned to the earth great distances from their origin as components of rain or snow (EPA, 1979; Likens *et al.*, 1979). Among other pollutants released into the atmosphere during the combustion of fossil fuels are toxic and potentially toxic trace elements such as As, Cd, Hg, Se, Pb, and Tl (Natusch, 1978), and polycyclic aromatic hydrocarbons (PAHs) such as benzo(a)pyrene and benzo(a)anthracene which have been shown to be mutagenic or carcinogenic to many groups of biota including humans (National Academy of Sciences, 1972). The aim of this study is to determine to what degree these two classes of pollutants have been deposited in remote lakes known to be affected by acid rainfall and to provide data on historical changes in the levels of these substances in the environment.

2. Materials and Methods

2.1. LAKE DESCRIPTIONS

Both of the lakes investigated (Sagamore and Woods) are located in the Adirondack State Park on the western slope of the Adirondack Mountains. Several investigators have shown that this area receives acid rainfall (Cogbill, 1976; Galvin and Cline, 1978). Sagamore Lake is a 'brown water' lake due to high levels of humic materials while Woods Lake is generally clear. Both lakes are acidic throughout the year, although Sagamore is less so than Woods. A complete description of the lakes has been presented (Heit *et al.,* 1980), and the general physical characteristics are summarized in Table I.

TABLE I

General lake characteristics[a]

	Sagamore Lake	Woods Lake
Location	43°46′ N	43°53′ N
	74°38′ W	74°57′ W
Elevation (m)	586	615
Watershed area (km²)	49.7	2.1
Lake surface area (km²)	0.72	0.23
Surface: watershed ratio	1 : 69	1 : 9
Volume (10^6 m³)	7.5	0.8
Mean depth (m)	11.6	4.2
Maximum depth (m)	23	12
Flushing time (days)	65	180
pH range	5.0–6.4	4.7–5.1
Alkalinity range	−60–+20	−45–+45

[a] Hendrey (1979).

Sagamore and Woods are among the lakes presently being studied by various research organizations in a program known as the Integrated-Watershed Acidification Investigation (Hendrey, 1979). The aim of this program is to discover the mechanisms by which lakes become acidic as well as to describe the effects of acid precipitation on aquatic ecosystems.

2.2. SEDIMENT SAMPLING AND DESCRIPTION OF THE SEDIMENTS

Sediment cores were taken from each of the lakes during March 1978 with a 21 cm diameter coring device equipped with a sphincter core retainer (Burke, 1968). The method of core extrusion has been described elsewhere (Heit and Burke, 1977). The sediment throughout the cores from both lakes was dark brown in color, contained considerable amounts of water (92 to 87% for Woods and 92 to 81% for Sagamore Lake) and was loose and sticky, adhering to the extrusion tools. Electron microscopy revealed considerable amounts of pollen which may explain the stickiness. Also observed were large numbers of siliceous diatom skeletons.

2.3. SEDIMENT DATING

Measurements of ^{137}Cs and ^{210}Pb were used to determine the sedimentation rates within the lakes and hence the age of the sediment at any depth within the cores relative to the surface. The methodology for ^{137}Cs analysis is described in Ritchie *et al.* (1973) and for ^{210}Pb in Eakins and Morrison (1970).

It should be noted that there were some age discrepancies between the ^{210}Pb and ^{137}Cs methods of dating, particularly for depths greater than 10 cm. However, for reasons described elsewhere (Heit *et al.*, 1980), we believe the results of the ^{137}Cs dating procedure to be more realistic and hence only these values are presented here.

2.4. TRACE ELEMENT ANALYSES

The sediment samples were sent to Analytical Bio Chemistry Laboratories, Columbia, MO, where they were freeze dried and analyzed by various atomic absorption procedures for Ag, Al, As, Be, Cd, Cr, Cu, Hg, Ni, Pb, Se, Sn, Tl, V, and Zn. Special techniques were used for As and Se in which the matrix was modified through the addition of Ni. In cases where interferences occurred, As was converted to arsine gas prior to analyses. A complete description of the methods of digestion and analyses is given in Heit *et al.* (1980).

TABLE II

Polycyclic aromatic hydrocarbons measured

Compound	Abbreviation
Phenanthrene	Ph
Anthracene	A
C_1 isomers of phenanthrene and anthracene	$C_{15}H_{12}$
C_2 isomers of phenanthrene and anthracene	$C_{16}H_{14}$
Fluoranthene	F
Pyrene	P
C_1 isomers of pyrene and fluoranthene	$C_{17}H_{12}$
Benzo(a)fluorene	BaFl
Benzo(a)anthracene	BaA
Chrysene/triphenylene	Ch/Tr
Benzo(b)fluoranthene	BbF
Benzo(k)fluoranthene	BkF
Benzo(e)pyrene	BeP
Benzo(a)pyrene	BaP
Perylene	Per
Indeno(cd)pyrene	IndP
Dibenzo(*a, c/a, h*)anthracene	DBacahA
Benzo(ghi)perylene	BghiPer
Coronene	Cor
Dibenzo(*a, e*)pyrene	DBaeP
Retene	Retene
7,12a-dimethyl-1, 2, 3, 4, 4a, 11 12, 12a-octahydrochrysene	DMOHC
3, 4, 7-trimethyl-1, 2, 3, 4-tetrahydrochrysene	TMTHC

2.5. POLYCYCLIC AROMATIC HYDROCARBON ANALYSES

At the Environmental Measurements Laboratory (EML) the sediment was freeze dried and the PAHs removed by Soxhlet extraction and isolated by solvent partition and silica gel column-chromatography. Detection and quantification were performed by gas chromatography – mass spectrometry using multiple ion and flame ionization detectors and capillary gas chromatography using a flame ionization detector (Tan, 1979). The PAHs measured and the abbreviations used for these compounds are shown in Table II.

2.6. EML QUALITY CONTROL

For this study an EML homogenized sediment reference material, blind duplicates and commercially available reference materials were used to assess trace element precision and accuracy. The precision was excellent ($< 10\%$) for all of the elements except As, Cr, and Hg. For these three elements the precision was good ($\geqslant 10$ to 20%). The accuracy was excellent ($100 \pm 10\%$) for As, Cd, Cr, Hg, Ni, Pb, V, and Zn;

TABL]

