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The historical residue trend of PCBs in the Agassiz Ice Cap, Ellesmere Island, Canada

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Abstract

Current detailed measurements of contaminant deposition cannot provide a historical perspective except through long-term monitoring programs. In the Arctic, ice caps provide an alternative to lake sediments, in that the annual snow layers reflect atmospheric deposition. As a result of the remoteness of the ice cap and the limited summer melt, annual layers undergo little chemical change, especially after the first summer season, and therefore provide a well-defined historical record. Initial work was undertaken at the Agassiz Ice Cap (80°49′50″ N, 72°56′30″ W) beginning in 1986, but a major effort was undertaken in 1993, during which snow samples covering 30 years were taken from a snow pit. Large volume snow samples were obtained for the determination of PCB congeners. Mean ΣPCB deposition to the ice cap ranged from 930 ng/m^2/year in the winter of 1967–1968 to a minimum of 91 ng/m^2/year in 1980–1981. Since 1980–1981, deposition has again increased to a local maximum of 848 ng/m^2/year (in 1989–1990). The mean deposition for the 30 years of record was 406 ng/m^2/year, with no evidence of a consistent long-term trend.

Keywords: Arctic; Snow; PCB's, Deposition; Trend

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1. Introduction

Semi-volatile persistent organic pollutants have been observed in remote locations of the world, including the High Arctic of Canada. Recently, much of the available information has been summarized in a special volume of the Science of the Total Environment (vol. 122). That volume not only considers the sources, transport pathways, and environmental fate of these contaminants, but also examines some of their potential ecological and human health effects in this sensitive environment, which, until recently, was so often described as 'pristine'.

One of the questions that obviously occurs with respect to these contaminants in the Arctic is that of the depositional trend over time. The temporal trend is directly relevant to assessing the value of current policies in managing the problem or the need for further action and remediation. Con-

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taminant trend work in the Arctic has utilized lake sediments as a means of integrating contaminant accumulation (Lockhart, 1993) as well as determining contaminant residues in tissues of fresh-water and marine fish and marine mammals (Addison et al., 1986; Addison, 1993; Muir, 1993; Norstrom, 1993). Due to the very low sediment accumulation rates in arctic lakes, time slices are of the order of 5–10 years. Tissues for residue analyses are also collected periodically and compared over time. Neither method can be directly linked to atmospheric contaminant delivery, due to the intervening factors that affect accumulation, although it is assumed that these trends are indicative of overall contaminant delivery to the region.

As indicated by Barrie et al. (1992), the winter season is an important period for atmospheric transport of contaminants into the north, due to the strong Siberian anticyclone, which flows mainly from the Eurasian continent into the Arctic. As snow is an effective scavenger of contaminants, probably even of hydrophobic compounds at low temperatures, it is arguable that cold polar ice caps should retain a very detailed record of contaminant deposition to the Arctic. Further, for recent times (i.e. the last 30 years), it is possible to sample ice caps by annual layer with little ambiguity in dating. Moreover, as the ice cap receives snow directly during deposition of fresh snow, at least there are no terrestrial delivery processes that must be considered when interpreting the apparent trend.

The Agassiz Ice Cap on Ellesmere Island, Canada, should provide a credible historical record of anthropogenic pollutant deposition, because (1) average summer melt affects only about 3% of the winter snow layer, resulting in negligible redistribution of ions between adjacent snow layers (Barrie et al., 1985) and (2) annual snow accumulation at this lattitude and elevation represents, on average, more than 75% of the total annual precipitation (Woo et al., 1983). The seasonal specific conductance record of the firn has been shown by Barrie et al. (1985) to reflect the trace constituent composition of the atmospheric aerosol, which undergoes a strong seasonal variation with maximum ion concentrations and therefore specific conductance during the arctic winter (Rahn and Shaw, 1982; Hoff and Barrie, 1986).

