Environment Canada
Environmental Protection Branch
Pacific and Yukon Region

Temporal Trends in PCB Concentrations in the Canadian Environment

- A Summary of Current Information

Regional Program Report 01-02

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By
C. Garrett and D. Goyette

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ABSTRACT

The objective of this report was to provide a summary of available information on temporal trends of PCBs in the Canadian environment and current sources to the environment. Particular emphasis was placed on reviewing studies which documented a stabilization or increase in environmental levels of PCBs. The report presents Canadian temporal trend information for selected regions including the West Coast, Arctic, Great Lakes, Atlantic Coast and the St. Lawrence River system. Trend information on environmental concentrations of PCBs in other areas of the world was also presented.

RESUME

L'objectif de ce rapport est de fournir un résumé sur l'information disponible concernant les tendances temporelles de BPC dans l'environnement canadien et les sources courantes de l'environnement. L'emphase a été placée sur la révision des études dont lesquelles indiquaient la stabilisation ou l'augmentation des niveaux de BPC dans l'environnement. Le rapport présente l'information sur les tendances temporelles canadiennes des régions sélectionnées incluant la Côte ouest, Arctique, les Grands Lacs, Côte de l'Atlantique et le fleuve St.-Laurent. De l'information axée sur les concentrations environnementales de BPC dans d'autres régions dans le monde a aussi été présentée.

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SUMMARY AND CONCLUSIONS

- Environmental information for most areas of Canada was not adequate to determine temporal trends. The importance of temporal trend monitoring for environmental contaminants should be recognized. In addition to its importance in a regulatory and compliance context, long-term monitoring for temporal trends provides insight into how environmental contaminants respond to changes in environmental conditions such as reductions in point source releases, remobilization, alterations to the structure of biological communities, and climate changes (Whittle, M., personal communication).

- The availability of archived environmental samples was valuable in determining temporal trends in the Great Lakes and certain other areas. The routine archiving of samples (sediments and biota) from environmental surveys should be encouraged in order to better assess temporal trends in the future.

- Most of the information on temporal trends for PCBs in the Canadian environment pertains to the Great Lakes and Arctic regions. Information for other regions including the west and east coasts was limited.

- Industrial discharges were the main historical sources of PCB contamination in the Great Lakes region. In Arctic regions, however, atmospheric deposition was largely responsible for the widespread presence of PCBs. PCB releases from DEW lines in the Arctic were substantial but the impacts were localized, and it was determined that these releases contributed only 1% of the total PCB loadings in the Arctic environment during a 30 year period of study. Ocean currents and north-flowing rivers also conveyed PCBs to Arctic regions.

- Although dated sediment core information on PCB trends was limited, cores collected in the Georgia Strait, Fraser Basin lakes, Great Lakes and the St. Lawrence estuary generally reflected past PCB use with concentrations increasing through the 1940s until the 1970s when declines were noted. Sediment cores from Arctic lakes showed the delayed appearance of PCBs, and indicated a lag time between PCB use and long-range transport to Arctic regions.

- PCB concentrations in the Canadian environment peaked in the 1960s and early 1970s but declined rapidly in many areas following the introduction of stringent regulations on PCB use and manufacture in the 1970s. Similar declines were also detected in other areas of the world.

- The largest environmental decreases occurred in areas where high PCB concentrations had previously been attributed to the close proximity of point sources. Decreases in less contaminated areas (removed from point sources and urban areas) were less apparent.

- The rapid decline in environmental concentrations in the 1970s indicated that the federal regulations were very effective in reducing environmental levels, however, areas of high PCB contamination still exist.

- Although point sources of PCB releases to the environment have been almost eliminated through the introduction of controls in most western countries, the large repositories of PCBs in soils and bottom sediments leads to recycling of PCBs in the environment. Volatilization from soils and surface waters results in the atmospheric transport and redistribution of PCBs to other areas and is thought to be the dominant source of PCB redistribution in the environment. Long-range atmospheric transport of PCBs via processes such as 'cold condensation' and 'global fractionation' result in the contamination of Arctic regions with specific PCB congeners.

- Environmental monitoring conducted since the 1980s showed that PCB concentrations in the environment were decreasing much more slowly than in the 1970s. In some areas the environmental levels appeared to have stopped decreasing or were decreasing at such a slow rate that they appeared to have stabilized. However, it is likely that PCB concentrations in the environment were still declining overall, albeit at a slower rate than was observed in the 1970s. Although existing sampling programs were adequate to detect the prominent decreases in concentrations observed in the late 1970s, a more subtle rate of change in PCB concentrations would be more difficult to determine with currently available data.

- Adequate time frames and sample numbers are crucial in the interpretation of temporal trends of PCBs. In one instance, where environmental concentrations appeared to have stabilized, the reanalysis of data using a longer time period showed concentrations continuing to decline.

- In future monitoring programs large sample sizes will likely be required in order to detect the more subtle changes in overall PCB concentrations which are now occurring and to compensate for the high variability observed in PCB concentrations among individual organisms.

- Methods of data analysis can affect the ultimate interpretation. For example, Pekarik and Weseloh (1998) analyzed herring gull data from the Great Lakes using change point regression analysis. For some of the Great Lakes herring gull colonies, their findings on temporal trends were different from those of other researchers using different methods of data analysis (Stow, 1995).

- Some researchers reported that recent PCB concentrations were higher than 1980s concentrations in some areas. However, these researchers have not attributed these apparent increases to new sources or to increased ambient environmental levels, but suggested that such factors as weather conditions, overwintering behaviour, changing predator-prey dynamics and changes in diet may be largely responsible.

- Predator-prey dynamics appeared to play an important role in determining PCB concentrations in higher trophic level predators. It was suggested that the presence of a long food-web in lakes can result in higher PCB concentrations in high trophic level predator fish species. Some researchers suggested using fisheries management practices as a method to lower the PCB concentrations in Great Lakes sport fish.

- It was suggested that future reductions in environmental levels will occur slowly, particularly in humans and other long-lived species such as whales. PCBs persist in tissues for a long time and significant transfers of PCBs from parent to young have been observed.

- Available information to date does not support the theory that coplanar PCBs are declining less rapidly in the environment than other PCB congeners. However, some researchers reported an enrichment of the more highly chlorinated congeners in the environment. This was attributed to the more highly chlorinated congeners being more stabile and less volatile and, therefore, less prone to environmental degradation and atmospheric transport.

1. Introduction

PCB concentrations in the Canadian environment peaked in the 1960s and early 1970s, but declined rapidly in many areas following the introduction of stringent regulations on the use and release of PCBs. Similar declines were observed in other areas of the world as point sources of PCBs to the environment have almost been eliminated in most western countries through the introduction of controls. However, environmental monitoring conducted from the 1980s to the present indicated that PCB concentrations in the Canadian environment are now decreasing much more slowly than in the 1970s. Researchers suggested that, in some areas, environmental levels have stopped decreasing or are decreasing at such a slow rate that they appear to have stabilized. In a few instances, researchers reported that current PCB levels are somewhat higher than those observed in the 1980s. The reason for the apparent stabilization of PCB concentrations has been the subject of some dispute, and regulatory agencies have questioned the need to amend existing PCB regulations.