Trace element concentration in Sagamor

Depth (cm)	Ag	Al*	As	Be	Cd	Cr	Cu
0–1	0.38 ± 0.11	22 ± 0.4	5.3 ± 0	5.1 ± 0.1	1.5 ± 0.1	8.0 ± 0.1	8.7 ± 0
1–2	0.14	20	4.3	4.7	1.7	8.0	7.1
2–3	0.24 ± 0.01	23 ± 0.1	3.8 ± 0.1	5.0 ± 0	1.7 ± 0.4	9.5 ± 1.0	7.0 ± 0.4
3–4	0.31	21	6.5	5.6	2.1	9.4	9.9
4–5	0.31	23	3.5	5.7	2.0	10	9.0
5–6	0.27 ± 0.07	22	3.5 ± 0.5	7.8 ± 0.4	1.8	9.0	7.5
6–7	0.44	21	2.9	7.2	2.1	9.7	6.4
7–8	0.18	19	2.8	4.6	1.1	10	5.6
8–9	0.72	19	2.4	4.4	1.1	11	4.8
9–10	0.14 ± 0.05	19 ± 0.3	3.5 ± 0.6	4.4 ± 0.6	1.0 ± 0.4	9.1 ± 0.3	5.6 ± 1.0
10–11	0.58	19	1.6	6.5	0.5	9.4	4.8
12–13	0.70	20	—	5.7	0.5	8.1	5.1
14–15	0.12	19	1.7	4.3	0.3	8.5	5.8
16–17	0.26	20	0.9	6.8	0.6	8.9	4.3
18–19	0.66	20	1.2	6.5	0.5	9.2	5.1
19–20	0.11 ± 0.03	21 ± 1	1.0 ± 0.1	4.0 ± 0.6	0.4 ± 0.3	10 ± 2	5.3 ± 0.3
20–21	0.58	19	1.4	6.3	0.7	10	4.2
29–31	0.11 ± 0.02	20	2.0	6.2 ± 0	0.5	11	4.5
31–33	0.20	24	1.1	4.0	0.5	12	5.1
39–41	0.14 ± 0.02	22 ± 1	1.4 ± 0.3	4.4 ± 0.3	0.6 ± 0.1	10 ± 0.6	5.0 ± 0.2
41–43	0.46	25	1.0	3.8	0.5	11	5.8
49–51	0.72	31	1.5	4.9	0.8	14	6.7
51–53	0.23	21	0.9	4.4	0.6	9.3	6.4
59–61	0.15 ± 0.02	21 ± 1	0.9 ± 0.4	5 ± 1	0.7 ± 0.1	10 ± 1	5.2 ± 0.5
61–63	0.55	22	1.1	4.1	0.8	11	4.8
69–71	0.20	22	0.7	4.6	0.5	10	5.6
71–73	0.07	20	1.2	4.1	0.6	12	7.0
77–79	0.12	22	1.0	4.4	0.6	11	5.7

[a] $Al = \mu g\, g^{-1} \times 10^3$.

good (100 ± 20%) for Cu and Se; and acceptable (100 ± 30%) for Ag. Poor accuracy was found for Be (37%) and Tl (145%). However, as the precision for these elements was excellent it was decided that the values should not be corrected and the data are presented as reported.

An EML homogenized reference sediment was used to assess the precision of the PAH analyses. Excellent precision was found for all of the compounds except fluoranthene, pyrene and dibenzo(a, c/a, h)pyrene where the precision was good. Since a reference material was not available for assessing PAH accuracy, an intercomparison of an EML reference sediment was conducted between EML, the Massachusetts Institute of Technology and Battelle-Columbus Laboratories. Our data showed good agreement with these laboratories. Subsequent re-analysis of this reference material has confirmed our values.

III

Lake as a function of depth ($\mu g\ g^{-1}$ dry wt)

Hg	Ni	Pb	Se	Sn	Tl	V	Zn
0.38 ± 0.01	6.4 ± 0.6	53 ± 4.2	1.6 ± 0.1	< 25	10 ± 0.8	105 ± 0	139 ± 1
0.69	8.5	47	1.5	"	9.5	102	126
0.41 ± 0.01	7.9 ± 0.6	47 ± 3	1.3 ± 0.2	"	9.7 ± 0.4	108 ± 3	148 ± 6
0.39	8.3	46	1.9	"	9.8	103	154
0.42	8.5	46	1.6	"	10	99	160
0.43	9.0 ± 0.7	41 ± 4	1.4 ± 0	"	8.6	142 ± 38	155
0.45	7.9	30	1.2	"	9.9	103	154
0.37	8.2	19	1.2	"	9.1	93	126
0.46	8.4	10	0.98	"	8.1	92	111
0.39 ± 0.04	6.3 ± 0.5	9.4 ± 0.5	0.85 ± 0.04	"	10 ± 1	82 ± 6	90 ± 20
0.46	6.0	8.5	—	"	9.7	100	85
0.38	6.5	4.4	0.44	"	9.7	90	77
0.30	5.1	4.0	0.65	"	9.1	79	68
0.34	6.4	3.7	0.65	"	9.8	90	70
0.31	6.5	2.3 ± 0.3	0.73	"	9.4	90	60 ± 20
0.24 ± 0.06	5.5	3.8	0.69 ± 0.23	"	10	90 ± 6	65
0.31	7.2	3.9 ± 0.1	0.60	"	10 ± 3	94	64
0.26	5.4 ± 2.2	4.0	0.68	"	9.3	97	84
0.17	6.1	4.2 ± 0.2	0.75	"	10	88	64 ± 1
0.24	8.4 ± 0.4	4.0	0.82 ± 0.11	"	11	104 ± 1	
0.24	7.1	4.2 ± 0.2	0.98	"	11 ± 1	92	98
0.35	8.8	—	0.69	"	9.5	112	74
0.42	7.0	4.0	0.78	"	11	96	71 ± 7
0.23 ± 0.01	6.7	4 ± 2	0.87 ± 0.01	"	12	94 ± 10	64
0.27	8.1	3.1	0.67	"	10	107	76
0.22	7.0	3.8	1.1	"	10	92	77
0.26	6.8	4.2	0.79	"	12	97	70
0.20	6.6	4.2	0.78	"	11	97	81

3 Results

3.1. TRACE ELEMENT CONCENTRATIONS

As shown in Figure 1 and Tables III and IV, As, Cd, Cu, Pb, Se, and Zn decreased in concentration with depth until reaching baseline levels, while Al and Tl remained at fairly constant levels throughout the length of the cores. Tin was not detected in the sediment from either lake ($< 25 \mu g g^{-1}$ dry wt). The trends of Ag, Be, Cr, Hg, Ni, and V were not as clear since differences were found between the two lakes. These elements decreased in concentration with depth until reaching baseline levels in Woods Lake, while remaining fairly constant in Sagamore Lake.

Silver, As, Be, Cd, Cu, Hg, Ni, Pb, Se, V, and Zn were generally higher in concentration in the surface (0 to 4 cm) of Woods Lake as compared to Sagamore Lake. Below this depth the concentrations were either about the same or slightly higher in one lake or the other depending upon the element. Chromium and Tl were higher in concentration in Sagamore than in Woods Lake. Since these two elements remained constant throughout the length of the Sagamore core, we believe that this may indicate a higher natural background in Sagamore Lake.

