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Sedimentary Record of Polychlorinated Naphthalene Concentrations and Deposition Fluxes in a Dated Lake Core

BONDI GEVAO, TOM HARNER,[†] AND KEVIN C. JONES*

Department of Environmental Science, Institute of Environmental and Natural Sciences, Lancaster University, Lancaster, LA1 4YQ, United Kingdom

Polychlorinated naphthalene (PCN) concentrations and fluxes were measured in a dated core from the profundal sediments of Esthwaite Water, a seasonally anoxic semirural lake in northwest England. The vertical profile shows that the ΣPCN flux remained fairly constant at 0.4−0.6 μg m^{-2} y^{-1} from depth until the early 1940s, escalating sharply thereafter to a subsurface maximum of ∼12 μg m^{-2} y^{-1} in the late 1950s to mid-1960s followed by a 4-fold decrease to the sediment−water interface. The ΣPCN maximum predates the ΣPCB maximum by ∼20 years, broadly consistent with the time lapse in the production and wide use of both compounds. Furthermore, there was a secondary peak in the profiles of both compound classes corresponding to the period of maximum input of the other compound class. This may be a result of PCNs present as impurities in the manufacture of PCBs and vice versa. The vertical profiles of the individual PCN homologue groups closely followed that of the ΣPCN profile with no overall change in the mass percent contribution of individual PCN congeners throughout the core. To our knowledge, this is the first study to reconstruct the historical record of PCNs in the environment.

Introduction

Polychlorinated naphthalenes (PCNs) are a group of 75 compounds, with physical and chemical properties similar to those of PCBs. Due to their high thermal stability and inertness, they were used in the electric industry as dielectric fluids in transformers and capacitors and as cable insulators (*1, 2*). In addition to their industrial synthesis, PCNs are byproducts of a number of combustion processes, such as municipal solid waste incineration (*3, 4*) and metallurgical processes such as copper smelting (*1, 2*). Other potential sources of PCNs in the environment include the pyrolysis of chlorinated solvents (*4*), their presence as impurities in PCB mixtures (*1, 5*), and formation during chlorination of tap water (*1*). In the United Kingdom, it is thought that PCNs were produced from around 1919 until the mid-1960s, although no production data were available. The precise worldwide production of PCNs is not known, but it has been estimated to be approximately 1 500 000 tonnes, which is 10% of the total global production of PCBs (*6*). The industrial production of technical PCN formulations began to decline in the 1960s and voluntarily ceased in the United States in 1977, but their production in Europe continued until the mid-1980s (*1*).

PCNs, like other persistent organic pollutants (POPs), are ubiquitous in the environment and have been measured in remote areas such as the Arctic (*7, 8*). Their toxicity equivalent (TEQ) contribution in the atmosphere is similar in magnitude to the contribution of PCBs (*3, 8*). POPs are delivered to lacustrine and marine systems by atmospheric transport and deposition, direct and indirect discharges, and riverine inputs. As a result of their low aqueous solubilities they partition primarily onto abiotic and biotic carbon-rich particles in the water column.

The use of dated sediment cores is well-established as a means of reconstructing historical chronologies of contaminant inputs (*9*). Lacustrine sediments are particularly useful for this purpose provided they are undisturbed and if diagenetic processes of bioturbation, pore water transport, and biotransformation are absent or negligible (*10*). In this paper, we examine the sedimentary profiles of PCNs in Esthwaite Water (EW), a semirural lake in the English Lake District. Although various sediment studies have been carried out in different regions of the world (*11, 12*), this study is the first attempt at reconstructing the historical inputs of PCNs in Europe and—as far as we are aware—is the first such study of these compounds worldwide.

Materials and Methods

Description of Study Area. EW has a surface area of 1 km^2, maximum and mean depths of 15.5 and 6.4 m, respectively, and a hydraulic residence time of 13 weeks (*13*). It has a catchment area to lake surface ratio of 17:1. The catchment area for the lake is mostly grazing land typical of the lower valleys of the English Lake District. The lake is situated at latitude 54°N and an altitude of 65 m in a region of cool oceanic climate and high annual rainfall (∼ 2 m y^{-1}) and has been extensively used for scientific purposes (*14, 15*). The sediments are anoxic through much of the year, although a surface oxidized layer (∼1 cm) develops during complete water column mixing in the winter. The absence of a well-mixed surface-sediment layer, as indicated by various radionuclide profiles, suggests that bioturbation is relatively unimportant (*16*) and this aids the reconstruction of contaminant time trends.