The objective of this report was to provide a summary of available information on temporal trends in the Canadian environment and current sources to the environment. Particular emphasis was placed on reviewing studies which documented a stabilization or increase in environmental levels of PCBs.

The importance of PCB recycling through the release of PCBs from contaminated soils and sediments and the subsequent redistribution of PCBs in the environment through such mechanisms as long-range transport and global fractionation are discussed in Section 2. Section 3 of the report presents temporal trend information for selected regions of Canada including the West Coast, Arctic, Great Lakes, Atlantic Coast and the St. Lawrence River system, and the overall findings for the Canadian environment are summarized in Section 4. Section 5 discusses temporal trend information on environmental concentrations of PCBs in other areas of the world.

2. Recycling and Redistribution of PCBs in the Environment

During the 1960s and early 1970s, the widespread use of PCBs and the lack of regulatory controls on releases to the environment resulted in new and ongoing releases to the atmosphere and aquatic systems. The use of PCBs in new products, machinery and electrical equipment was prohibited in Canada in 1980, but PCBs are still present in some older in-service equipment. Occasional spills from electrical equipment still occur and the Emergency Division of Environment Canada, Pacific and Yukon Region received ten reports of spills from pole top transformers in British Columbia in 1999. The average fluid volume loss in these incidents was approximately 100 litres, however, the PCB concentrations were low (50 to 100 mg/L). Upon identification, these spills were cleaned up immediately by the owner and the PCB contaminated oil in the transformer was replaced with PCB-free oil. B.C. Hydro is currently in the process of identifying the remaining in-service transformers containing low levels of PCBs. Another possible source of release to the environment are the old transformers containing PCB contaminated oil which are occasionally found during the investigation of abandoned contaminated sites (Mendoza, E., personal communication).

Current losses from these sources would be minor in comparison to losses which occurred prior to the introduction of regulations, and significant point sources have now been virtually eliminated. For example, Smith (1995) reported that point sources were no longer considered important in the mass balance of PCBs in the Great Lakes. Estimates from both the U.S. Environmental Protection Agency and the International Joint Commission suggested that point sources accounted for less than 10% of the total external loading, while dominant sources included atmospheric deposition and release from contaminated sediments.

PCB spills, improper disposal of waste PCBs, and numerous discharges to aquatic systems, which occurred prior to the introduction of government regulations, resulted in a large repository of PCBs in soils and sediments available for recycling and redistribution in the environment. The U.S. Environmental Protection Agency estimated that 40% of the total PCBs manufactured in the United States were lost to the environment in plasticizers, lubricating and heat transfer fluids, carbonless copypaper, and through improper disposal of fluids and equipment, dumping and accidental releases. It is expected that a similar percentage of the 40,000 tonnes of PCBs imported into Canada would also have been lost to the environment (Doug Wilson, personal communication). Tanabe (1988) estimated that approximately 35% of the global PCBs produced now exist in environmental reservoirs, such as soils, plants and aquatic systems.

The remobilization through volatilization of previously released PCBs in the environment results in 'outgassing' back to the atmosphere (Jones *et al.*, 1995). With decreased PCB emissions to the atmosphere from industrial sources, volatilization from soils and surface waters have now become important potential sources of PCBs to the atmosphere.

It was estimated that 90 to 95% of the PCBs present in soil when environmental concentrations in Asia, Europe, and North America peaked in the early 1970s, have since revolatilized and become available for long-range atmospheric transport to northern and Arctic areas (Gregor, 1994). Lee *et al.* (1998) reported that, climatic conditions determine the direction of flux, with PCBs being deposited to soil and vegetation during cool temperatures and volatilized from soil and vegetation during warm temperatures. Some studies attributed decreased PCB concentrations in soils in the United Kingdom (Alcock *et al.*, 1993) and the surface waters of Lake Superior (Jeremiason *et al.*, 1994) to the volatilization of previously deposited PCBs.

Lake Superior had no significant point sources of PCBs and it was estimated that atmospheric deposition accounts for approximately 90% of the PCB burden in this lake. However, mass balance calculations for Lake Superior indicated that volatilization (87-92%) rather than sedimentation (5-11%) was also the main source of loss from the lake (Baker and Eisenreich, 1990; Jeremiason *et al.*, 1994; Strachan and Eisenreich, 1988). According to Hornbuckle *et al.* (1994), whether there was a net loss or net gain of PCBs to Lake Superior depended on the season. This was supported by the findings of other researchers (Swackhamer *et al.*, 1988). One would expect volatilization to be dominant only during the warmer summer months, however, Hornbuckle *et al.* (1994) observed that, while a net loss of PCBs through volatilization occurred throughout most of the year, there was a net PCB deposition to the lake only during the spring. This was explained by the fact that the PCB flux depends on both water temperatures and vapour-phase concentrations, which drive the PCB exchange at the air-water interface in opposite directions. They estimated that the annual flux of total PCBs out of Lake Superior in 1992 was 250 kg/yr. Hoff *et al.* (1996) also studied the atmospheric loadings of PCBs to the Great Lakes and concluded that there was a net loss from the lakes to the atmosphere via volatilization.

PCBs associated with particulate material in the water column can become permanently removed from aquatic systems as a result of sedimentation and subsequent sediment burial. However, PCBs adhering to suspended solids are not always lost from the system as a result of permanent sediment burial. For example, Jeremiason *et al.* (1998) concluded that only 2 to 5% of the PCBs associated with settling solids in the water column of Lake Superior accumulated in the bottom sediments. The majority of the PCB was recycled in the benthic region just above the bottom sediments. Most of the PCBs in the water column, therefore, remained available for uptake into biota or volatilization to the atmosphere.

In addition, PCBs in bottom sediments can be remobilized to the water column (Gevao *et al.*, 1997; Oliver *et al.*, 1989; Bushart *et al.*, 1998), where they become available for recycling in the environment through uptake into biota or volatilization to the atmosphere. It was suggested that post-depositional mobility of PCBs in sediments favours the lower chlorinated congeners which tend to be more soluble and more volatile (Gevao *et al.*, 1997).

Atmospheric transport is acknowledged as the main route of global PCB movement and redistribution. The unexpectedly high PCB levels detected in some species of biota in Arctic regions, which were relatively devoid of direct PCB sources, have been attributed to processes known as 'cold condensation' and 'global fractionation'. PCB and other contaminants volatilize in the warmer temperate regions, move northward, and condense in the colder Arctic regions. A latitudinal fractionation is thought to occur due to the differences in the volatility of the different compounds. The less chlorinated and more volatile PCB congeners would be transported to farther distances than the more highly chlorinated congeners. The highly chlorinated and less volatile PCBs tend to condense relatively close to temperate regions with little being transferred to colder areas (Lead *et al.*, 1996; Wania and Mackay, 1993). The theory of 'cold condensation' is supported by the fact that sediment cores, obtained from lakes in high Arctic regions, indicated that PCBs did not appear in the Arctic environment until many years after they appeared in regions at lower latitudes. Similarly, the presence of higher proportions of the lower chlorinated PCB congeners in high Arctic regions supports the theory of 'global fractionation' (Muir *et al.*, 1996a).