The temporal trend of persistent organic pollutants in northern ice caps has received much less attention than the trend for inorganic compounds. The earliest known work on persistent organochlorine compounds in ice caps from the Canadian Arctic was in 1970 (Stengle et al., 1973), at Mount Logan in the Yukon Territory. Nineteen snow samples were taken at depths of 1–15 m, with DDT not quantified in any of these samples, at a detection limit of 5 ng/l. Subsequently, limited data have been used to speculate on contaminant trends for the Agassiz Ice Cap on northcentral Ellesmere Island, Canada (Barrie et al., 1992; Gregor et al., in press). The conclusions from this work suggested that total PCB deposition rates to the Ice Cap may have decreased by a factor of about two from the mid-1970s through the mid-1980s. These authors clearly recognized that a 17-year record from the Ice Cap was too limited to provide data that could confidently show a trend; consequently, they recommended follow-up work.

This investigation set about to extend the depositional record of selected chlorohydrocarbons in the glacial snow of the Agassiz Ice Cap on Ellesmere Island at 80°49′50″ N, 72°56′30″ W (Fig. 1) and, if possible, to confirm the trend that had been observed in the previous study. The goal of this expedition was to provide a detailed annual record for the last 30 years of deposition by means of large volume snow samples collected in a surface pit.

In March 1993, more than 5 t of equipment and nine people were moved to the field site on the Agassiz Ice Cap at an elevation of approximately 1600 m above sea level. In 3 weeks, nearly 20 t of snow were excavated from a pit 8.3 m deep, which provided the basis for the first 30 years of samples. Below this depth, hand coring was intended to provide uncontaminated samples. Unfortunately, the corer did not work successfully in March. The corer was subsequently modified and a smaller team returned to the site in May to attempt the work, but again without success. Therefore, until the site can again be revisited with other equipment, we will report only on the

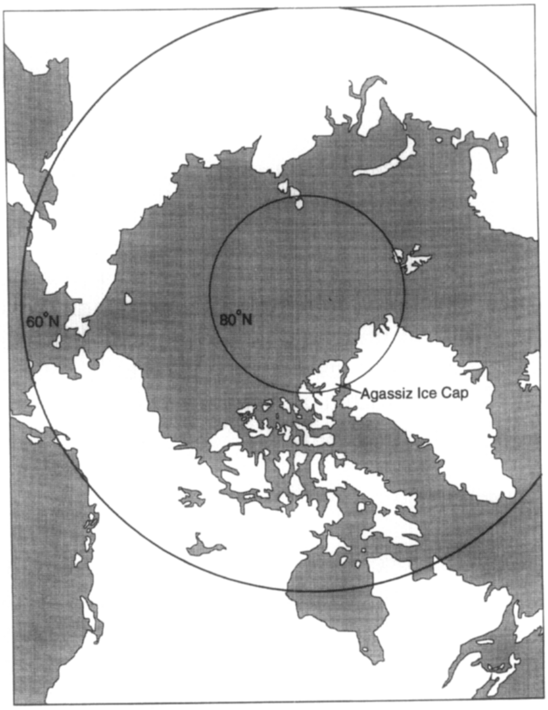

Fig. 1. Location map of Agassiz Ice Cap, Ellesmere Island, Canada.

results from the last 30 years. While the samples have been analyzed for a broad spectrum of persistent organic pollutants, this report is limited to polychlorinated biphenyls (PCBs).

2. Methods

2.1. Field sampling

The sampling site was located 1.7 km north of the semi-permanent field camp and was approached from downwind by snowmobiles. The snowmobiles were not taken closer than 50 m to the actual sampling site. The sampling pit was covered by a large portable shelter to provide protection for field personnel and to prevent windblown surface snow from entering the pit.

The snow was excavated manually and lifted to the surface using a battery powered winch. Annual layers were identified on the basis of visual inspection of the firn, the presence of ice layers, plus the conductivity profile in the snow determined from subsamples down the face of the wall at approximately 2-cm intervals, and measured at the field camp at 0°C, as outlined in Gregor et al. (in press) and Peters et al. (1994). Annual layers were sampled from autumn (i.e. immediately above the summer melt layer, if present) to the following autumn. This method ensured that any contaminant migration during the summer melt was contained within the layer for the winter season preceding that melt.

Samples were collected using a stainless steel shovel that had been rinsed with high purity acetone and hexane. Annually integrated samples were placed into custom-made aluminum snow cases that were also rinsed with high purity acetone and hexane at the sampling site. These samples were collected for the analysis of PCBs as reported here, organochlorine pesticides (which will be reported on at a later date), and PAHs as reported in this volume by Peters et al. (1994).