Sampling. A sediment core of approximately 1 m in length was obtained from the deepest part (north basin) of EW with a Mackereth minicorer (*17*) in July 1995. The Plexiglas core barrel was washed thoroughly with soap and water and rinsed with Milli-Q water prior to use. Upon retrieval, the core showed an undisturbed mud−water interface. It was transported in a vertical position to the laboratory, stored at 4 °C within 4 h, and sectioned within a week of collection. The sectioning materials were washed and solvent rinsed between each section. Material from each section was immediately transferred to a preweighed solvent rinsed glass jar lined with solvent rinsed aluminum foil, which was reweighed to obtain the wet weight of sediment. All subsamples were stored frozen until analysis.

Extraction and Analysis of PCNs. Sediments for PCN analyses (15−20 g wet weight) were mixed with anhydrous sodium sulfate which had been previously baked out at 450 °C to remove residual water. The mixture was placed in pre-extracted thimbles and extracted in a Soxhlet apparatus with pesticide-grade hexane for 12−16 h on a Buchi 810 fat

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* Corresponding author phone: +44 1524 593972; fax: +44 1524 593985; e-mail: k.c.jones@lancaster.ac.uk.

[†] Present address: Atmospheric Environment Service, Downsview, Ontario, M3H 5T4, Canada.

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extraction unit. Copper turnings were added to the beakers during the extraction stage to remove elemental sulfur.

The extracts were fractionated on a liquid−solid chromatographic column (i.d. 0.7 mm) containing 3 g of silica gel that had been activated at 130 °C for 3 h. The silica gel was wet loaded as a slurry in hexane and capped with approximately 0.2 g of anhydrous sodium sulfate to prevent the column from going dry or trapping air bubbles. The silica gel used throughout this work was pre-extracted with hexane for 24 h, changing solvents every 6 h, dried under vacuum at 90 mbar of pressure, and stored in solvent-rinsed glass jars lined with solvent-rinsed aluminum foil.

The samples were loaded onto the column, and 31 mL of hexane was added (fraction 1, F1), followed by 15 mL of 50:50 hexane:DCM (fraction 2, F2). F1 contained PCBs and PCNs, and F2 contained PAHs *(18)*. Approximately 1 mL of dodecane spiked with internal standards was added to F1, and the sample was blown down under a gentle stream of N_2 to remove hexane. The F1 portions were further fractionated on minicarbon columns, as described in Harner and Bidleman (1997) *(18)*, in order to separate the PCNs from the bulk of the PCB congeners and to allow further reduction in volume for extracts stored in dodecane. The first fraction (F1-1) was eluted with 5 mL of 30% DCM in cyclohexane and contained the multi-ortho PCBs and a portion of the mono-ortho PCBs. The second fraction (F1-2) was eluted using 5 mL of toluene and contained the PCNs, non-ortho PCBs, and the remainder of the mono-ortho congeners. Fraction F1-2 was volume reduced to 25 μL under a gentle stream of N_2. PCNs were determined by gas chromatography electron impact mass spectrometry (GC-EIMS, Fisons MD-800) in SIM using a 30 m DB-5 MS column (0.25 mm i.d., 0.25 μm film).

The mass of individual PCN congeners present in sediment extracts was determined by comparison to a standard calibration mix containing internal standards (PCB 208+209) and Halowax 1014, a commercial mixture of 2-Cl to 8-Cl PCNs. Response factors for individual congeners were determined using known mass percent contributions *(18)*. The concentrations of PCNs were calculated by dividing their masses by the dry weight of the extracted sediment (μg kg^{-1} dry weight), determined gravimetrically after drying subsamples of the sediment in an oven at 105 °C to constant weight.

QA/QC. Blank levels for the analytical method were based on extractions of 10 g of sodium sulfate ($n = 12$). The limit of detection (LOD) was calculated as the blank value + 3 SD. When peaks were not detectable, an area of ~2000 was used which corresponds to integration of background noise. Peaks were only quantified if they exceeded the LOD; the mean blank value was then subtracted. Analytical recoveries were carried out by spiking 10 g of sodium sulfate with Halowax 1014, extracted, and cleaned up as described for sediment samples. Average recoveries for six replicates were >90% for all homologues except hepta- and octa-CNs which were ~82%. The coefficient of variation (CV) for replicate analysis were ≤10% for all homologues except for the hepta- and octa-CN which were 12 and 25%, respectively.

Organic Carbon Determination and Dating of the Core. The determination of organic C was carried out by the use of a Carlo Erba 1108 elemental analyzer. The core has previously been dated using the radioisotopes ^{210}Pb and ^{137}Cs. The detailed methodology used and discussion of the radionuclide profiles is provided elsewhere *(16)*. The dates shown in Figure 2 are based on ^{210}Pb mean sedimentation rates, details of which have been previously reported *(16)*. The mean sedimentation rate for the dated portion of the core was 0.995 ± 0.153 kg m^{-2} y^{-1}.