3. A Review of Regional Information on Temporal Trends in Canada

This section provides a summary of temporal trend information for various regions in Canada. Existing information on temporal trends for PCB concentrations in the Canadian environment pertains primarily to the Great Lakes and Arctic regions. Information on temporal trends of PCB concentrations for the west and east coasts of the country was limited. The Great Lakes region received PCB contaminated discharges from many industrial facilities, but few point sources existed in Arctic areas. PCB releases from DEW line radar sites in the Arctic were substantial and very high PCB concentrations were found in their immediate vicinity. However, the impacts were localized and releases were considered to be small (approximately 1% of the total over a 30 year period) in comparison to the total burden of PCBs reaching Arctic areas (Gregor and Reimer, 1995). The majority of PCBs in Arctic regions were transported from global sources via long-range atmospheric transport, ocean currents and north-flowing rivers (Government of Canada, 1997).

3.1 West Coast

3.1.1 Sediments

Sediment cores collected in the Georgia Basin and Puget Sound regions showed similar PCB trends. A rapid rise in PCB concentrations was observed starting in the 1930 to 1940 horizon with maximum concentrations occurring in sediments in the 1950s and 1960s. Significant decreases in PCB concentrations observed in the horizons deposited since 1970 coincided with the introduction of stringent controls on the use and manufacture of PCBs. The detection of very low concentrations of PCBs in horizons (pre-1940s) deposited prior to their commercial manufacture was attributed to one or more of the following factors: bioturbation, bias in the age dating technique, and/or false positive results caused by interfering compounds (Macdonald and Crecelius, 1994).

In cores collected from the Strait of Georgia in 1990, the profile of PCB congener 77 matches that of OCDD and was consistent with atmospheric deposition patterns found elsewhere. Concentrations increased until the 1970s and then appeared to decrease in the Balenas Basin core and plateau in the Howe Sound core. Differences observed at the two sites may be due to differences in sources or to mixing within the cores (Macdonald *et al.*, 1992; Macdonald, personal communication).

In an attempt to assess the extent of long-range transport of contaminants in the Fraser Basin, Macdonald *et al.* (1999) collected sediment cores from lakes in the headwater systems above the influence of point sources. They detected very low concentrations of PCBs in the sediments, but observed that the PCB patterns in the cores were consistent with atmospheric deposition of contaminants. Macdonald (personal communication) observed that the cores indicated that PCBs entered the Fraser Basin aquatic systems in the 1930s and increased after this time. Concentrations seemed to have plateaued or decreased in recent years but, due to the low PCB concentrations combined with analytical sensitivity limits, a clear trend analysis was not possible. Cores from Kamloops lake differed somewhat from the other lakes in the study in that an industrial source of PCBs was also suggested. Cores indicated atmospheric deposition of PCBs and organochlorines to the lake in the early years with the appearance of an additional source following the construction of the pulp mill in 1965. PCBs (congeners 13, 15, and 37) were generated in the pulp chlorination process at the mill and discharged in wastewaters to the lake from 1965 until 1990, when chlorine bleaching was phased out (Macdonald *et al.*, 1999).

High concentrations of PCBs were detected in sediments at several sites in Vancouver and Victoria harbours in the late 1970s and early 1980s (Garrett, 1983). Although high PCB concentrations were still present in many areas of both harbours, concentrations in sediment samples collected in the late 1980s and early 1990s were generally lower than in earlier samples (Garrett, in preparation). The heterogeneity of the bottom sediments can cause the concentrations of PCBs in the surface layer of bottom sediments to vary widely in samples collected within metres of each other. Repeat sampling of sites can be especially difficult in harbours as access to sampling stations is often obstructed by the presence of large ships. As a result, variations in the location of sampling stations may occur between surveys. In addition, past dredging activity would have altered sediments, making the assessment of trends difficult. Complete dredging histories for many sites were not available.

Brewer *et al.* (1998) reported a declining trend for PCB concentrations in sediments from the Fraser River system. Substantial declines were observed in sediments from the Fraser River estuary, since 1985, and in the Brunette River watershed compared to levels reported 20 years ago.

3.1.2 Biota

3.1.2.1 Fish

Raymond *et al.* (1998) reported that peamouth chub collected from the Fraser basin in 1995 and 1996 contained lower PCB concentrations in liver tissue than did chub collected in the 1970s and 1980s.

Macdonald *et al.* (1999) collected fish from lakes in the headwater systems of the Fraser Basin above the influence of point sources. High PCB concentrations detected in the liver tissue of burbot from one of these lakes (Moose Lake) was unexpected considering the very low PCB concentrations in the sediments. While the source of the high PCBs to fish in this lake was not determined, one of possible sources considered was the melt-back of glacial ice and snow in warm years (Macdonald *et al.*, 1999; Blais *et al.*, 1998).

3.1.2.2 Birds

PCB concentrations in eggs collected from heron colonies in the Fraser estuary declined rapidly a few years after the introduction of strict regulations on the use of PCBs, and then fluctuated at lower levels from the early 1980s until 1994. Eggs collected in 1996, however, contained the highest PCB concentrations detected since 1983. The researchers stated that it was too early to determine whether this increase represented an increasing trend (Wilson *et al.*, 1999).

PCB concentrations in the eggs from seabird colonies in industrialized regions of coastal B.C. (Strait of Georgia and Fraser Estuary) have declined significantly since the restrictions on PCB use and manufacture were introduced in the 1970s. However, changes in PCB concentrations were much less apparent in eggs from bird colonies from remote areas such as the Queen Charlotte Islands. Storm petrel eggs maintained virtually unchanged levels of PCBs over a 20 year period. The authors observed that the continued presence of elevated PCB levels in offshore foraging species probably reflects the ongoing atmospheric dispersal of PCBs globally (Elliott *et al.*, 1997).

3.1.2.3 Terrestrial Mammals

Elliott *et al.* (1999) reported that PCB concentrations in the liver tissue of otters collected from the lower Columbia River between 1990 and 1992 were 10 fold lower than were concentrations measured a decade earlier.

3.1.2.4 Marine Mammals

Very little information was available on PCB trends in marine mammals on the west coast of Canada. Information on concentrations of PCB congeners in juvenile harbour seals from 1992 and 1996 was obtained but analysis was still in progress (Addison, personal communication).

High concentrations of PCBs were detected in killer whales from the Strait of Georgia, however, these animals are very long-lived and no trend information was available (Ross, personal communication).

3.2 Arctic Regions

Temporal trend data for Arctic species are limited and are often based on small sample sizes and few sampling locations. In many cases, the lack of standardized methodology and changing analytical methodologies over time makes the interpretation of the existing data difficult. The Canadian Arctic Contaminant Assessment Report (Government of Canada, 1997) and the Arctic Monitoring and Assessment Programme report (AMAP, 1998) recommend the design and implementation of temporal trend studies to analyze both abiotic and biotic samples from specimen banks from CACAR and from other Arctic nations (Bard, 1999).

3.2.1 Atmosphere

Elevated concentrations of PCBs in the Arctic environment were attributed to long-range atmospheric transport from industrialized regions in warmer areas. The analysis of snow samples from the Agassiz Ice Cap in 1993 revealed that PCB concentrations ranged from 1.2 to 6.7 ng/L over a 30 year period. There was little post-depositional PCB loss from the snowpack. However, it was not possible to identify long-term trends in PCB deposition to the ice cap due to the high variability in PCB concentrations in the samples. PCBs were not present in the snowpack prior to 1957, but low amounts appeared in glacial snow samples between 1957 and 1963 (9 $ng/m^2/year$). PCB deposition to glaciers increased significantly between 1963 and 1993 (406 ± 187 $ng/m^2/year$) (Gregor, 1994; Gregor *et al.,* 1995).