For the elements which decreased in concentration with depth, so called baseline or constant background levels (Table V) generally occurred at depths greater than 9 to 11 cm in Sagamore Lake and 6 to 9 cm in Woods Lake. These depths correspond to

TABLE

Trace element concentration in Woods Lake

Depth (cm)	Ag	Al*	As	Be	Cd	Cr	Cu
0–1	0.56 ± 0.05	27 ± 1	13 ± 4	5.7 ± 0.2	2.6 ± 0	7.6 ± 0.1	15 ± 1
1–2	0.64	27	10	5.5	2.8	7.6	13
2–3	0.56 ± 0.01	26 ± 2	7 ± 2	5.8 ± 0.2	2.2 ± 0.2	6.5 ± 0.5	12 ± 1
3–4	0.51	27	8.2	5.4	1.4	3.1	10
4–5	0.31	30	6.4	5.1	1.0	6.3	7.6
5–6	0.21	36	3.0	5.4	0.55	5.9	7.4
6–7	0.26	23	2.6	—	0.72	4.0	4.0
7–8	0.13	26	1.9	4.3	0.58	4.6	7.1
8–9	0.34	24	2.0	4.1	0.59	2.4	5.2
9–10	0.21 ± 0.04	24 ± 1	1.3 ± 0.1	4.2 ± 0.1	0.4 ± 0.2	3.4 ± 0.5	5.9 ± 0.4
10–11	0.33	26	1.8	4.5	0.62	3.3	5.9
12–13	0.25	23	2.7	4.2	0.85	5.3	6.3
16–17	0.48	25	1.0	3.9	0.27	4.4	7.3
18–19	0.30	29	1.5	4.7	0.64	6.8	7.1
20–21	0.19	26	1.1	4.1	0.58	5.8	5.8
40–42	0.25	20	1.2	4.1	0.56	4.5	6.8
50–52	0.19	23	1.4	4.2	0.58	3.8	6.4
58–60	0.28	22	0.8	3.3	0.27	4.3	6.0
60–62	0.31	26	0.9	4.0	0.55	4.5	5.9
70–72	0.84	32	0.9	4.1	0.54	5.1	5.4
80–82	0.49	25	1.3	4.3	0.57	3.0	6.9

[a] $Al = \mu g g^{-1} \times 10^3$.

sediment deposited in the lakes about 30 yr ago according to the ^{137}Cs method of dating (see Table VI). It should be noted that the terms baseline, natural and background are used synonymously, and refer to the fairly constant levels reached in the cores below these depths. It is possible that lower levels would be reached in cores of greater length.

## 3.2. POLYCYCLIC AROMATIC HYDROCARBON CONCENTRATIONS

All of the 3 to 7 ring parental PAHs with the exception of perylene decreased in concentration with depth in the cores taken from both lakes (see Figure 2 and Tables VII and VIII). Concentration ranges (ng g^{-1} dry wt) for the different isomers of the methyl ($C_{15}H_{12}$) and dimethyl ($C_{16}H_{12}$) phenanthrenes and methyl ($C_{17}H_{12}$) fluoranthenes and pyrenes were measured only in the 0 to 4 cm layers of the sediment. Retene, octahydrochrysene and tetrahydrochrysene fluctuated in concentration with depth or remained fairly constant throughout the length of the cores (see Figure 2).

With the exception of perylene, all of the parental PAHs were three to four times higher in concentration in the surface sediments of Woods Lake than in the surface sediments of Sagamore Lake. It is important to note that the baseline levels (Table V) for the PAHs occurred at approximately the same depths as the baselines for the metals which decreased in concentration, i.e., sediment deposited $\sim$30 yr ago according to the ^{137}Cs method of dating. In addition, the surface concentrations of many of

IV

as a function of depth (µg g^{-1} dry wt)