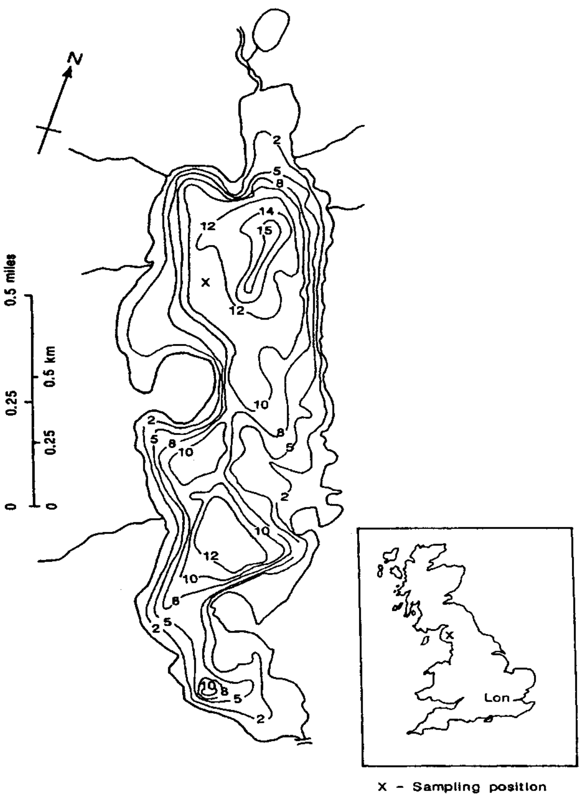


FIGURE 1. Location and bathymetry of Esthwaite Water (from ref *13*), showing its position relative to London. Depth contours are in meters.

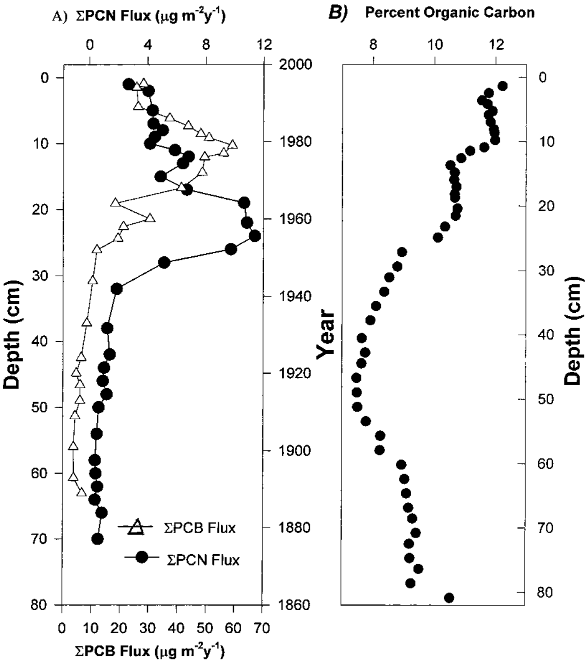


FIGURE 2. (A) Fluxes of ΣPCNs and ΣPCBs to Esthwaite Water sediments and (B) organic carbon profile in Esthwaite Water sediments.

Results and Discussion

ΣPCN Time Trends and Fluxes. The down-core variation in the ΣPCN fluxes to EW is shown in Figure 2 together with the ΣPCB depth profile from the same core *(16)*. Table 1 gives the concentrations of PCNs on a congener basis for repre-

TABLE 1: Concentrations of Congeners, ΣPCN Concentration (ng kg^{-1}), and Fluxes (μg m^{-2} y^{-1}) for Selected Sediment Sections