3.2.2 Sediments

Sediment cores revealed that PCBs were not present in high Arctic lakes until the 1960s (± 10 years), indicating a delay of approximately 20 years between PCB production and use and the appearance of these chemicals in high Arctic areas. In comparison, the PCB profiles in sediment cores collected from lakes in sub-arctic areas more closely followed the history of PCB production, with PCBs appearing in sediment horizons from the 1940s (Government of Canada, 1997; Muir *et al.*, 1996a).

Core samples collected from Lake Laberge in the Yukon indicated that peak PCB loadings occurred in the 1940s (Muir *et al.*, 1994). The profile of the PCB congeners changed with depth in the cores. In the older horizons, which corresponded to the 1945 to 1955 time period, higher chlorinated hexachlorobiphenyls dominated , while tri- and tetrachlorobiphenyls dominated in the more recent horizons. These findings differ from those of Muir *et al.* (1996a) who reported high proportions of the lower chlorinated compounds in Arctic lakes due to preferential atmospheric transport. Similarly, atmospheric and snow congener patterns were dominated by the lower chlorinated congeners (Stern *et al.*, 1997). This suggests that a local source of Aroclors was present in the Lake Laberge area in the 1940s and 1950s.

Cores from a lake in the Mackenzie River basin with undisturbed sediments indicated a slight increase in PCB concentrations over the last 30 years (Government of Canada, 1997; Graf Pannatier, 1996).

3.2.3 Biota

3.2.3.1 Fish

A comparison of organochlorine concentrations in round and lake whitefish in Lake Laberge over an 18 year period revealed decreases in some contaminants, such as DDT, but increases in others, such as PCBs and toxaphene (Government of Canada, 1997). The very high concentrations of PCB and other organochlorines in Lake Laberge fish may have been due, at least in part, to the higher trophic level species on which these top predator fish were feeding. Longer food webs were shown to result in higher PCB concentrations in predator fish species (Rasmussen *et al.*, 1990).

3.2.3.2 Birds

Migratory peregrine falcons breeding in the Canadian Arctic were sampled in 1982-1984 and 1991-1994, but no significant changes in PCB concentrations were observed in eggs over this time period. In addition, the proportion of clutches exceeding the 'critical PCB level' did not change over this time. Studies of falcons from Greenland and Alaska, however, indicated that PCB concentrations have decreased in these areas (Jarman *et al.*, 1992; Swen, 1994).

PCB concentrations in seabird eggs from Prince Leopold Island in Lancaster Sound declined by 69-86% between 1976 and 1987. Declining PCB concentrations were also noted in the liver of northern fulmar collected from Prince Leopold Island between 1975 and 1993. (Nettleship and Peakall, 1987; Noble and Elliott 1986; Muir *et al.*, 1999). Data for 1998 showed continued declines in PCB levels in Arctic seabirds (Braune, B., personal communication).

3.2.3.3 Marine Mammals

Ringed seal data from the western Arctic (Holman Island in the Northwest Territories) indicated that PCB concentrations declined significantly (approximately 40%) between 1972 and 1981 and then stayed relatively constant until 1991 (Addison and Smith, 1998; Addison, personal communication). Similarly, PCB concentrations in female ringed seals from Admiralty Inlet declined by approximately 50% between 1975/6 and 1983 (Muir *et al.*, 1988). However, PCB concentrations in female ringed seal blubber from Cumberland Sound at Baffin Island or Barrow Strait in Lancaster Sound did not decrease over six and nine year study periods, respectively (Muir, 1996).

No significant decreases in total PCB concentrations were detected in blubber samples collected from beluga whales in the Mackenzie delta region over a 10 year period (Addison and Brodie, 1973). Calculations based on models predicted that it would take 20 years, or two generations, before changes in PCB concentrations in beluga whales became obvious. This was due to the intergenerational transfer of PCBs and to their slow elimination rate (Hickie, 1995). Slightly higher PCB concentrations were detected in male narwhal from Lancaster Sound in 1994 than in 1982 (Muir, 1996). Similarly, Norstrom *et al.* (1988) reported that polar bears from the central Arctic contained higher PCB concentrations in 1983-1984 than in 1969-1971.

3.2.4 Humans

PCB concentrations in human milk samples collected from women living in southern areas of Canada have declined since the 1980s. No comparable information was available for women from Arctic regions (Newsome *et al.*, 1995), despite the fact that PCBs are among the contaminants of primary concern in the diets of Inuit people. Long-range atmospheric transport has resulted in high levels of PCBs in the higher trophic level Arctic species such as whales and seals, which are routinely consumed by Inuit people. Studies have shown that the PCB concentrations in human tissue were substantially higher in Arctic people than in southern Canadians. This difference reflects the higher consumption of high trophic level species for food by northern people. In addition, the concentration of organochlorine compounds were much higher in Inuit people from Nunavik in northern Quebec compared to southern non-aboriginal people. Newborn cord blood samples from some Inuit babies exceeded the concern level for PCBs and cord blood samples from Nunavik were twice as high as those from Inuit people from the Northwest Territories (Government of Canada, 1997; Dewailly *et al.*, 1989; 1992; 1993) .

3.3 Great Lakes

3.3.1 Atmosphere

Simcik *et al.* (1999) reported that gas-phase PCB concentrations decreased near Lakes Michigan and Erie between 1991 and 1997. Gas-phase concentrations near Lake Superior showed no trend over the same period, however, some individual PCB congeners increased. The authors suggested that global distillation may account for the difference between the Lake Superior site and the other sites. Since Lake Superior was the northernmost site sampled, and experienced the coldest temperatures, it is possible that global distillation could transport PCBs from more temperate locations to this site. Significant half-lives for individual PCB congeners ranged from 0.5 to 7.5 years compared to half-lives of 2.8 to 3.3 years for total PCBs.

3.3.2 Sediments

Lake Superior has not received significant point source inputs of PCBs and atmospheric inputs are thought to account for the bulk of the PCB loading (90%) to this lake. However, it was also reported that volatilization of PCB, rather than sedimentation, was the main source of PCB loss from Lake Superior. Whether there was a net loss of PCBs from the lake via volatilization or a net gain through atmospheric deposition varied with the season (Baker and Eisenreich, 1990; Jeremiason *et al.*, 1994; Strachan and Eisenreich, 1988; Swackhamer *et al.*, 1988; Hornbuckle *et al.*, 1994). Hornbuckle *et al.* (1994) concluded there was a net loss of PCB from the lake annually (250 kg/yr in 1992). Jeremiason *et al.* (1994) reported that PCB concentrations in Lake Superior water decreased between 1980 and 1992. Subsequent studies on sediment trap samples from Lake Superior indicated that PCB concentrations in settling solids had declined from 1984 to 1991. The authors concluded that only minor amounts of PCBs were permanently removed from the system by sediment burial, with the bulk being recycled in the benthic region (Jeremiason *et al.*, 1998).