The snow was melted inside the sealed sampling cases at the field camp in a circulating water bath maintained at 17°C. The melt water was extracted with a Goulden large volume extractor (GLVE), using a centrifugal pump to mix and emulsify the sample with the extraction solvent (Goulden and Anthony, 1985; Neilson et al., 1988). The flow rate in the GLVE was 250 ml/min and the extraction solvent was dichloromethane (DCM). The GLVE was thoroughly cleaned with high purity acetone and hexane between samples. The initial 150 ml of DCM added to the GLVE was spiked with 60 ng of 2,3,4,5-tetrachlorobiphenyl (PCB 70). Makeup DCM, metered into the GLVE at a nominal rate of 3 ml/min, was further spiked with 10 ng δ-HCH and 20.32 ng 1,3,5-tribromobenzene in methanol. The graduated cylinder containing this spike was always thoroughly rinsed with DCM, which was subsequently metered to the GLVE to ensure a quantitative transfer of the spike during the course of the extraction. At the end of the extraction, the DCM was drained from the GLVE into a precleaned 500-ml amber glass bottle. The water remaining in the GLVE was further separated from the DCM using a separatory funnel. The DCM and a small quantity of the water, to minimize volatilization of the DCM, were added to the bottle. The bottles were sealed and kept refrigerated during shipment to, and storage at, the analytical laboratory. Finally, the volume of water extracted was measured and recorded. Sample volumes ranged from 14.8 to 20 l of meltwater.

2.2. Analytical procedures

The sample extract was allowed to reach room temperature prior to processing, generally following the procedure developed at Environment Canada's National Water Quality Laboratory (Anonymous, 1987). Thereafter, excess water was removed by passing the sample through anhydrous Na_2SO_4 in an Alihn filter funnel. A 20-ml volume of DCM was then passed through the Na_2SO_4 with gentle suction. The sample was then spiked with 56.52 ng of 1,3-dibromobenzene and reduced to approximately 5 ml by reduced pressure rotary evaporation at 30°C. Following solvent exchange with hexane and base wash with 0.1 M K_2CO_3 (aq.), the extract was again passed through Na_2SO_4 with three 5-ml hexane rinses. Subsequently, the sample was spiked with 40 ng of 2,3,5,6-tetrachlorobiphenyl (PCB 65) and 235 ng of perthane prior to concentration by reduced pressure rotary evaporation to approximately 2 ml. This concentrate was applied to a silica gel column prepared with 10 g of 60-mesh silica gel, fully activated at 500°C for 12 h. The sample was eluted to obtain four fractions: F1, 80 ml hexane; F2, 85 ml 35% (v/v) DCM in hexane; F3, 85 ml 60% (v/v) DCM in hexane; and F4, 50 ml DCM followed by 50 ml MeOH. Each fraction was then spiked with 65 ng of octachloronaphthalene (OCN) and concentrated by reduced pressure rotary evaporation to approximately 2 ml, solvent exchanged to iso-octane, and finally concentrated to 1.0 ml under a nitrogen stream. The solvent was then transferred to vials and stored at 4°C until analysis.

Samples were analyzed with an HP 5890 gas chromatograph equipped with dual ECD detectors. The two 30-m × 0.25-mm fused silica columns were SPB1 (polydimethyl siloxane) and SPB5 (5% phenyl and 95% methyl polysiloxane) (listed as Supelco products), with a phase thickness of 0.25 μm. The sample was injected using an autosampler in splitless mode with a time delay of 0.5 min, after which the injector was purged. An initial oven temperature of 80°C was maintained for 2 min, increased at 4°C/min to a maximum of 280°C, and held for 12 min. Injector and detector temperatures were 220°C and 350°C, respectively. The carrier gas was helium at 1.0 ml/min with nitrogen as the makeup gas at 30 ml/min.

PCB congeners reported here had to be confirmed on both columns with retention times within ± 0.02 min relative to the external standard. Concentrations determined on both columns had to agree within a factor of two in order to quantify the compound.

3. Results

Mean recovery of the initial DCM spike (2,3,4,5-tetrachlorobiphenyl) is 89%. Mean recoveries for δ-HCH and 1,3,5-tribromobenzene, added during the field extraction, are 107% and 46%, respectively; 1,3-dibromobenzene, 2,3,5,6-tetrachlorobiphenyl and perthane, spiked during the cleanup and fractionation procedures, have mean recoveries of 46%, 90%, and 125%, respectively. Mean recovery of OCN for the 34 samples analyzed is 78%. Although recoveries of individual compounds are variable, there is good consistency for each compound and no evidence of major methodological problems.