		section	**1**	**5**	**10**	**15**	**20**	**25**	**30**	**35**	**40**	**45**
		depth (cm)	0–1	7–8	10–11	15–16	20–21	30–32	40–42	50–52	60–62	70–72
		year[b]	1995	1989	1981	1972	1962	1948	1927	1909	1889	1870
labeling system	IUPAC no.	congener										
3–1	CN-19	1,3,5-	66	131	90	127	302	66	32	18	14	18
3–2	CN-14/24	1,2,4-/1,4,6-	545	791	565	763	1486	375	222	108	113	162
3–5	*a*	*a*	40	65	47	77	187	35	40	15	16	
3–7	*a*	*a*	125	148	45	124	280	99	60	47	10	
3–10	*a*	*a*	288	418	413	450	767	179	148	82	71	109
4–1	CN-42	1,3,5,7-	58	95	72	93	207	40	30	9	8	6
4–6	CN-33/34/37	1,2,4,6-/1,2,4,7-/1,2,5,7-	351	566	510	630	1127	207	174	78	58	58
4–7	CN-47	1,4,6,7-	97	165	148	194	500	80	56	25	13	19
4–10	CN-28/43	1,2,3,5-/1,3,5,8-	193	381	368	444	922	118	112	48	33	43
4–15	CN-35	1,2,4,8-	220	261	263	241	390	93	85	73	49	83
4–16	CN-38	1,2,5,8-	302	389	420	432	614	118	168	95	52	43
4–18	CN-46	1,4,5,8-	145	195	177	196	306	64	82	52	30	31
5–1	CN-52/60	1,2,3,5,7-/1,2,4,6,7-	47	130	146	151	540	78	43	19		
5–3	CN-61	1,2,4,6,8-	97	160	121	161	539	63	45	42	23	100
5–4	CN-50	1,2,3,4,6-		47	6	55	169	24				
5–8	CN-57	1,2,4,5,6-	57	72	103	102	347	29	10			
5–9	CN-62	1,2,4,7,8-	45	100	122	131	428	39	27			
5–10	CN-53	1,2,3,5,8-	38	84	117	158	580	56	17			
5–11	CN-59	1,2,4,5,8-	49	78	104	121	320	31	8			
6–1	CN-66/67	1,2,3,4,6,7-/1,2,3,5,6,7-	4	100	191	111	390	105	99			
6–2	CN-64/68	1,2,3,4,5,7-/1,2,3,5,6,8-	54		116	81	252	41	24			
6–3	CN-69	1,2,3,5,7,8-		76	95	89	334	29	22			
6–4	CN-71/72	1,2,4,5,6,8-/1,2,4,5,7,8-		55	87	77	265	21	7			
6–5	CN-63	1,2,3,4,5,6-		53	61	41	173					
7–1	CN-73	11,2,3,4,5,6,7-					554	278				
7–2	CN-74	1,2,3,4,5,6,8-					170	78				
ΣPCNs (ng kg^{-1})			2821	4560	4385	5047	12148	2347	1509	709	490	672
ΣPCN flux (μg m^{-2} y^{-1})			2.7	4.3	4.2	4.8	11.5	2.2	1.4	0.7	0.5	0.6

[a] Unidentified peaks. [b] The year shown is the midpoint of the core section.

sentative sections of the core. The ΣPCN flux recorded in the core is fairly constant at 0.4–0.6 μg m^{-2} y^{-1} from the base (~1870) until the early 1940s. Over this period, the PCN concentrations, although low, were invariably well above detection, especially for the low chlorinated homologues. This was followed by a marked increase in fluxes to a well-defined maximum of ~12 μg m^{-2} y^{-1} in the late 1950s to mid-1960s and then a gradual decrease to the sediment–water interface. The current ΣPCN fluxes (~3 μg m^{-2} y^{-1}) are a factor of 4 less than the maximum flux but around five times higher than the preindustrial background levels. The reduced levels observed in surface sediments are largely the response of the system to restrictions/reductions of PCN production/use and their decreased input into the lake.

Comparison of ΣPCB and ΣPCN Input Profiles. PCNs and PCBs have similar properties and consequently have had similar industrial applications (1, 2). The flux profiles for both compounds is typical of that for many banned man-made chemicals, characterized by an exponential rise in fluxes at depth to a subsurface peak followed by a decrease to a surface. The rise from background and peak in the inputs of PCNs recorded in the core predates that of PCBs by ~20 years, as shown by the sedimentary records. This is consistent with the time difference in the production and use of PCBs and PCNs (1, 19). PCN usage was reduced when suitable alternatives were found. PCBs were the primary replacements for PCNs because of their markedly similar properties (thermal stability and inertness). PCBs were first produced in the 1930s, mass-produced by the early 1950s, peaked in usage from the mid- to late-1960s, and were banned in the U.S. and Europe in the early 1970s. A detailed evaluation of the PCB production and use data in the U.K. has been carried out (19), and a detailed discussion of the input profile in the U.K. environment has been reported previously (16). PCNs were used extensively in the manufacture of electrical insulation materials between the 1930s and 1950s, and in 1956 it was estimated that ~3200 tonnes y^{-1} were produced in the U.S. By 1978, production in the U.S. had fallen to ~320 tonnes y^{-1} due to their replacement by a variety of substitutes, predominantly PCBs. In the U.K, PCNs were produced from around 1919 until the mid-1960s (1, 2), but data on production volumes are not available. The use of both compounds were restricted in the late-1970s in the U.S. and Europe and the inputs of the compounds to sediments have responded accordingly. Another interesting similarity in both ΣPCB and ΣPCN deposition records is that there appears to be two distinct peaks in both their profiles. There is a major peak corresponding to the periods of maximum input for each compound class. However, in addition there is a smaller, distinct peak at the corresponding input maximum of the other compound class. PCNs are known to be present as impurities in PCB mixtures (1, 2) and the release of PCBs will inevitably result in the release of PCNs and vice versa. This probably explains the secondary peak in the profiles of both compounds.