PCB concentrations in the horizons of dated sediment cores collected in Lake Ontario and Lake Michigan reflected the decline in PCB use and manufacture through 1980, but little or no decrease was observed since this time (Oliver *et al.*, 1989; Eisenreich *et al.*, 1989; Golden *et al.*, 1993). Eisenreich *et al.* (1989) reported that PCB patterns in sediment cores from Lake Ontario paralleled the use of PCBs in the United States. Concentrations rose from 1950 to 1966/69, when they reached peak levels, and then declined in 1989.

PCB concentrations in suspended sediments in the Niagara River decreased from 650 ng/kg in 1979 to 150 ng/kg in 1991 (Kuntz, 1993).

3.3.3 Biota

3.3.3.1 Fish

Archived samples of whole lake trout (4 years old) collected from eastern Lake Ontario showed an 80% decline in PCB concentrations between 1973 and 1993 (Huestis *et al.*, 1997). Total PCBs and many congeners demonstrated a rapid drop in concentration between 1977 and 1981, followed by a more gradual decline and leveling off at present levels. All congeners except 169 declined 30% between 1977 and 1981. This early rapid decline was followed by a slower additional 50% decrease between 1981 to 1993. Between 1977 and 1993 there was a change in relative congener proportions. A slight increase in the proportion of hexa- and hepta- substituted congeners and a corresponding decrease in the proportion of penta- and tetra- substituted congeners was observed. Hexa- and penta- substituted PCBs accounted for about 70% of the total PCBs. There was no observable change in the overall proportion of tri-, octa- and nona- congeners, which accounted for less than 3% of the total PCBs measured (Huestis *et al.*, 1996). The analysis of archived lake trout samples collected between 1977 and 1993 indicated that coplanar PCB levels did not appear to be increasing over time in relation to levels of other PCB congeners (Huestis *et al.*, 1996).

Borgmann and Whittle (1991) reported that, although PCB concentrations in lake trout from Lake Ontario have decreased, the downward trend was not smooth. PCB concentrations decreased from 1977 to 1981, increased between 1982 and 1984 and then decreased again in 1985. The increases in PCB concentrations in 1981 to 1982 were not observed in fish from Lake Michigan, Huron and Superior (DeVault *et al.*, 1986), suggesting a phenomenon unique to Lake Ontario. The authors suggested that the deviations from the overall downward trend were related to prey dynamics and changes in diet. PCB concentrations in trout were observed to decline one to two years after major die-offs occurred in alewife (primary food source). It was estimated that the half-life of PCBs in lake trout in Lake Ontario was approximately 10 years.

PCB concentrations in fish from Lake Michigan declined in all seven species examined (lake trout, rainbow trout, brown trout, chinook, coho, alewife, and bloater chub) after the introduction of controls on PCB use and release, but then appeared to stabilize in the mid- to late-1980s. PCB concentrations in chinook and coho salmon appeared to increase since the late 1980s, although all species contained PCB levels well below those detected in the 1970s (Stow *et al.*, 1995). The authors suggested that the apparent stabilizing of PCB concentrations in the other fish species was likely due to the large pools of PCBs that are being recycled in the environment. Atmospheric input was shown to be an important PCB source to Lake Michigan and the sediments are known to contain a large PCB reservoir. The authors suggested that the apparent increases in the PCB concentrations in chinook and coho were probably the result of changing growth dynamics caused by alterations in the food web. As alewife (important prey species) availability decreased there was a decline in the growth of chinook and coho. This could have resulted in the increased PCB concentrations in these species. Changes in prey PCBs would not be reflected as quickly in lake trout as in the slower growing and longer-lived species such as coho and chinook salmon. Median PCB concentrations were estimated to be only slightly below the FDA action level of 2 mg/kg wet weight. Stow *et al.* (1995) suggested that, in order to reduce PCB concentrations further, it may be necessary to address management practices to maximize fish growth rates. For example, Jackson (1996) stated that any changes occurring within a lake that would alter forage fish growth rates or standing stocks, would also affect PCB concentrations in sport fish species. Fisheries management may be a possible mechanism to further reduce PCB concentrations in sport fish.

Due to the high variability in PCB concentrations among individual fish, it was suggested that it may be necessary to collect very large numbers of fish in future surveys in order to detect the small changes in overall PCB concentrations. Stow *et al.* (1995) concluded that declines in PCB concentrations were probably still occurring in the Lake Michigan system overall; however, they may have been occurring at a rate too slow to be detected with the data available. Smith (1995) observed that, since PCB and DDT concentrations in Lake Michigan lake trout and coho salmon varied at about the same time and the same magnitude, the same force must be driving the changes in both chemicals. For this reason, it was unlikely that the observed changes in concentrations were related to a point source or to changes in loadings or ambient concentrations but, more probably, to internal forces such as food chain dynamics. PCB concentrations in Lake Michigan lake trout were observed to drop significantly in years following major declines in adult alewives. A similar observation was made for Lake Ontario lake trout (Borgmann and Whittle, 1991).

In addition, Smith (1995) stated that the current data did not support the theory that PCB concentrations in Lake Michigan lake trout were approaching a new equilibrium. He argued that a new equilibrium for PCBs is not possible as there are no significant new sources or any natural sources of PCBs. PCBs are recycled in the environment and atmospheric deposition and other non-point sources are important sources to the Great Lakes. However, PCBs are also undergoing continual irreversible loss from the system through processes such as burial and degradation. He conceded that it is possible that we are now observing extremely slow depuration, however, he maintained that this is fundamentally different from no depuration or 'equilibrium'. Smith (1995) supported the theory, proposed by Stow *et al.* (1995), that current sampling rates are not adequate to detect the presence or absence of changes in PCB concentrations in Lake Michigan trout.

PCB concentrations in spottail shiners in the Niagara River decreased by a factor of five between 1975 and 1991, however, concentrations still exceeded the 100 ng/kg objective which was considered to be a threshold level for the protection of fish-eating birds (Suns *et al.,* 1991).

3.3.3.2 Birds

Stow (1995) reported that PCB concentrations in Great Lakes herring gull eggs decreased following the introduction of regulations on the use and manufacture of PCBs, however, concentrations in eggs from Lakes Superior, Michigan, Huron and Ontario appeared to have stabilized or slightly increased in recent years. Only the herring gull colonies on Lake Erie appeared to be experiencing a continuing decline in PCB concentrations.

Change point analysis was useful in pinpointing the specific year when changes in long term trends occurred and also in establishing the statistical significance of these changes. Pekarik and Weseloh (1998) analyzed temporal trends for herring gull eggs from 13 Great Lakes colonies using change point analysis. The results showed that a logarithmic decline of PCBs in herring gull eggs from western Lake Ontario was slower from 1987 to 1995 than from 1974 to 1986. However, continued decreases in PCB concentrations were observed at both Lake Ontario colonies. PCB levels in herring gull eggs have shown no temporal trend in the Green Bay Lake Michigan colony since 1976 or in Lake Superior colony since the mid-1980s. Analysis of data up to 1995 for the Niagara River colony indicated that PCB concentrations had not declined since the mid-1980s, however, the addition of 1996 data demonstrated that the decline in PCB concentrations in eggs from this colony was continuing at the previous rate. PCB levels in herring gull eggs collected at the other eight colonies continued a logarithmic decline in recent years at the same rate as, or faster than, in previous years. The findings of Pekarik and Weseloh (1998) agreed with those of Stow (1995) for Lakes Erie and Superior but not for Lakes Ontario, Michigan and Huron. Both authors reported that PCB levels were declining at steady rates in Lake Erie but had stopped declining in Lake Superior.