Hg	Ni	Pb	Se	Sn	Tl	V	Zn
0.46 ± 0.11	11 ± 0.11	138 ± 8	4.6	< 25	5.8 ± 0.1	114 ± 1	345 ± 8
0.41	9.7	128	4.5	"	6.2	116	327
0.35 ± 0.02	8.9 ± 0.9	114 ± 4	3.1 ± 0.1	"	6.3 ± 0.8	120 ± 10	300 ± 30
0.34	8.1	82	3.7	"	6.9	126	239
0.35	4.9	57	1.3	"	6.3	114	140
0.28	5.0	31	2.0	"	5.5	131	129
0.32	4.9	12	2.8	"	4.6	112	145
0.20	5.1	16	1.9	"	—	102	48
0.29	4.7	11	1.4	"	5.3	102	38
0.20 ± 0.08	4.9 ± 0.4	5.1 ± 1.1	1.6 ± 0.1	"	6.3 ± 1.2	103 ± 2	50 ± 2
0.29	5.3	5.4	1.3	"	6.2	105	71
0.27	—	5.6	1.3	"	5.6	102	83
0.31	4.6	5.0	1.2	"	3.8	99	69
0.28	6.0	4.9	—	"	7.0	116	64
0.38	5.1	4.9	1.5	"	4.6	99	50
0.56	5.6	4.8	1.9	"	4.7	96	71
0.24	5.6	4.7	1.9	"	7.5	92	48
0.48	4.1	3.1	1.5	"	4.6	75	49
0.29	5.8	4.2	1.3	"	5.0	90	68
0.20	5.5	5.3	2.4	"	5.9	97	59
0.23	5.3	5.8	1.6	"	5.7	99	54
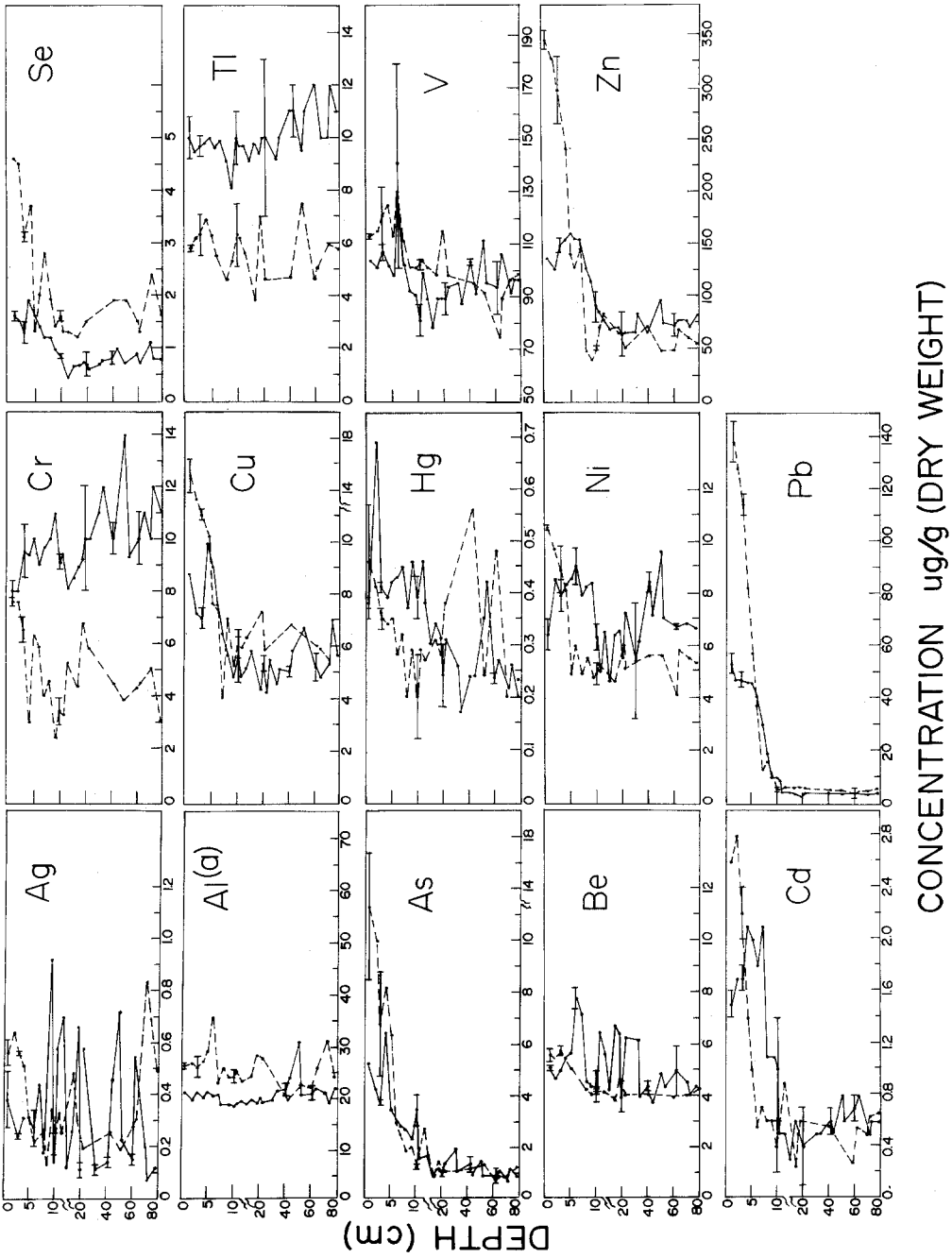
Fig. 1. Trace element concentration with sediment depth in Sagamore and Woods Lakes. Symbols: (——) Sagamore Lake, (— —) Woods Lake, (a) Al = µg/g × 10³.
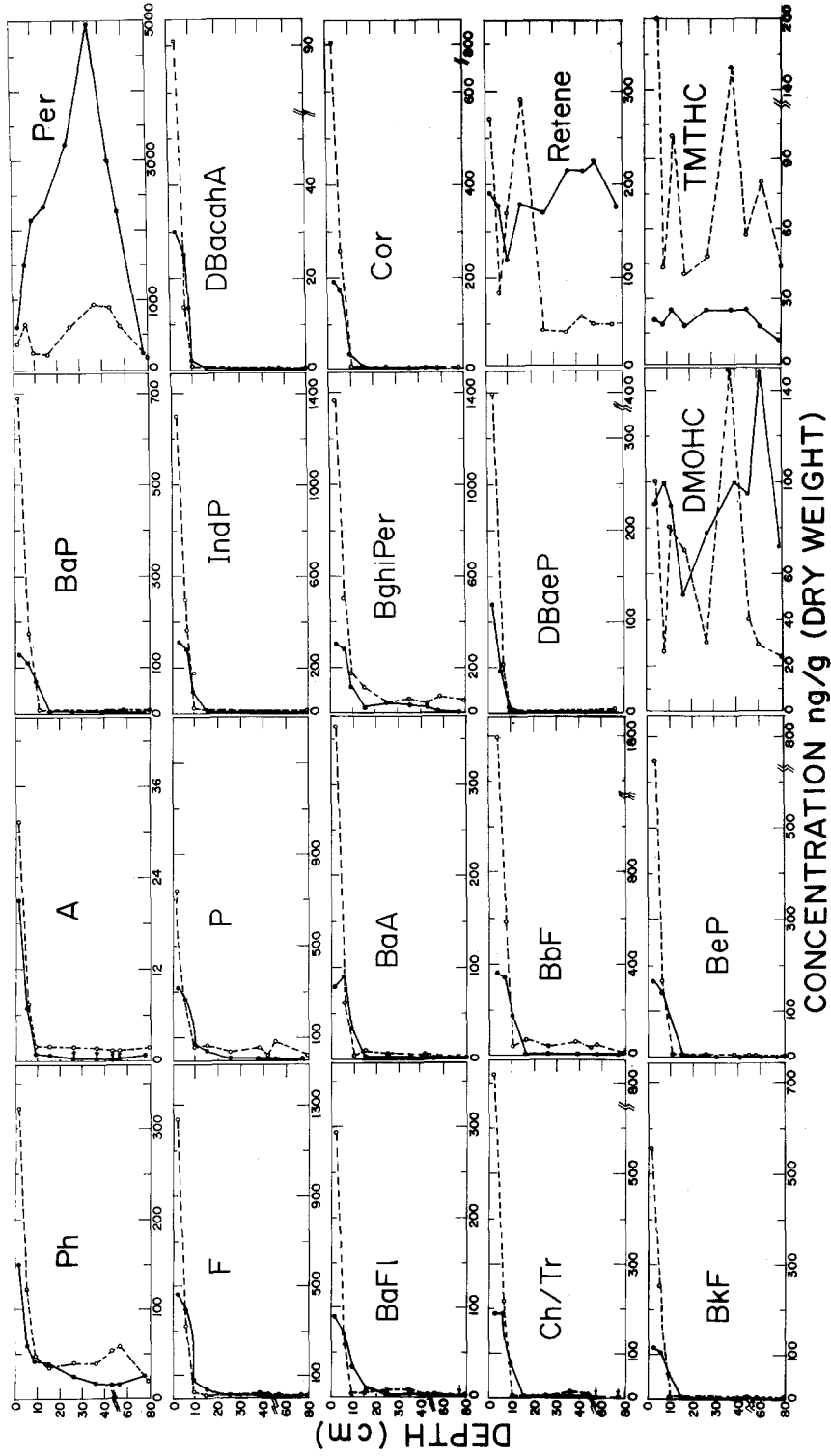
Fig. 2. PAH concentrations with sediment depth in Sagamore and Woods Lakes. Symbols: (———) Sagamore Lake, (– – –) Woods Lake.

TABLE V

Background average concentrations of trace elements
and polycyclic aromatic hydrocarbons*

Element	Sagamore Lake (μg g^{-1} dry wt)	Woods Lake (μg g^{-1} dry wt)
Ag	0.3 ± 0.2	0.3 ± 0.2
Al	22000 ± 3000	22000 ± 3000
As	1.2 ± 0.3	1.4 ± 0.5
Be	5 ± 1	4.1 ± 0.3
Cd	0.6 ± 0.1	0.5 ± 0.2
Cr	10 ± 2	4 ± 1
Cu	5.4 ± 0.8	6.3 ± 0.7
Hg	0.28 ± 0.07	0.3 ± 0.1
Ni	7 ± 1	5.2 ± 0.5
Pb	3.9 ± 0.5	5.4 ± 1.8
Se	0.8 ± 0.2	1.6 ± 0.3
Sn	< 25	< 25
Tl	10 ± 1	6 ± 1
V	95 ± 8	98 ± 9
Zn	74 ± 9	59 ± 13

PAH	Sagamore Lake (ng g^{-1} dry wt)	Woods Lake (ng g^{-1} dry wt)
Ph	20 ± 6	40 ± 10
A	< 2	3 ± 1
F	10 ± 6	15 ± 5
P	8 ± 4	50 ± 30
BaFl	< 4	5 ± 2
B(a)A	< 1	2 ± 2
Ch/Tr	< 4	6 ± 5
BbF	4 ± 2	40 ± 20
BkF	2 ± 1	7 ± 2
BeP	< 2	7 ± 3
BaP	< 2	4 ± 3
Per	2600 ± 1400	500 ± 300
IndP	3 ± 1	6 ± 6
DBacahP	< 4	< 3
BghiPer	29 ± 16	83 ± 47
Cor	< 3	< 3
DBaeP	< 3	< 3

* Background levels determined from the average con-
centrations of trace elements and PAHs at depths greater
than 9–11 cm in Sagamore and greater than 6–9 cm in
Woods Lake.

these compounds, especially in Woods Lake, approach or exceed levels reported for
sediments taken from much more urban or industrialized areas such as Lake Con-
stance, West Germany (Müller *et al.,* 1977), Buzzards Bay, MA (Hites *et al.,* 1977;
Heit and Tan, 1979) and Lake Griefensee, Switzerland (Geiger and Schaffner, 1978);
while the baseline levels were similar to those reported for pristine areas such as
northern Ontario (Brown and Starnes, 1978).