A total of 67 PCB congeners were analyzed in this study. Of these, only six congeners (5, 7, 8, 16, 52, 54) were present in the background corrected field blanks at significant concentrations (Fig. 2) and these have not been included in this analysis. A total of 60 PCB congeners are considered to comprise ΣPCB and these are listed by homologs in Table 1. Reported concentrations of individual congeners were not adjusted for recoveries based on individual sample surrogate recoveries or mean recoveries of laboratory samples spiked with standards. The first procedure introduces a degree of subjectivity into the results that is variable within and among samples. The second procedure corrects for variable recoveries of different congeners, assuming all things constant, but cannot account for human factors affecting real samples. Therefore, adjustments of this nature may intro-

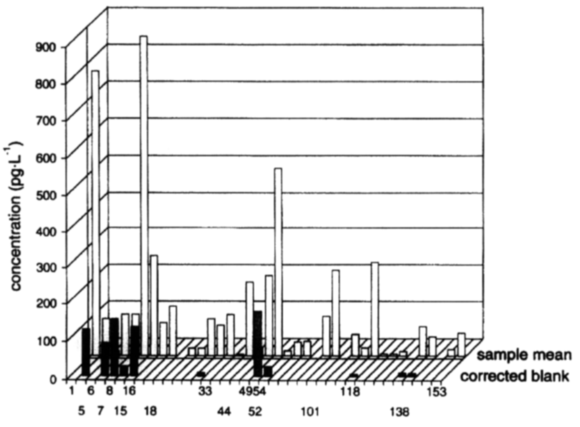

Fig. 2. Sample mean PCB congener concentration compared to mean corrected field blank for congeners analyzed up to IUPAC 153. Detection of congeners heavier than 153 was infrequent. Some of the dominant congeners, especially those present on the blanks, are numbered.

duce additional and unnecessary inconsistencies into the data set while achieving little more than increasing the reported concentrations.

At least five of the congeners considered here have been found to be absent from commercial Aroclors (Schulz et al., 1989). The presence of any of these compounds in the samples would therefore not be expected. Three of these congeners, PCB 86, 103, and 154 were not detected. PCB 121 was detected once, while PCB 182 was detected twice. In these three cases, the concentrations were low (< 100 pg/l); although they were not eliminated from the data set, they would not significantly affect interpretation.

The ΣPCB concentration profile for the 30 years of samples from the Ice Cap is shown in Fig. 3. The layers for the winter seasons of 1964–1965 and 1965–1966 were sampled as one in the field and are treated here as a combined layer (see Peters et al., 1994, for further discussion on annual layers). ΣPCB concentrations over the period of record range from 1.2 to 6.7 ng/l, in 1968–1969 and 1983–1984, respectively. This is not indicative of any kind of a trend as evidenced by the high inter-annual variability. Mean concentration of ΣPCB over the 30-year period is 3.5 ng/l (±1.3). The top two surface layers were sampled contiguously in triplicate. The mean con-

Table 1
Homolog distribution of PCB congeners measured in this study showing no evidence of contamination on procedural blanks

Homolog	Congeners[a] analyzed
Mono and di	1, 6, 15
Tri	17, 18, 19, 22, 28, 31, 32, 33
Tetra	40, 44, 45, 49, 60
Penta	86, 87, 101, 103, 105, 110/77, 114, 118/149, 121
Hexa and hepta	128, 129, 137, 138, 141, 143, 151, 153, 154, 156/171, 157, 159, 170, 173, 180, 182, 183, 185, 187, 189, 191, 193
Octa, nona and deca	194, 195, 196, 199, 200, 201, 202, 203, 205, 206, 207, 208, 209

[a] Numbering of congeners is based on IUPAC.