Hebert *et al.* (1997) reported that PCB concentrations in Lake Ontario herring gull eggs were influenced by weather conditions in the region. In years with cold weather, PCB concentrations in eggs were greater because the proportion of fish in the herring gull diet increased as a result of alewife overwinter mortality. This emphasized the importance of food web interactions in regulating annual PCB concentrations in eggs. The authors concluded that PCB levels in herring gull eggs from Lake Ontario continued to decline but at a slower rate than was observed in the 1970s and early 1980s. PCB levels in herring gull eggs from other Great Lakes also showed relationships with the weather, but the significance of the relationship varied among the colonies (Hebert *et al.*, 1998).

Fox *et al.* (1998) compared the concentrations of PCBs and other contaminants in the pooled livers of adult herring gulls sampled at eight Great Lakes colonies and at one reference colony on the Atlantic coast. Between 1974 and 1993, PCBs declined in 7 of the 8 colonies. The largest decreases were observed before 1985. The highest concentrations detected in the 1990s occurred in the Middle Island and Saginaw Bay colonies. No decreases in PCB concentrations were observed in the Middle Island herring gull colony over this time period.

Ryckman *et al.* (1998) reported that PCB concentrations declined in the double-crested cormorant eggs collected from the Canadian Great Lakes, Lake Nipigon and Lake of the Woods between 1970 and 1995. Significant declines were observed at colonies located on Lake Ontario, Lake Superior, Georgian Bay and North Channel. However, PCB levels in eggs from the Lake Erie colony remained stable.

PCB concentrations in bald eagle eggs from Lake Erie declined significantly between 1974 and 1994, however, concentrations in plasma collected from eagle chicks at Lake Erie did not change significantly between 1990 and 1996.

Insufficient information was available to determine temporal trends in plasma PCB concentrations in chicks collected from other lakes in the area. Donaldson *et al.* (1999) suggested that the short time period examined, combined with the numerous sources of variation, could prevent the detection of a slow rate of PCB decline in chick plasma.

Red-breasted merganser eggs were collected from nests in the Great Lakes area in 1977, 1978, and 1990. Concurrent analysis of archival and new samples demonstrated that PCBs decreased by approximately 60% between 1977 and 1990 and by 81% between 1975 and 1990 (Stromberg *et al.*, 1993).

Harris *et al.* (1993) reported that median PCB levels in Forster's tern eggs from Green Bay, Lake Michigan decreased by 67% between 1983 and 1988.

3.4 Atlantic Coast/St. Lawrence

3.4.1 Atmosphere

Brun *et al.* (1991) reported on PCB concentrations in wet precipitation samples collected from three locations in Atlantic Canada between 1981 and 1989. Although concentrations decreased over this time period, the authors noted that historical data was insufficient to determine whether the PCB regulations and restrictions introduced in the 1970s affected the atmospheric transport and loading of PCBs to the Atlantic region.

3.4.2 Sediments

As a result of the regulations on PCBs which were introduced in the 1970s and the ban on PCB imports into Canada as of 1980, PCB inputs to the St. Lawrence River have decreased by 80% since 1985 (Coakley *et al.*, 1993). Analysis of sediment samples indicated that PCB concentrations have decreased since the 1970s (Lebeuf *et al.*, 1995; Cossa, 1990). Sediment cores recently collected from the St. Lawrence estuary contained maximum PCB concentrations in the 1968 to 1971 horizons, but concentrations decreased since this time and appeared to have leveled off. Although, no significant changes in PCB concentrations were observed in horizons deposited since 1988, the analytical variability (10 to 20%) and the possibility of mixing due to bioturbation in the surface layers, complicated the interpretation of trends in the more recent horizons. Cores were also collected in the Gulf of St. Lawrence, however, the low sedimentation rates in this area make it more difficult to determine clear trends, particularly over the last 10 to 20 years (Lebeuf, personal communication).

3.4.3 Biota

3.4.3.1 Fish

Harding *et al.* (1997) reported that PCB concentrations in pelagic fish (herring, blueback and gaspereau) declined steadily from the late 1970s to the 1990s. PCB concentrations in liver tissue from cod collected in the southern Gulf decreased between 1977 and 1985 (Misra and Nicholson, 1994). Declining PCB concentrations were also observed in eel from the St. Lawrence area. Between 1982 and 1990, the PCB concentrations in muscle tissue of adult eels decreased by 68% (Hodson *et al.*, 1992; 1994; Castonguay *et al.,* 1989).

3.4.3.2 Plankton

Harding *et al.* (1997) reported that PCBs may have declined by a couple of orders of magnitude in phytoplankton collected from the Gulf of St. Lawrence between the early 1970s and the mid-1980s.

3.4.3.3 Birds

Eggs were collected from seabird colonies (cormorants, petrel and puffin) in eastern Canada at 4 year intervals from 1968 to 1984. The data indicated that PCB concentrations declined significantly in eggs of all species from the Bay of Fundy during the 1970s, however, declines in PCB levels were only observed in petrels from the Atlantic coast of Newfoundland. PCB concentrations in cormorant eggs decreased significantly since 1972 but, between the mid-1970s and 1992, concentrations stabilized at low levels. In general, studies indicated that PCB concentrations in marine birds in the near shore ecosystem of the North American continental shelf decreased as a result of the introduction of regulations on the manufacture and use of PCBs (Government of Canada, 1998; Pearce *et al.*, 1989; Elliott *et al.*, 1992). With the reduction of PCB releases to the environment, long-range transport of PCBs has become an increasingly important source on the East Coast (Burgess, N., personal communication).

Significant decreases in PCB concentrations were observed in double-crested cormorant eggs collected from the Gulf of St. Lawrence and the estuary between 1972 and 1992 (White and Johns, 1997). Similarly, Elliott *et al.* (1988) reported that significant declines in PCBs and other organochlorines were recorded in gannet eggs from Bonaventure Island in the Gulf of St. Lawrence between 1968 and 1984. However, unlike the Great Lakes region, no significant declines occurred in PCB concentrations in herring gulls from a colony in the St. Lawrence estuary between 1976 and 1984 (Pearce *et al.*, 1989).

3.4.3.4 Marine Mammals

PCB concentrations declined by approximately 55% in harp seals from the Gulf of St. Lawrence between 1971 and 1982 (Addison *et al.*, 1984). Similarly, PCB concentrations in male harp seal from Les Escoumins in the St. Lawrence Estuary declined 1.7 fold between 1982 and 1989 (Beck *et al.*, 1994). Significant declines in PCB concentrations were also observed in male Beluga whales from the Gulf of St. Lawrence between 1982-1985 and 1993-1994. According to the authors, this trend paralleled trends observed in seals, eels and seabirds in the St. Lawrence estuary during the 1980s (Muir *et al.*, 1996b).

PCB concentrations (Aroclor 1254) in grey seals from Sable Island, Nova Scotia remained relatively steady from 1976 until 1988 and then began to decline (Addison *et al.,* 1998; Addison, personal communication).