TABLE VI

Time frame for Sagamore and Woods Lake
by ^{137}Cs dating[a]

Sediment depth (cm)	Approximate age relative to surface	
	Sagamore[b]	Woods[c]
1	1976	1975
2	1973	1971
3	1971	1968
4	1968	1965
5	1966	1962
6	1963	1958
7	1961	1955
8	1958	1952
9	1956	—
10	1953	—

[a] All data from Heit et al. (1980).
[b] Sedimentation rate Sagamore Lake
$\simeq 0.4$ cm yr^{-1} at 0–10 cm.
[c] Sedimentation rate Woods Lake
$\simeq 0.3$ cm yr^{-1} at 0–8 cm.

TABLE VII

Depth distribution of PAHs in Sagamore Lake sediment (ng g^{-1} dry wt)

PAH	Depth (cm)								
	0–4	4–8	8–11	12–17	25–27	35–37	43–47	51–55	71–75
Ph	154	67	42	38	25	15	15	17	27
A	21	7	2	2	1	1	1	1	2
$C_{15}H_{12}$	40–70								
$C_{16}H_{14}$	25–70								
F	463	399	81	38	14	15	5	3	11
P	320	263	66	42	9	9	5	3	14
$C_{17}H_{12}$	15–40								
BaFl	91	70	34	10	2	4	3	3	1
BaA	78	89	33	1	1	1	1	1	1
Ch/Tr ·	191	191	77	7	6	6	2	1	1
BbF	358	339	172	6	6	6	2	2	1
BkF	115	106	57	2	2	2	1	1	1
BeP	165	140	87	3	2	3	1	1	1
BaP	128	120	70	2	1	1	1	1	1
Per	605	1550	2123	2341	3205	4985	2988	2260	207
IndP	315	279	99	3	3	4	2	2	2
DBacahA	30	25	4	2	2	2	2	2	2
BghiPer	303	280	106	26	48	38	39	20	5
Cor	188	174	39	< 3	< 3	< 3	< 3	< 3	< 3
DBaeP	115	44	6	< 3	< 3	< 3	< 3	< 3	< 3
Retene	191	176	118	179	169	215	215	224	174
DMOHC	90	99	89	50	78	100	95	149	172
TMTHC	20	18	25	18	25	25	25	18	12

TABLE VIII

PAHs in Woods Lake sediment (ng g^{-1} dry wt)

PAH	Depth (cm)								
	0–4	4–8	8–11	12–17	24–26	34–38	42–44	50–54	80–84
Ph	324	126	48	33	38	36	55	59	24
A	32	8	3	3	3	3	2	2	3
$C_{15}H_{12}$	60–100								
$C_{16}H_{14}$	45–60								
F	1236	319	25	11	15	25	13	15	11
P	934	286	56	67	37	60	21	89	16
$C_{17}H_{12}$	6–30								
BaFl	294	58	4	4	7	8	5	5	2
BaA	362	62	2	7	2	3	1	1	1
Ch/Tr	888	219	4	5	6	17	8	3	2
BbF	1784	575	33	72	37	55	31	48	8
BkF	558	179	11	4	7	6	5	8	8
BeP	768	254	8	5	9	10	6	10	3
BaP	690	175	3	2	2	3	11	3	1
Per	280	485	251	230	622	952	813	767	116
IndP	1294	358	20	4	3	5	< 2	8	2
DBacahA	92	17	< 2	3	< 2	< 2	< 2	< 2	< 2
BghiPer	1356	498	177	111	44	65	46	76	59
Cor	801	258	< 3	< 3	< 3	< 3	< 3	< 3	< 3
DBaeP	394	58	< 3	< 3	< 3	< 3	< 3	< 3	< 3
Retene	270	83	144	289	42	40	56	49	53
DMOHC	100	26	80	70	30	150	40	29	23
TMTHC	200	43	100	40	48	150	57	80	44

4. Discussion

4.1. CONCENTRATION DIFFERENCES BETWEEN THE LAKES

Differences in PAH and metal concentration between the two lakes appear to be due to the higher rate of sedimentation in Sagamore Lake ($\sim$0.4 cm yr^{-1}) as compared to Woods Lake ($\sim$0.3 cm yr^{-1}), Table VI. The resulting greater amounts of natural materials deposited in the Sagamore sediment would be expected to dilute the anthropogenically deposited metals and PAHs to a greater degree than for Woods Lake, leading to relatively higher concentrations in the latter lake. Hendrey (1980) has in fact shown that the top 1 cm of Sagamore Lake contains significantly more organic C than Woods Lake which supports this dilution hypothesis.

Comparing the total anthropogenic deposition of the metals and PAHs on a per unit area basis (μg cm^{-2} or ng cm^{-2}) eliminates the biases from such dilution effects that occur when comparing concentrations (μg g^{-1} or ng g^{-1}). The total anthropogenic deposition of an element or compound per unit area is calculated according to the following formula:

$$Q = \sum_{i=1}^{n} (X_i)(D_i)(C_i)$$

where

Q = the total integrated deposition,

X_i = the concentration of substance X in the ith section of sediment minus the average baseline concentration of that substance ($ng\ g^{-1}$ dry wt),

D_i = the density of the ith section of sediment ($g\ cm^{-3}$ dry wt), and

C_i = the thickness of the ith incremental slice of the sediment (cm).

By this method of comparison, the agreement of the total integrated anthropogenic trace element deposition between the two lakes was found to be very good (see Table IX). For example, the total anthropogenic As deposition was found to be 3600 $ng\ cm^{-2}$ for both lakes although the concentrations in the 0 to 1 cm sections of the two lakes were quite different, 13 000 $ng\ g^{-1}$ and 5300 $ng\ g^{-1}$, respectively. Other examples include: (a) Pb, 40 000 $ng\ cm^{-2}$ deposited in Sagamore Lake and 48 000 $ng\ cm^{-2}$ deposited in Woods Lake; (b) Se, 700 $ng\ cm^{-2}$ deposited in Sagamore Lake and 800 $ng\ cm^{-2}$ deposited in Woods Lake; and (c) Hg, 300 $ng\ cm^{-2}$ deposited in both lakes. In fact, the ratios of the anthropogenic deposition between the lakes (Table IX) were ∼1 for almost all of the elements, indicating that approximately the same amounts of these materials were deposited in both lakes even though they are some 30 km apart.