Hydrophobic organic compounds (HOCs) are thought to be associated mainly with organic-rich particles, and for this reason their concentrations are often normalized with respect to organic carbon (OC) (20). In EW, however, it appears that PCN and PCB concentrations and loadings are effectively independent of those of OC. For example, the contrasting sediment profiles of C and ΣPCNs/ΣPCBs (Figure 2a,b) indicate that the historical records of PCNs/PCBs and OC deposition are quite different and that normalizing to C would

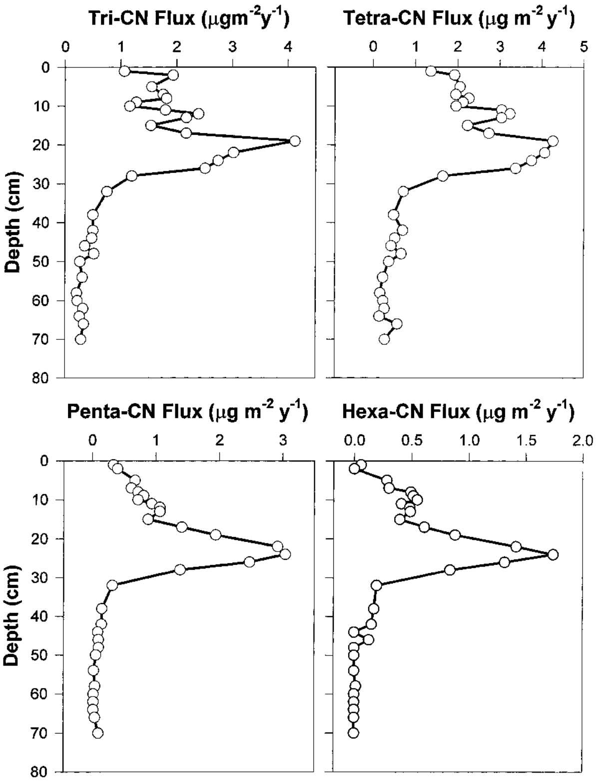


FIGURE 3. PCN homologue fluxes to Esthwaite Water sediments.

in fact confuse the true historical records. This independence is attributed to two main factors. First OC is effectively present in excess due to the eutrophic nature of the lake, resulting in high OC loadings of ∼14−25% by wt in the profundal sediments. Second the concentrations (per unit mass of particles) and fluxes of trace substances tend to be dependent on carrier-phase behavior (i.e. "scavenging") only in large systems with long residence times, such as the oceans and large inland waters. In smaller water bodies with short particle-, pollutant-, and hydraulic residence times, temporal variations in external inputs are more likely to be important, as seen with trace metals (21).