Total PCB concentrations in porpoises from the Bay of Fundy decreased by over 4 fold between the 1970s and 1991 (Gaskin *et al.*, 1983; Westgate *et al.*, 1997).

4.0 A Summary of Information on PCB Temporal Trends in the Canadian Environment

4.1 Atmosphere

Although long-range atmospheric transport and deposition was recognized as an important source of PCBs to the environment, particularly to Arctic regions, very little temporal trend information on PCBs in the atmosphere was available.

Between 1991 and 1997, gas-phase PCB concentrations decreased near Lakes Michigan and Erie but not Lake Superior (Simcik *et al.*, 1999), and wet precipitation samples collected in Atlantic Canada contained lower PCB concentrations in 1989 than in 1981 (Brun *et al.,* 1991).

Studies on polar ice caps could not identify long-term trends for PCB deposition. However, PCBs first appeared in snowpack in 1957 and PCB deposition rates were much greater between 1963 and 1993 than between 1957 and 1963 (Gregor, 1994; Gregor *et al.*, 1995).

4.2 Sediments

PCB trend data for Canadian sediments was very limited. However, sediment cores collected from Georgia Basin on the west coast (and Puget Sound near Seattle, Washington) (Macdonald and Crecelius, 1994; Macdonald *et al.*, 1992), lakes located above the headwaters in the Fraser Basin (Macdonald *et al.*, 1999), Lakes Ontario and Michigan (Oliver *et al.*, 1989; Eisenreich *et al.*, 1989; Golden *et al.*, 1993), and the St. Lawrence Estuary (Lebeuf, M. personal communication) generally reflected past PCB use. Maximum PCB concentrations were found in 1950/60s horizons with significant decreases throughout the 1970s.

Sediment cores collected from lakes in remote areas of Canada indicated that the onset of elevated PCB deposition in the high Arctic (1950-1960s) was delayed in comparison to lakes located at lower latitudes (1930-1940s). This delay in the appearance of PCBs in Arctic areas, combined with observations of higher proportions of the lower chlorinated PCB congeners in sediment cores, supports the theory of global fractionation (Muir *et al.*, 1996a).

The interpretation of PCB trends in the more recent surface layers of sediment cores can be difficult. The variability of analytical results, combined with the lower concentrations of PCBs which are often found in recently deposited sediments, makes trends less obvious in the surface horizons of sediment cores. In addition, evidence of PCB redistribution through bioturbation within some cores was observed. The mixing of sediments as a result of bioturbation can result in a more even distribution of PCB concentrations in recent horizons.

4.3 Biota

In most regions studied environmental PCB concentrations in biota peaked in the1960s and declined rapidly through the 1970s and early 1980s, following the introduction of stringent regulations on PCB use. For example, PCB concentrations decreased by 80% between 1973 and 1993 in lake trout from eastern Lake Ontario (Huestis *et al.*, 1997), 69 to 86% in seabird eggs from Lancaster Sound between 1976 and 1987 (Nettleship and Peakall, 1987), 81% in red-breasted merganser eggs from the Great Lakes area between 1977 and 1990 (Stromberg *et al.*, 1993), 40% in ringed seals from the North West Territories between 1972 and 1981 (Addison and Smith, 1998), and 55% in harp seals from the Gulf of St. Lawrence between 1971 and 1982 (Addison *et al.*, 1984).

The largest decreases in environmental concentrations of PCBs were observed in areas where the environmental levels were high in the 1970s due to nearby point sources discharges. Decreases in less contaminated areas (areas removed from point sources and urban areas) were less apparent. For example, Elliott *et al.* (1997) noted significant decreases in PCB concentrations in seabirds collected from the Strait of Georgia between 1968 and 1990, however, declines were less apparent in remote offshore colonies. PCB concentrations in storm petrels (offshore foraging species) have remained virtually unchanged over a 20 year period, probably due to the ongoing atmospheric dispersal of global levels of PCBs.

Although PCB concentrations remained elevated in some localized regions of Canada, the rapid decline in PCB concentrations observed in the 1970s indicated that the regulations were very effective in reducing environmental levels of PCBs. However, declines in environmental PCB levels since the 1980s have been far less apparent. For example, PCB concentrations in Great Lakes lake trout decreased rapidly between 1977 and 1981, but decreased at a much slower rate between 1981 and 1993 (Huestis *et al.*, 1997). Similarly, several researchers reported that decreases in PCB concentrations in herring gull eggs from Great Lakes colonies were slower since the mid-1980s than between the 1970s and the mid-1980s (Pekarik and Weseloh, 1998; Stow, 1995; Fox *et al.*, 1998). Addison and Smith (1998) observed that, although PCB concentrations in ringed seals decreased significantly between 1972 and 1981, they appeared to have stayed constant since that time.

These findings led many researchers to suggest that the environmental levels of PCBs have leveled off and have either stopped decreasing or are decreasing at such a slow rate that they appear to have stabilized. Some researchers suggested that environmental concentrations may be reaching a new equilibrium. Others argue that PCBs have not stabilized and are continuing to decline in most areas, but that declines are slower and, therefore, more difficult to detect with the data currently available. For example, Smith (1995) suggested that the analysis of PCB concentrations in Lake Michigan lake trout does not support the theory that PCB levels are approaching equilibrium and that a first order decline model may not be appropriate for assessing the decline of PCB levels in fish. He argued that the PCB concentrations could not be at equilibrium since PCBs are continually being lost through degradation and irreversible burying by sedimentation, while new PCB releases to the environment are very minor. Smith (1995) suggested that, since the variations in PCB and DDT concentrations in Lake Michigan lake trout over time were similar in both time and magnitude, the concentrations of these chemicals in fish were being controlled by the same factor. For this reason, it was more likely that internal forces such as food chain dynamics were influencing the PCB and DDT concentrations in fish, rather than changes in loading or an increase in ambient PCB environmental concentrations. Smith (1995) conceded that it is possible that current rates of depuration are extremely slow, however, he maintained that this is fundamentally different from no depuration (equilibrium).

Some researchers reported that recent PCB concentrations were higher in some areas than were concentrations observed in the 1980s, but have not attributed these recent increases to new sources or increased ambient environmental levels. For example, Borgmann and Whittle (1991) suggested that observed deviations from the overall downward trend for PCB concentrations in lake trout from Lake Ontario between 1977 and 1985 were related to prey dynamics and changes in diet (primarily alewife). Stow *et al.* (1995) reported that PCB concentrations in chinook and coho salmon from Lake Michigan appeared to have increased slightly since the late 1980s, while PCB concentrations in other fish species appeared to have stabilized. They attributed the apparent increase in PCB levels in chinook and coho salmon to changing growth dynamics caused by alterations in the food web. In addition, chinook and coho salmon are relatively short-lived compared to many other species, and would likely reflect changes in prey PCB levels more quickly than other longer-lived species. Stow *et al.* (1995) concluded that PCB concentrations were probably still declining in Lake Michigan, but at a rate too slow to be detected with the data available. The authors suggested that it may be necessary to collect very large numbers of fish in order to detect subtle changes in overall PCB concentrations.