When the same technique was applied to the parental PAHs, the agreement in the total deposition was also found to be good for all of the 3 and 4 ring compounds except chrysene/triphenylene. The ratio of the anthropogenic deposition between Woods and Sagamore Lake for these compounds was again ∼1 (Table IX). However, the 4 ring chrysene/triphenylene and all of the 5, 6, and 7 ring compounds with the exception of dibenzo ($a, c/a, h$)anthracene had ratios of ∼2. The reason(s) for these differences are not clear although we do not believe that they are due to selective anthropogenic deposition of higher molecular weight PAHs into Woods Lake. One possible explanation for this phenomenon may be differences in the amount of naturally occurring humic organic materials entering into the two lakes. Since Sagamore Lake contains high levels of humic materials which are known to bind with particulates (Braunstein *et al.*, 1977), atmospherically deposited substances may have a longer residence time in the water column of Sagamore Lake before settling into the sediment as compared to Woods Lake. This longer residence time may result in some PAHs being degraded to a greater extent in Sagamore Lake as compared to Woods Lake since it has been established that these compounds are susceptible in varying degrees to photolysis (National Academy of Sciences, 1972). Therefore, the elevated ratios for the PAHs above chrysene/triphenylene in ring size may be a reflection of their relative stabilities.

4.2. HISTORIC TRENDS

The concept of a cultural enrichment factor (CEF) may be used to indicate whether the rate of entry of suspected pollutants into a body of water has increased in relation to an earlier period or baseline value (Robins and Edgington, 1977). The CEF is

TABLE IX

Integrated Anthropogenic and Natural Deposition (ng cm^{-2})

Element	Sagamore[a]			Woods[a]			Ratio of anthropogenic input Woods/Sagamore[b]
	Anthropogenic	Natural or baseline	Anthro/natural ratio	Anthropogenic	Natural or baseline	Anthro/natural ratio	
Ag	60	500	0.1	30	210	0.1	1
As	3600	1900	1.9	3600	910	4.0	1
Be	1000	7800	0.1	800	2700	0.3	1
Cd	1400	900	1.5	700	350	2.0	1
Cr	~0	16000	<0.01	600	2900	0.2	—
Cu	2000	8600	0.2	2100	4100	0.5	1
Hg	300	440	0.7	300	200	1.5	1
Ni	1200	11000	0.1	1400	3400	0.4	1
Pb	40000	6200	6.4	48000	3500	14	1
Se	700	1200	0.6	800	1000	0.8	1
Tl	~0	16000	<0.1	~0	3600	<0.1	1
V	12000	143000	0.1	14000	64000	0.2	1
Zn	82000	118000	0.7	104000	38000	2.7	1

a Deposition integrated to 9–11 cm in Sagamore and 6–9 cm in Woods Lake.
b Ratios rounded to the nearest whole number.

TABLE IX (Cont'd)

PAH	Sagamore			Woods			Ratio of anthropogenic input Woods/Sagamore[a]
	Anthropogenic	Natural or baseline	Anthro/natural ratio	Anthropogenic	Natural or baseline	Anthro/natural ratio	
Ph	120	30	4	140	40	4	1
A	14	<1	≥14	13	2	7	1
F	500	25	20	550	15	35	1
P	360	20	18	420	40	10	1
BaFl	100	<10	≥10	130	4	30	1
BaA	110	<1	≥100	150	2	75	1
Ch/Tr	250	<3	≥80	400	5	80	2
BbF	450	5	90	830	30	30	2
BkF	140	2	70	260	5	50	2
BeP	200	<5	≥40	370	5	70	2
BaP	170	<5	≥30	290	3	100	2
IndenP	360	3	120	600	4	150	2
DBacahA	30	<1	≥30	30	2	15	1
BghiPer	320	50	≥6	660	60	11	2
Cor	210	<3	≥70	380	<2	≥190	2
DBaeP	80	<5	≥16	160	2	≥80	2

[a] Ratios rounded to the nearest whole number.

defined as the ratio of the concentration of the suspected material in recent sediment (usually the layer(s) at which the maximum concentration occurs) to the constant baseline concentration of the material (see Table V). These values may be interpreted as follows: a CEF of $\sim$1 indicates no significant anthropogenic input, whereas a CEF of 1 to 2 is taken as evidence of some anthropogenic contribution. CEFs > 2 correspond to increasingly greater amounts of anthropogenic input.

The CEFs calculated for this study (Table X) indicate that compared to its baseline input Pb had the greatest anthropogenic deposit of any of the trace elements entering

TABLE X

Cultural enrichment factors CEF[a]

Element	Sagamore Lake	Woods Lake
Ag	1.2	1.7
Al	1.1	1.2
As	4.4	9.0
Be	1.0	1.4
Cd	2.6	4.8
Cr	0.8	1.7
Cu	1.6	2.4
Hg	1.4	1.5
Ni	0.9	2.0
Pb	14	26
Se	2.1	2.8
Sn	—	—
Tl	1.0	1.1
V	1.1	1.2
Zn	1.9	5.9
PAH		
Ph	6.4	7.9
A	$\geq$11	11
F	33	82
P	27	19
BaF	$\geq$23	59
BaA	$\geq$78	180
Ch/Tr	$\geq$48	150
BbF	90	44
BkP	58	80
BeP	$\geq$83	110
BaP	$\geq$64	170
Per	0.2	0.6
IndP	110	220
DBacahA	$\geq$8	$\geq$31
BghiPer	10	16
Cor	$\geq$63	$\geq$270
DBaeP	$\geq$38	$\geq$130

[a] CEF = concentration of element or compound in recent sediment average baseline concentration of element or compound (from Table V).

the two lakes i.e. the Pb CEF was 26 for Woods Lake and 14 for Sagamore Lake. Other elements which appear to have mainly an anthropogenic origin include: As, CEF 4 to 9; Cd, CEF 3 to 5; Se, CEF 2 to 3; Zn, CEF 2 to 6; and Cu, CEF 2 for both lakes. This interpretation is also supported by comparing the ratios of the total integrated anthropogenic deposition per unit area for each element to their respective integrated natural deposition (see Table IX). These ratios again indicate that the greatest anthropogenic input of any metal into these lakes was Pb, with ratios of 6 for Sagamore Lake and 14 for Woods Lake, followed by As and Cd, with ratios of between 2 and 4. It is also important to note that both the CEFs and the deposition ratios indicate that several of the element including the toxic elements Be, Hg, and Tl (Braunstein *et al.,* 1977) have not entered the sediments of these lakes in amounts significantly different from their natural levels of input.

It should be pointed out that although the anthropogenic input of Pb exceeded its natural input into the lakes to a greater degree than any of the metals measured, Zn had a higher total anthropogenic deposition, 82 000 ng cm^{-2} versus 40 000 ng cm^{-2} Pb in Sagamore, and 104 000 ng cm^{-2} Zn versus 48 000 ng cm^{-2} Pb in Woods (Table IX). Interestingly, the element with the third highest anthropogenic input was V, an element associated with combustion of high S containing fuels (Heit, 1977). The V anthropogenic deposition ranged from 12 000 ng cm^{-2} in Sagamore Lake to 14 000 ng cm^{-2} in Woods Lake. .However, the natural V deposition was found to greatly exceed its anthropogenic input, 64 000 ng cm^{-2} for Woods Lake and 143 000 ng cm^{-2} for Sagamore Lake (Table IX).