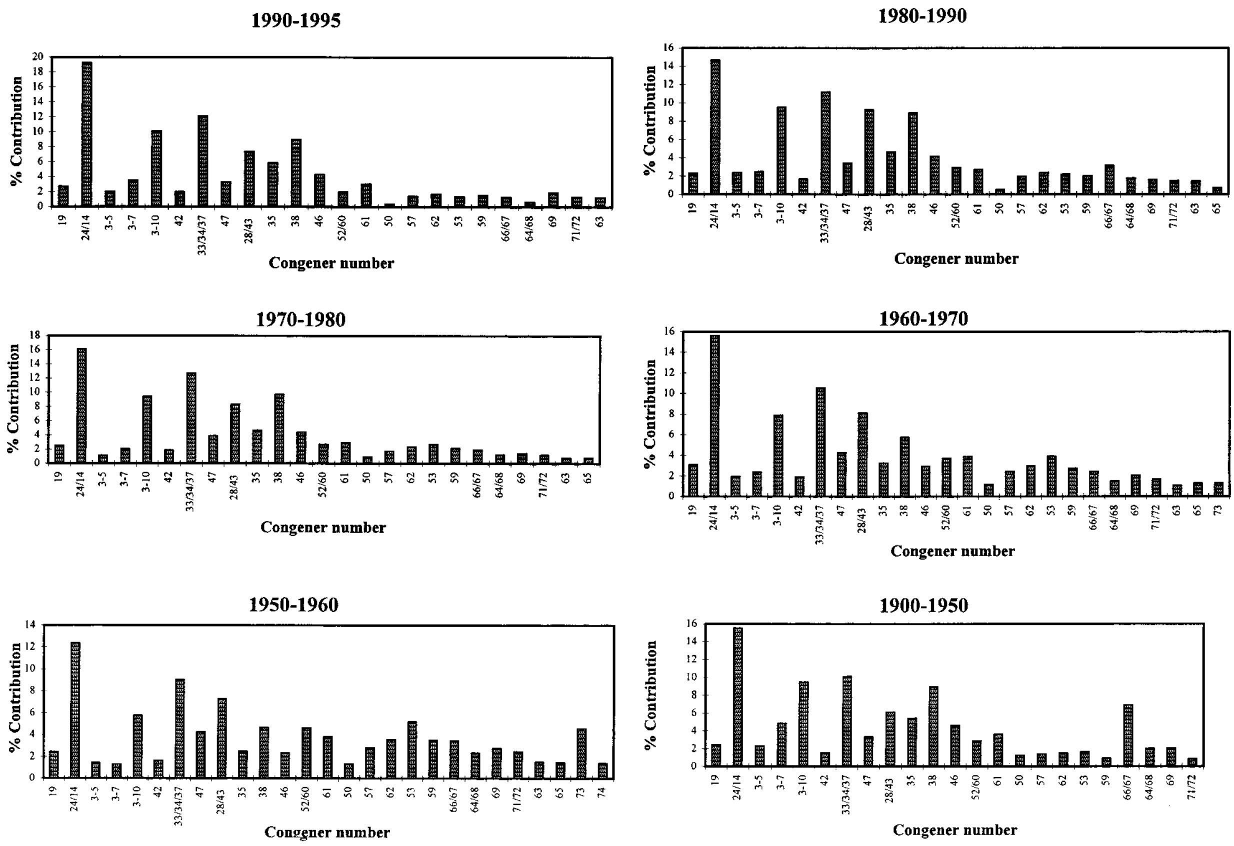
Homologue Trends and Observations of Congener Patterns. Sedimentary profiles for some PCN homologues are presented in Figure 3a−d. The profiles closely track that of the ΣPCN profile with an increase from depth to a subsurface peak in the late-1950s to early-1960s, before an exponential decrease to the surface. The homologue patterns have remained fairly constant throughout the core (Figure 4), and the concentrations/fluxes decrease in the following order: tetra- > tri- > penta- > hexa-PCNs. If the inputs were solely a function of long-range transport and deposition to the lake, rather than a source related function, the specific percent composition would have favored the lower molecular weight congeners as is the case for PCBs (16). The observed order in homologue abundance, however, suggests that the input of PCNs to EW may be a signature of the composition of the mixture released into the environment and not solely due to long-range transport. Congeners with ≥7 chlorines were below detection in most sections of the core. The hepta-PCNs were only encountered in sections corresponding to the period of maximum input to the lake. In these sections CN-73 was a factor of 3 higher than CN-74, the reverse in the technical Halowax mixture released into the environment. This phenomenon has been reported before for uncontaminated sediment samples (5, 24). A similar observation in both particulate and gas phases from a municipal air sample (18) led to speculations that it may be related to some source process, i.e., thermal sources (24).

There are dominant congeners within every homologue group throughout the core: CN-24/14 and peak 3-10 dominate the tri-CNs; CN-33/34/37, CN-28/43, and CN-38 dominate the tetra-CNs; CN-52/60, CN-61, CN-53, and CN-59 dominate the penta-CNs; CN-66/67, CN-69, and CN-71/72 are dominant within the hexa-CN group; and CN-73 > CN-74 for the hepta-CNs as mentioned above (see Figure 4). In a recent study aimed at comparing congener profiles in several environmental media, biological samples, and various source-related samples in Sweden, Jänberg et al. (1997) (5) observed that some congeners were more abundant than others in sediment samples collected at sites remote from any known point sources compared to potential source samples and air samples. Although aerobic microbial degradation has been shown to be unimportant for PCNs (2) the likelihood of anaerobic dechlorination similar to that reported for PCBs in Hudson River sediments (22, 23) may be important for the alterations observed in sediments as opposed to their source signatures. Although this phenomenon has not yet been observed for PCNs, it cannot be ruled out. There is therefore the possibility that some lower chlorinated congeners could be degradation products of higher chlorinated congeners. In another study, Järnberg et al. (1999) (24) provided evidence for photolytic dechlorination of higher chlorinated congeners, particularly those substituted at the 1 and 8 positions. They suggested that photolysis may account for the increase in abundance of CN-33/34 and CN-52/60 in sediments collected from remote locations.