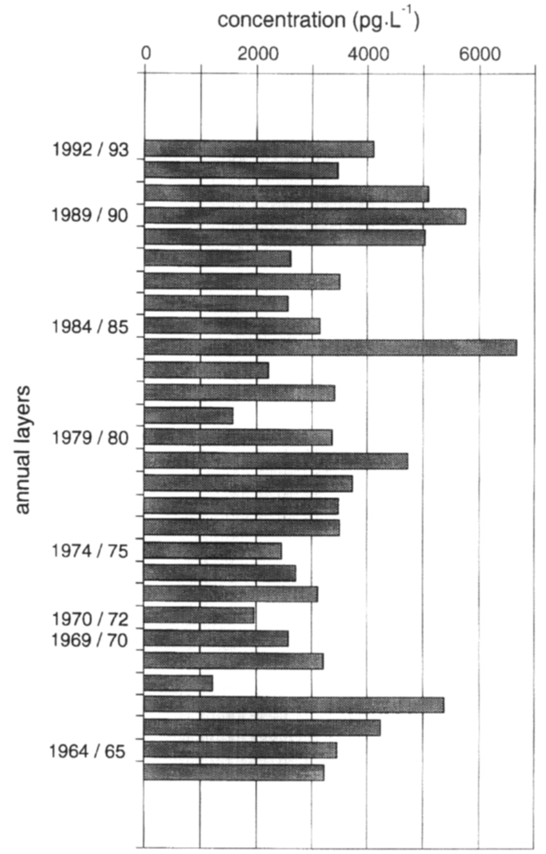

Fig. 3. Temporal profile of ΣPCB congener concentration for Agassiz Ice Cap from surface (1992–1993) to bottom of pit (1963–1964). Note that the layers for winter seasons 1970–1971 and 1971–1972 have been sampled as a single layer.

centrations (± S.D.) for each layer, respectively, are 4.1 (± 0.6) and 3.5 (± 0.5) ng/l. The coefficients of variation for both of these samples are of the order of 15%, much less than the variation among the annual samples.

The concentrations of the PCB homologs over the period of record are shown in Fig. 4. It is evident that the lower chlorinated congeners dominate, with only infrequent detections of congeners with eight or more chlorines. As well, much of the yearly variability in concentrations noted above is derived from the mono- and dichlorinated PCBs.

4. Discussion

PCBs have been shown to be between 80% and 100% in the vapor phase in urban atmospheres in the area of the Laurentian Great Lakes (Murphy and Rzeszutko, 1977; Doskey and Andren, 1981; Eisenreich et al., 1981). Duinker and Bouchertall (1989) investigated the partitioning of several PCB congeners and concluded that while only a very small fraction of total PCB was found in aerosols, the aerosol compartment accounted for more than 99% of the PCBs in rain. More highly chlorinated congeners were particle-associated, whereas the lower chlorinated congeners remained essentially in the vapor phase. The partitioning of PCBs between vapor and particles in the atmosphere is inversely related to temperature (Bidleman, 1988). Consequently, at ambient arctic temperatures, a greater proportion of lower chlorinated PCBs are likely to be particle associated than in the temperate source regions. Thus it can be speculated that for the arctic winter, ice crystals, fresh snowfall, and some unknown component of dry deposition scavenge rather effectively the full range of PCB congeners. Nevertheless, during the winter season in the Arctic, atmospheric concentrations of higher chlorinated PCBs will be less than those of lower chlorinated PCBs, as they will have been preferentially scavenged during transport (Lunde et al., 1977). The distribution of PCBs in the Agassiz Ice Cap is entirely consistent with this previous evidence, with concentrations being in-