The CEFs and the ratios of the total anthropogenic/natural deposition also indicate that all of the parental PAH compounds, with the exception of perylene, have entered these lakes primarily through anthropogenic rather than natural processes. For example, the CEFs ranged from 6 to 8 for phenanthrene in both lakes to > 270 for coronene in Woods Lake. The carcinogenic BaA and BaP ranged from > 60 in Sagamore to > 170 in Woods. These high CEFs also indicate that the parental PAHs have entered these lakes at significantly greater rates than the metals, with the possible exception of Pb. This is the first data that we are aware of which indicates that anthropogenically derived PAHs may be entering remote environments at rates higher than those of toxic or potentially toxic trace elements.

4.3. POLLUTANT FORMATION

We are convinced that the main source of the excess (above natural background levels) trace elements and polynuclear aromatic hydrocarbons is high temperature combustion. We base this hypothesis on the following evidence.

4.3.1. *Parental PAH/Alkyl homologue ratios*

Both lakes exhibited PAH patterns in their surface sediment (0 to 4 cm) in which the parental PAHs predominated over their alkylated homologues (Table VII and VIII). For example, the combined level of parental fluoranthene and pyrene in Sagamore Lake was 783 ng g^{-1}, while their alkylated C$_{17}$H$_{12}$ homologues ranged in concentration

from 15 to 40 ng g^{-1}. In Woods Lake the concentrations of total fluoranthene and pyrene was 2170 ng g^{-1} compared to their $C_{17}H_{12}$ homologues which ranged from 6 to 30 ng g^{-1}. Similar, although less dramatic results were also found for phenanthrene/anthracene when compared to their $C_{15}H_{12}$ and $C_{16}H_{12}$ alkyl homologues. Ratios in which the parental/alkylated homologues are > 1 have been shown to be indicators of high temperature combustion derived PAHs, whereas ratios that are < 1 has been shown to be associated with PAHs produced by diagenic or biogenic processes (Hites *et al.*, 1977; Heit and Tan, 1979; Brown and Starnes, 1978). Clearly the ratios for these lakes fall into the former category indicating a high temperature combustion origin for the parental PAHs.

4.3.2. *Biogenic PAH distribution*

All of the parental PAHs, with the exception of perylene, decreased simultaneously in both lakes to their respective baseline levels. Perylene, although a parental PAH, is believed to be produced primarily by biogenesis in sediments from an as yet unknown precursor(s) under reducing conditions (Wakem *et al.*, 1979, 1980). Since none of the other parental PAHs followed the distribution of perylene in the sediment, i.e. rising in concentration with depth and then falling in concentration in the deeper sediment (see Figure 2), we interpret this as evidence that the parental PAHs in these lakes have an origin different from perylene. In addition, retene, DMOHC and TMTHC, derived from terpene precursors in higher plants (Laflamme and Hites, 1978; Wakem *et al.*, 1980), also had different sediment distribution patterns than the parental PAHs in that they remained at fairly constant levels or fluctuated up and down in concentration throughout the length of the cores (see Figure 2). We conclude that the excess parental PAHs were not produced by known biogenetic processes.

4.3.3. *Correlations*

The use of the correlation coefficient, r, may serve as an indicator of whether trace substances have a common origin (Heit *et al.*, 1981). Here the concentrations of all of the trace elements and PAHs in the sediment cores were correlated with Pb and fluoranthene. These substances were chosen because they have been shown to be associated with various types of combustion related anthropogenic pollution (Heit, 1977; Braunstein *et al.*, 1977) and were among the substances with the highest concentrations measured in the lake sediments.

At the 1% level of significance (Pearson and Hartley, 1954), both Pb and fluoranthene correlated with As, Cd, Se, and Zn and all of the parental PAHs with the exception of perylene in Sagamore Lake. In addition, at this level of significance Pb correlated with Cu, Hg, Se, and V. For Woods Lake, at the 1% level of significance, Pb and fluoranthene correlated with As, Be, Cd, Cu, Ni, Se, V, and Zn and all of the parental PAHs except perylene. Lead also correlated with Cr and Hg. As expected, fluoranthene did not correlate with the biogenetically derived retene, DMOHC or TMTHC in either lake. A complete listing of all of the correlation values is given in Heit *et al.* (1981).

These data suggest that all of the parental PAHs (except perylene) and the elements As, Cd, Pb, Se, and Zn have a similar origin in both lakes. Although the data are not as clear, it also suggests that Be, Cr, Cu, Hg, Ni, and V may also be considered in this group based upon their significant correlations with Pb or fluoranthene in Woods Lake. Since both fluoranthene and Pb are primarily released into the environment by high temperature combustion processes, it would appear that these elements also have a similar origin. Interestingly, Tl, an element predicted to be released by industrial combustion processes (Heit, 1977; Braunstein *et al.*, 1977), did not correlate with Pb or fluoranthene and appeared to have no measurable anthropogenic deposition in either lake (Heit *et al.*, 1980).

4.4. SPECIFIC COMBUSTION SOURCES

Combustion origins for these pollutants are limited to the following sources: forest fires; input from local anthropogenic sources, such as industry, space heating, automobile and street runoff; and long distance atmospheric transport of anthropogenically derived pollutants into the region.

4.4.1. *Forest fires*

Although it is quite difficult to evaluate whether the source of parental PAHs and metals is forest fires or anthropogenic combustion, we believe that natural fires may be ruled out as major sources of these pollutants in the two lakes. This statement is based on the distribution of the parental PAHs in the sediment in which these compounds decreased with depth in the cores until reaching fairly constant levels which were much lower in concentration than in the surface layers. If forest fires were important sources, one would expect both the concentrations and natural fluxes of parental PAHs in core segments corresponding to the inputs of past fires to reach levels that approach the surface values. Also, since forest fires can be expected to have occurred from time to time in both basins one would expect intermittent periods of high levels of PAHs throughout the length of both cores; this however does not occur (Heit *et al.*, 1980). For example, a major forest fire is said to have occurred about 80 yr ago in the Woods Lake basin (Marleau, 1980), but our data show the levels of the parental PAHs to be significantly lower at the corresponding depth in the Woods core, 11 cm by ^{137}Cs dating (Heit *et al.*, 1980), than at the surface (Table VIII).

Although several metals, including Pb, have been shown to be mobilized and transported by forest fires (Young and Jan, 1977), we believe that the same arguments as apply to the PAHs apply to the trace elements; the trace elements remain at fairly constant low levels in the deeper sediments as compared to the elevated levels at the surface.