Preindustrial PCN Inputs to EW. Another interesting feature of the EW core is the presence of PCNs in deep sediment sections, dated prior to their industrial synthesis. This has previously been observed for PCBs (16) at this site and elsewhere (20, 25, 26). Several possible explanations include the following: (a) incorrect dating of the core; (25) (b) physical mixing by bioturbation or by smearing during core extrusion; (26) (c) sample contamination during preparation and analysis (27); (d) selective downward migration of compounds in the sediment column (20, 25); (e) production from natural and/or anthropogenic combustion sources (28); and (f) other "natural" production (27).

Incorrect dating probably can be discounted, because the sedimentation rate obtained in this study is similar to those obtained for EW sediments by a number of workers, using a range of techniques (29, 30). The increasing downward enrichment of the less chlorinated groups relative to the more chlorinated homologues indicates that physical mixing of particles alone cannot explain the trends, because presumably all congeners would be affected equally if migration was just associated with particles. Since the absolute concentrations of the individual congeners in this zone are low, the possible effects of laboratory contamination might be important. However, it is reasonable to suggest that all preproduction sediments would have been contaminated to the same relative extent by different homologue groups since the entire analysis was carried out using the same procedure, in the same laboratory, around the same time. It is also worth noting that the concentrations reported were blank corrected. Contamination during sample preparation should therefore have been removed since the blanks were taken through the entire analytical procedure. Additionally, a detailed experiment was carried out to investigate whether smearing and/or laboratory contamination could explain the presence of compounds in preindustrial sediments. The study conclusively showed that it could not be due solely to these factors (16).

PCNs are known to be a byproduct of combustion reactions such as solid waste incineration (3) and in metallurgical processes such as copper roasting (1, 2). There


1990-1995

1980-1990

1970-1980

1960-1970

1950-1960

1900-1950

FIGURE 4. Mass percent contribution of individual congeners to the ΣPCN concentrations at different time periods in the Esthwaite Water core. Unidentified peaks include the following: 3-5, 3-7, 3-10.

is a long combustion history in the Lake District associated with metal exploration. Wood burning (ash and hazel) for the production of charcoal for use in iron ore smelting was locally common in the eighteenth century (31). The so-called “bloomery” method in open hearths was used before 1709, after which it was replaced by furnaces. Some of the bloomeries and subsequent furnaces were located within 1 km of EW. Between 1855 and 1877, a smelting works was built in Coniston, alongside copper mines, about 3 km from EW. Until about 1970, there were 32 blast furnaces operational between Barrow and Millom, utilizing wood burning methods in iron smelting. EW is along the SW−NW direction of the prevailing winds and rain. It is expected therefore that carbon debris associated with the process is likely to enter the lake. It is therefore likely that the metal mining and smelting in the Lake District environment before and during the early period of industrialization may have contributed to the preindustrial levels observed in the study. The increasing downward enrichment of CN-61 may add further credence to the theory since CN-61 has been shown to be predominant in MWI samples (32, 33).

In summary, this study provides the first detailed historical record for emissions and inputs of PCNs to the environment. The major findings in the study are as follows: (i) PCN inputs to the environment have declined in recent years in response to the restrictions imposed on their use in the mid-1970s. (ii) The peak in ΣPCN input predates that of the ΣPCBs by ∼20 years, consistent with the time difference in the production and wide use of both compounds. (iii) There was no significant difference in homologue profiles with time. (iv) Detectable levels of low chlorinated, more water soluble congeners are present in pre-1900 sediments, either because postdepositional mobility associated with porewater diffusive transport or production during metal smelting.