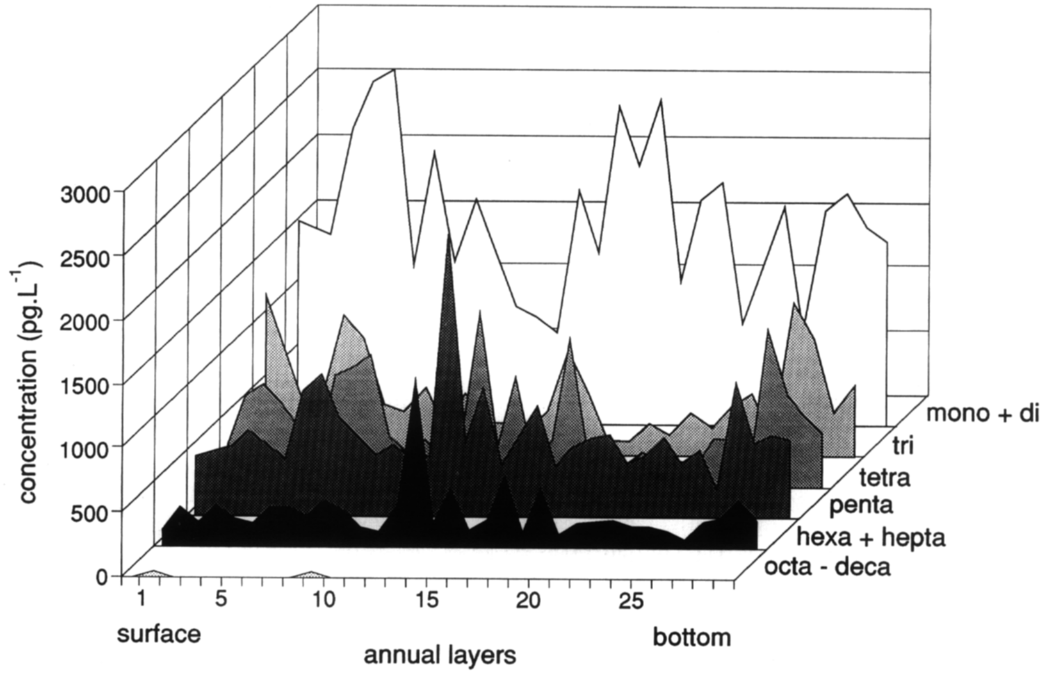

Fig. 4. Temporal profile of PCB homolog concentrations for Agassiz Ice Cap from surface (1992–1993) to bottom of pit (1963–1964).

versely proportional to the number of chlorine atoms. The presence of PCBs with eight or more chlorines is infrequent to the point of insignificance in the PCB profile.

PCB concentrations in the firn can vary for a number of reasons; thus temporal trends of contaminant fluxes may be more informative. The annual flux of PCBs to the Ice Cap is a function of the concentration, the snow thickness, and the water content of the annual layer. The annual fluxes over the period of record for the PCB homologs are shown in Fig. 5. Mean annual ΣPCB deposition to the Ice Cap over the 30 years is 406 (± 187) ng/m^2/year. The maximum estimated deposition for the period of record was 930 ng/m^2/year in 1967–1968. The general temporal pattern of deposition is a period of high deposition prior to 1968–1969, then in 1968–1969 a significant decrease in deposition, followed by a slow, consistent increase until 1979–1980 when a further decrease in deposition occurred, with the period of record minimum of 91 ng/m^2/year in 1980–1981. Subsequently, ΣPCB deposition again rose, with a local maximum of 848 ng/m^2/year in 1989–1990. The mean annual flux for the last 3 years of record (1990–1991, 1991–1992, and 1992–1993) was 465 ng/m^2/year, which is not significantly different from (but moderately higher than) the period of record mean flux (406 ng/m^2/year), indicating the absence of any overall temporal trend.

In contrast, Peters et al. (1994) indicate that ΣPAH deposition at this site has decreased over the period of record. ΣPAH deposition has remained relatively uniform for the past 20 years at approximately 11 (± 6) μg/m^2/year. This is much lower than the mean annual flux of 74 (± 20) μg/m^2/year measured between 1963–1964 and 1970–1971. The major decline in ΣPCB flux occurred in 1967–1968, 3 years before the ΣPAH decline, suggesting that these events were real rather than some unique phenomenon of a particular layer of the ice cap.