4.4.2. *Local versus long distance atmospheric transport*

We conclude that the inputs from local anthropogenic sources i.e. heating, automobiles and street runoff are also unimportant sources of PAH and trace elements compared to long distance transport. This conclusion is based on the facts that Woods

and Sagamore Lakes are remote, have remained relatively undisturbed by man for several decades and are away from any significant heating, industrial or local automotive insults. Most importantly, if the two lakes were receiving pollutants transported into this area on a regional basis, one would expect the total integrated anthropogenic input for these materials to be similar in both Sagamore and Woods. Whereas if local pollution sources were important factors, then one would expect the PAH and trace element inputs into the two lakes to be different due to various local factors. However, we have already shown that the total anthropogenic inputs (ng cm^{-2}) for the majority of the trace elements and 3-4 ring PAHs were approximately the same for both lakes (Table IX). Thus, our data support a regional deposition of atmospherically transported pollutants which appears to overshadow local anthropogenic inputs. It should be noted that the metals which showed the greatest total anthropogenic depositions (Table IX) i.e. Zn, Pb, V, and As have all been shown to be present in the atmospheric emissions of fossil fuel powered electric generating stations and other industry related processes (Heit, 1977; Braunstein *et al.*, 1977), none of which are located in the area.

4.4.3. *Industrial sources of pollution*

Industrial areas outside of the Adirondack park have been suggested as the major sources of pollution in the Adirondack region (Altshuller and McBean, 1978; Galvin *et al.*, 1978; EPA, 1979). One of the sites often mentioned is the Sudbury smelter complex located in Ontario, Canada. However, our data suggest that at least for the trace elements the influence of this area is small or masked by the inputs from other sources. We reach this conclusion based upon the relative total anthropogenic deposits of Ni and Cu, the two major elements refined at the smelter (Hutchinson and Whitby, 1974), which are small compared to Zn, Pb, and V, (Table IX). It should be noted that Zn, V, and Ni have approximately the same relative volatilities during combustion, thus the low levels of Ni in the sediments appears not to be due to this factor (Vaughan *et al.*, 1975). In fact, if one compares the average concentrations of these elements with their levels in the earth's crust (Clarke value) and normalized to Al, the resulting ratio called the enrichment factor, E, indicates that these two elements are naturally depleted in the lake sediment, not enhanced i.e. $E = 0.3$ to 0.5 for Ni and $E = 0.3$ to 0.8 for Cu (Table XI). One would expect the ratio E to be > 1 if there were significant non-geological inputs into the lake as is the case for Pb where E ranged from 2 to 15 above 11 cm in Sagamore Lake and $E = 3$ to 34 above 8 cm in Woods Lake. Below these depths $E = \sim 1$ for both lakes indicating the main source of the Pb to be crustal.

Interestingly, the level of Zn in the soil around Sudbury has been shown to be approximately constant from distances of 1.1 to 50 km from the complex (Hutchinson and Whitby, 1974) indicating nondistinguishable or insignificant Zn pollution from the smelter. However, we have found that Zn had the highest total anthropogenic deposition of any of the metals deposited into the lakes (Table IX) supporting our conclusion that the anthropogenic trace element contribution of Sudbury to these lakes is small.

TABLE XI

Enrichment factors (E) for Pb, Cu and Ni

Depth (cm)	Pb		Cu		Ni	
	Sagamore	Woods	Sagamore	Woods	Sagamore	Woods
0–1	15	34	0.6	0.8	0.3	0.4
1–2	15	31	0.5	0.7	0.5	0.4
2–3	14	28	0.5	0.7	0.4	0.4
3–4	14	20	0.6	0.4	0.4	0.3
4–5	13	13	0.5	0.3	0.4	0.2
5–6	13	6	0.5	0.3	0.5	0.2
6–7	9	3	0.5	0.3	0.4	0.2
7–8	7	4	0.4	0.4	0.5	0.2
8–9	3	3	0.4	0.3	0.5	0.2
9–10	3	1	0.4	0.2	0.4	0.2
10–11	2	1	0.4	0.3	0.3	0.2
> 11	1	1–2	0.3–0.5	0.3–0.5	0.3–0.4	0.2

E $= (Cx/CAl)/(Crx/CrAl)$
Cx   = concentration of the element measured in sediment
CAl   = concentration of Al in the sediment
Crx   = concentration of the element in the earth's crust
CrAl = concentration of Al in the earth's crust

Data in the literature also suggest that the lakes in the Adirondacks have been affected by pollutants emitted from the combustion of fossil fuels from other industrialized areas, most notably the Upper Ohio Valley – Great Lakes region (Altshuller and McBean, 1978). If meteorological conditions do in fact favor the transport of industrial pollutants generated in this region into the Adirondacks, then one may also expect to find similar ratios of these materials deposited in other areas downwind of the sites of origin. Unfortunately insufficient information is available for comparison. Thus at this time our data cannot prove or disprove that the pollutants originated primarily in the Great Lakes – Ohio Valley region.

Since baseline concentrations were reached for practically all of the anthropogenically produced PAHs and trace elements at depths slightly above or below (± 2 cm) the level at which the nuclear weapons produced ^{137}Cs was no longer detected in the sediments of the two lakes, we conclude that these pollutants began to enter these lakes in significant amounts at about the same time as the radioactive nuclide i.e. ~1950. This estimate agrees well with Galloway and Likens (1979) data for elements deposited in Woodhull Lake ~30 km south of Woods Lake, where a sediment core was also dated by the ^{137}Cs technique. The introduction of pollutants into the lakes at this time is not unrealistic since tall power plant smoke stacks, which have been associated with long distance atmospheric transport, were first built in the early 1950's (Thomas et al., 1963) reaching their peak heights in the late 1960's – early 1970's (Smith and Frankenberg, 1975). It should be noted that data have recently been published which show that a dramatic acceleration in the anthropogenic Pb flux into

Belgium has also taken place since the late 1950's (Oldfield *et al.,* 1980). However, Oldfield *et al.* ascribe the increase to the large rise in automobile usage which has taken place in Belgium since the early 1950's. As previously stated, while we believe the influence of local vehicular emissions to be minimal, we cannot assess at this time the portion of pollutants, i.e. Pb and PAHs, transported into the Adirondacks from automobile usage outside of the region.

5. Conclusion

Our data support the following conclusions on the levels and origins of trace elements and PAHs in the remote lakes of the Adirondack acid lake region.
— With the exception of perylene, the prime source of all the parental PAHs measured and the majority of the trace elements appears to be combustion.
— All of the combustion products appear to have primarily an anthropogenic origin as opposed to biogenesis produced perylene, retene and hydrochrysenes.
— With the exception of perylene, the levels of all the parental PAHs and several of the metals significantly increased in the surface sediments of both lakes compared to their background concentrations. The concentrations of many of the PAHs in Woods Lake were quite high and approached levels reported for more heavily populated and industrialized areas.
— Although the concentrations were generally 3 to 4 times higher in Woods Lake, the total anthropogenic integrated depositions for most of the metals and 3–4 ring PAHs were about the same in both lakes even though they are located some 30 km apart.
— Anthropogenically derived metals and PAHs decreased in concentration with depth to background levels. In both lakes, this baseline depth corresponded to about 30 yr ago according to the ^{137}Cs method of dating. Crustal or biogenically derived metals and PAHs increased in concentrations with depth, e.g. perylene, or remained fairly constant with depth, e.g. retene, DMOHC, TMTHC.
— Lead has increased to the greatest extent of any of the metals considered compared to its baseline levels in both lakes. The rates at which most of the other metals have entered these lakes have not increased significantly in relation to their baseline inputs.
— Long distance atmospheric transport and region-wide deposition of anthropogenically derived elements and PAHs into these remote lakes appears to be more significant than input from local sources.

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