Acknowledgments

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Literature Cited

(1) Crookes, M. J.; Howe, P. D. Environmental Hazard Assessment: Halogenated Naphthalenes; Toxic Substances Division, Directorate for Air, Climate and Toxic Substances, Department of Environment: 1993.
(2) Falandysz, J. Environ. Pollut. 1998, 101, 77−90.
(3) Abad, E.; Caixach, J.; Rivera, J. Organohalogen Compd. 1997, 32, 403−406.
(4) Oehme, M.; Mano, S.; Mikalsen, A. Chemosphere 1987, 16, 143.
(5) Järnberg, U.; Asplund, L.; de Wit, C.; Egeback, A.-I.; Widequist, U.; Jakobsson, E. Arch. Environ. Contam. Toxicol. 1997, 32, 232−245.
(6) Beland, F. A.; Greer, R. D. J. Chromatogr. 1973, 84, 59−65.
(7) Harner, T.; Bidleman, T. F.; Kylin, H.; Strachan, W.; Halsall, C. Organohalogen Compd. 1997, 33, 134−139.
(8) Harner, T.; Kylin, H.; Bidleman, T. F.; Halsall, C.; Strachan, W. M. J.; Barrie, L. A.; Fellin, P. Environ. Sci. Technol. 1998, 32, 3257−3265.
(9) Catallo, W. J.; Schlenker, M.; Gambrell, R. P.; Shane, B. S. Environ. Sci. Technol. 1995, 29, 1436−1445.
(10) Wong, C. S.; Sanders, G.; Engström, D. R.; Long, D. T.; Swackhamer, D. L.; Eisenreich, S. J. Environ. Sci. Technol. 1995, 29, 2661−2672.
(11) Eljarrat, E.; Caixach, J.; Jimenez, M. J.; Gonzalez, M. J.; Rivera, J. Chemosphere 1999, 38, 1901−1912.
(12) Falandysz, J.; Strandberg, L.; Bergqvist, P.; Kulp, S. E.; Strandberg, B.; Rappe, C. Environ. Sci. Technol. 1996, 30, 3266−3274.
(13) Ramsbottom, A. E. Depth charts of the Cumbrian Lakes; Sci. Publ.: 1976; Vol. 33, p 39.
(14) Davison, W.; Woof, C.; Rigg, E. Limnol. Oceanogr. 1982, 27, 987−1003.
(15) Tipping, E.; Woof, C. Limnol. Oceanogr. 1983, 28, 168−172.

(16) Gevao, B.; Hamilton-Taylor, J.; Murdoch, C.; Jones, K. C.; Kelly, M.; Tabner, B. J. Environ. Sci. Technol. 1997, 31, 3274−3280.
(17) Mackareth, F. J. H. Proc. R. Soc. (B) 1965, 41, 38−41.
(18) Harner, T.; Bidleman, T. F. Atmos. Environ. 1997, 31/32, 4009−4016.
(19) Harrad, S. J.; Sewart, A.; Alcock, R.; Boumphrey, R.; Burnett, V.; Duarte-Davison, R.; Halsall, C.; Sanders, G.; Waterhouse, K.; Wild, S.; Jones, K. C. Environ. Pollut. 1994, 85, 131−146.
(20) Muir, D. C. G.; Omelchenko, A.; Grift, N. P.; Savoie, D. A.; Lockhart, W. L.; Wilkinson, P.; Brunskill, G. J. Environ. Sci. Technol. 1996, 30, 3609−3617.
(21) Hamilton-Taylor, J.; Davison, W. In Physics and Chemistry of Lakes; Lerman, A., Imboden, D., Gat, J., Eds.; Springer-Verlag: New York, 1995; pp 217−263.
(22) Brown, J. F., Jr.; Wagner, R. E. Environ. Toxicol. Chem. 1990, 9, 1215−1233.
(23) Quensen, J. F.; Tiedje, J. M.; Boyd, S. A. Science 1988, 242, 752−754.
(24) Järnberg, U.; Asplund, L.; Egeback, A.-I.; Jansson, B.; Unger, M.; Widequist, U. Environ. Sci. Technol. 1999, 33, 1−6.
(25) Hermanson, M. H.; Christensen, E. R. J. Great Lakes Res. 1991, 17, 33−50.
(26) Oliver, B. G.; Charlton, M. N.; Durham, R. W. Environ. Sci. Technol. 1989, 23, 200−208.
(27) Alcock, R. E.; Halsall, C. J.; Harris, C. A.; Johnston, A. E.; Lead, W. A.; Sanders, G.; Jones, K. C. Environ. Sci. Technol. 1994, 28, 1838−1842.
(28) Fangmark, I.; Strömberg, B.; Berge, N.; Rappe, C. Environ. Sci. Technol. 1994, 28, 624−629.
(29) Hilton, J.; Gibbs, M. M. Chem. Geol. 1984−5, 47, 57−83.
(30) Sanders, G.; Jones, K. C.; Hamilton-Taylor, J.; Dorr, H. Environ. Sci. Technol. 1992, 26, 1815−1821.
(31) Millward, R.; Robinson, A. The Lake District; Eyre Methuen: London, 1974.
(32) Imagawa, T.; Yamashita, N. Organohalogen Compd. 1994, 19, 215−218
(33) Schneider, M.; Stieglitz, L.; Will, R.; Zwick, G. Chemosphere 1998, 37, 2055−2070.

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