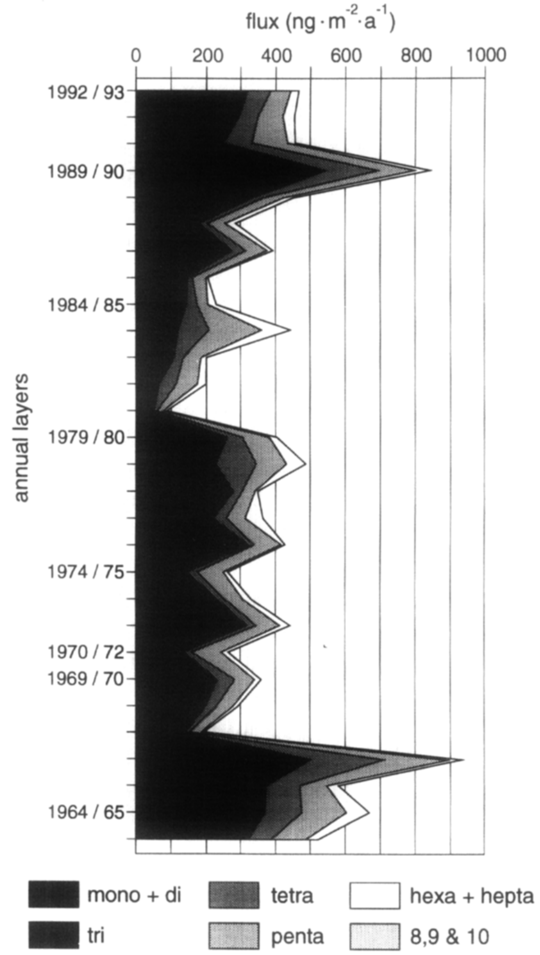

Fig. 5. Temporal profile of PCB homolog flux for Agassiz Ice Cap for period of record from 1992–1993 to 1963–1964.

Deposition of atmospherically transported, semi-volatile organic contaminants to the Ice Cap can vary annually for a variety of reasons. For example, the southward extent of the polar front over the continental land masses, which largely defines the potential source area for these pollutants, can vary. Emissions to the atmosphere from sources can be highly variable, as can the atmospheric transport and deposition processes. On average, the contributing air shed should not change and thus an observable decrease in deposition to the Ice Cap would have to result from a major change in emissions. Future work will attempt to normalize these ΣPCB fluxes for source, transport, and deposition variability.

Peters et al. (1994) note that the PAH flux trend may be indicative of changing global fossil fuel use. Analogously, the PCB data may therefore suggest that there has not been a major change in the PCB availability in the northern hemisphere over the past 30 years. This observation is indirectly supported by recent evidence from studies of agricultural soils in England. The UK study estimated that soil residues have decreased by as much as 90–95% since a peak in the early 1970s (Jones et al., 1992; Alcock et al., 1993). Further, volatilization has been determined to be a major cause of this loss, resulting in a shift toward greater proportions of the heavier homolog groups comprising the bulk of PCBs remaining in the soils. If northern hemisphere soils are releasing PCBs, a lag can be expected in a corresponding decline in deposition of PCBs in the Arctic. Add to this the fact that use in many Eurasian countries has continued past 1970 (Barrie et al., 1992), and the result may be that a measurable decrease in arctic PCB deposition will not occur for some time.

Gregor et al. (in press) published a 17-year trend of total PCB (analyses were by packed column gas chromatography using a 1:1:1 mixture of Aroclor 1242, 1254, and 1260 for quantification) in the Agassiz Ice Cap between 1969–1970 and 1986–1987. This Aroclor mixture consists largely of PCBs with more than three chlorines and thus a direct comparison with the ΣPCB data reported here is not possible. However, by summing only the congeners with more than three chlorine atoms, it is possible to make a crude comparison of the two sets of data (Fig. 6). Although there are some similarities between the two sets of data, the most notable differences occur in 1983–1984 and 1978–1979, when the 1993 data show higher fluxes, and for the period 1969–1970 through 1973–1974, when the flux for the 1987 samples was higher. Even with these differences, it is possible to see how this earlier work concluded that PCB deposition to the Ice Cap was decreasing, as the low flux rates for the mid-1980s concur in both cases; it could not be foreseen in 1987, however, that the minimum deposition had been reached in the early 1980s and that deposition would continue to increase following that time to near the levels of the early 1960s.

The sampling sites for 1987 and 1993 were approximately 9 km apart on the Ice Cap. The 1987 site was perhaps a better site, at approximately 7 km from the base camp. Consequently, a great deal of time was spent ‘commuting’ and it was not possible to work during the frequent periods of poor visibility. The 1993 site, approximately 2 km from the base camp, allowed work to be undertaken in all conditions. It may, however, have a lower accumulation rate and a greater chance of scouring. This cannot be confirmed; but scouring would conceivably reduce the measured flux rate, whereas most of the major differences between the two sites occurred as a result of higher calculated flux rates to the 1993 site. Therefore, perhaps the change to congener-

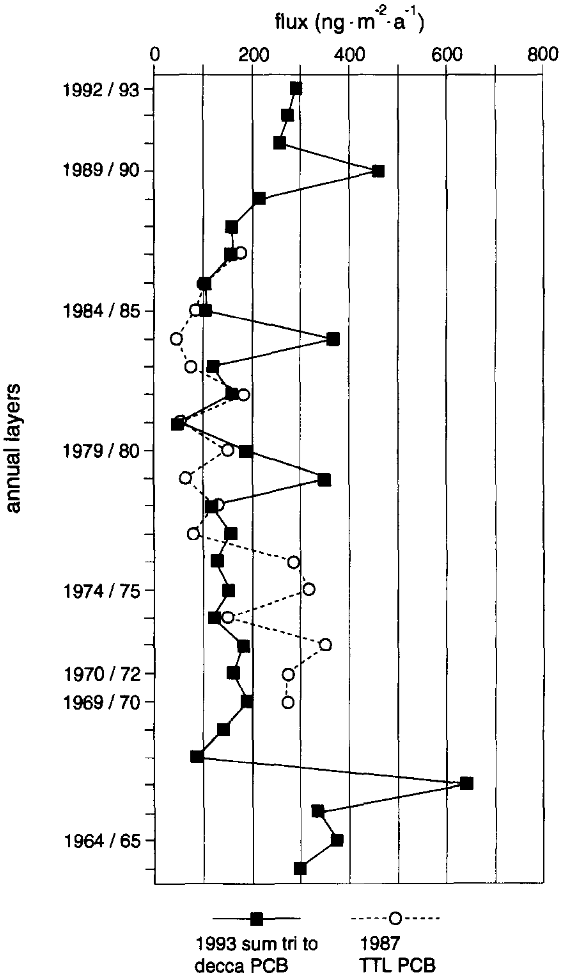

Fig. 6. Sum of tri- to deca-PCB congener flux from 1993 pit compared with total PCB flux from 1987 pit.

specific analyses is the primary reason for the increased variability seen at the 1993 site.

One final factor that could affect the temporal trend of PCBs in the firn is the impact of revolatilization of PCBs during the summer season. As demonstrated by ten Hulscher et al. (1992), as well as Jones et al. (1992) and Alcock et al. (1993), volatilization of PCBs can be an important process. This depends to some extent on where the PCBs are located on the snow crystal (i.e. particle associated as part of the condensation nuclei or adsorbed onto the surface). Previous work (Gregor et al., in press) has shown that roughly one-third of the total PCB deposition to the Ice Cap may be revolatilized. This has not been confirmed as yet in this data set and requires further investigation.

5. Conclusions

ΣPCB flux to the Agassiz Ice Cap on northcentral Ellesmere Island, Canada, shows no clear, continuing temporal trend for the last three decades, due in part to the inter-annual variability. Nevertheless, there is some evidence of a pattern to the deposition. Specifically, deposition was generally higher in the early 1960s, with a maximum of 930 $ng/m^2/year$ in 1967–1968; in 1968–1969, there was a significant decrease in deposition. Deposition then increased slowly and consistently until 1979–1980, when a further decrease in deposition occurred, with the period of record minimum of 91 $ng/m^2/year$ in 1980–1981. Subsequently, ΣPCB deposition again rose, with a local maximum of 848 $ng/m^2/year$ in 1989–1990 and a mean flux for the last 3 years of 465 $ng/m^2/year$, which is moderately higher than the period of record mean of 406 $ng/m^2/year$.

The PCBs on the Ice Cap are dominated by the lower chlorinated homologs, which also account for a great deal of the inter-annual variability. A normalization procedure that effectively accounts for inter-annual deposition variability would be very useful in assessing real trends. This topic should be investigated further. An earlier study that considered only 17 years of record between 1969–1970 and 1986–1987 and concluded that PCB deposition to this Ice Cap was decreasing, cannot be supported from this work. Recent reports of PCB volatilization from temperate soils indirectly support this observation, in that arctic PCB deposition considerably lags controls in temperate regions, due to the presence of the soil reservoir and the ongoing use and/or poor disposal practices in other regions. Determination of the PCB trend in arctic ice caps is complicated by variable source and delivery functions, scouring, and revolatilization. Further, longer term investigation of deposition to ice caps in association with process studies will add to the understanding of long-term trends of deposition in the Arctic.

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