Stow (1995) reported that while the PCB concentrations in herring gull eggs from other Great Lakes were declining or had stabilized, concentrations in eggs from Lake Superior and Lake Huron had increased slightly in recent years. Stow (1995) did not interpret the apparent increases in herring gull PCB concentrations in Lakes Superior and Huron as an indication of a sustained upward trend in PCB concentrations, but as evidence that the concentrations have ceased to decline or are now declining so slowly that the effects of noise (sampling error, random variation, analytical variability) are visible. Hebert (1998) states that it is necessary to improve our ability to separate noise from the underlying trend information in data sets if we are to continue using seabirds as environmental trend monitors. Hebert (1998) reported that local weather patterns can obscure temporal trends. These authors concluded that winter severity and overwinter migration affected the extent to which herring gull eggs from northern Great Lakes colonies reflected local contaminant bioavailability. This was especially true for Lake Superior and northern Lake Huron as herring gulls from these colonies tend to overwinter in the more highly contaminated southern areas of the Great Lakes. This could explain the apparent increases in PCB concentrations reported for these colonies by Stow (1995). Some methods of data analysis were particularly successful in identifying subtle temporal trends. In contrast to the findings of Stow (1995), who reported that PCB concentrations continued to decline only in the herring gull colony at Lake Erie, Pekarik and Weseloh (1998) reported that change point analysis of PCB concentrations in herring gull eggs from the Great Lakes revealed declining PCB concentrations in all colonies examined with the exception of those located at Lake Superior, Green Bay on Lake Michigan, and the Niagara River (up until 1995). Increasing concentrations were not observed in any of the colonies.

Wilson *et al.* (1999) observed that PCB concentrations in eggs collected from the UBC heron colony in the Fraser Estuary near Vancouver in 1996, were higher than concentrations detected in eggs collected in all other years since 1983. However, the authors stated that it was too early to explain the higher PCB concentrations in herons from the UBC colony.

The importance of adequate sample sizes and sampling period was emphasized by the findings of Pekarik and Weseloh (1998). These authors noted that, for the Niagara River herring gull colony, analysis of data to 1995 indicated that PCB concentrations were no longer decreasing, however, the addition of the 1996 data revealed that PCB concentrations were continuing to decline in this colony.

Further reductions in environmental concentrations will likely occur very slowly. It was suggested that reductions in PCB levels will be especially slow in humans and other biota with long life spans. The persistence of PCBs in biota results in body burdens reflecting PCB exposure from the past as well as the present. In addition, PCB transfer from parent to offspring was shown to be significant (Harrad *et al.*, 1994; Muir *et al.*, 1996b; Duinker and Hillebrand, 1979; Tanabe *et al.*, 1982).

Predator-prey dynamics appeared to be important in determining PCB concentrations in predator fish species and this has led some researchers to suggest fisheries management practices as a possible way of reducing PCB concentrations in sports fish in the Great Lakes (Jackson, 1996). Factors controlling forage fish growth rates or standing stocks will also affect PCB concentrations in sports fish. The possible effects of stocking lakes with certain species has been discussed (Jackson, 1996) and

Rasmussen *et al.* (1990) warned that stocking lakes with *Mysis* may result in higher PCB levels in fish by lengthening the food web and allowing for greater trophic transfer and possibly by functioning as a vector for the transport of PCB from sediments (Bentzen *et al.,* 1996).

Some researchers suggested that the more toxic coplanar (non-ortho) PCB congeners were declining less rapidly in the environment than were the other PCB congeners (Metcalf and Haffner, 1995). Hebert *et al.*, 1999 reported that the ecological half-lives for the non-ortho substituted congeners were longer than those for their respective homologues. It was suggested that the decreased volatility of non-ortho congeners and increased hydrophobicity of these congeners in comparison to the ortho-substituted congeners may account for longer half-lives in the environment (Bidleman, 1984; Dunnivant and Elzerman, 1988; Dunnivant *et al.*, 1988; Dunnivant *et al.*, 1992) Hesselberg *et al.* (1991) reported that the analysis of archived lake trout from Lakes Michigan and Ontario indicated that some of these AHH active PCB congeners (coplanar) declined at a slower rate than did total PCBs, but the decline was inconsistent for a given congener between lakes. For example, in Lake Michigan fish, PCB congener 77 declined 8% compared to 70% for total PCBs over an 11 year period while, in Lake Ontario fish, both PCB congener 77 and total PCBs declined by 60%. A study on PCB congener patterns in lake trout eggs by Mac *et al.* (1993) showed no significant enrichment of non-ortho congeners and the authors concluded that the concentrations of the individual PCB congeners were declining at similar rates. The analysis of archived lake trout samples collected from the Great Lakes between 1977 and 1993 showed no evidence of increased proportions of coplanar PCB congeners in relation to other congeners according to Huestis *et al.*, 1997, although from 1977 to 1993 there was a slight increase in the proportion of hexa- and hepta- substituted congeners and a corresponding decrease in the proportion of penta- and tetra- substituted congeners (Huestis *et al.*, 1996). Similarly, a study on herring gull eggs in the Great Lakes by Turle *et al.* (1991) reported that the proportions of monochloro- through pentachloro-substituted PCBs decreased over time. There was little change in the proportion of hexachloro- substituted congeners, while the proportions of heptachloro- to decachloro-substituted PCBs increased. Hebert *et al.*, 1999 also noted that PCB accumulation patterns in herring gull eggs from Lake Ontario and from Green Bay, Lake Michigan changed through time. Herring gull eggs collected in recent years contained a greater proportion of the more highly chlorinated PCB congeners. In Lake Ontario, this was attributed largely to a decrease in the source of the lower chlorinated congeners after 1976. However, in both colonies, the differences in the physicochemical properties of the individual congeners were thought to affect the removal processes such as sedimentation and volatilization, therefore, affecting the availability of specific congeners for biological uptake. In general, the ecological half-lives of the individual PCB congeners increased with the degree of chlorination.

Metcalfe and Metcalfe (1994) examined PCB congener accumulation in the Lake Ontario food web. They reported that there was a greater proportion of penta- to hepta- congeners at the upper trophic levels and concluded that the highly chlorinated PCBs would persist in the Great Lakes food webs.

4.4 Humans

Declines in PCB concentrations in human tissues also occurred over the last two decades. Fensterheim (1993) reported that PCB levels in the human diet have declined to less than 1% of the levels reported in the early 1970s. In addition, the percentage of people with more than 1 mg/kg of PCBs in adipose tissue declined from 62% in 1972 to 2% in 1984. However, PCB concentrations in the diet of Inuit people in the Arctic are of concern due to their greater reliance on high trophic level species as food. PCB concentrations in Inuit people from northern areas were much higher than those detected in southern Canadians (Dewailly *et al.*, 1989; 1992; 1993).

5. A Review of Current Information on PCB Temporal Trends in Other Areas of the World

Temporal trends in environmental levels of PCBs observed in the United States and many other areas of the world were similar to those reported in Canada.

In the highly contaminated Hudson River in the United States, PCB concentrations in sediments decreased in response to reduced discharges from local electrical equipment manufacturing plants (Kennish, 1992). Fowler (1990) reported that PCB levels in striped bass from the Hudson River decreased between 1978 and 1981, however, the decrease between 1978 and 1979 was greater than that between 1979 and 1981. Fowler (1990) concluded that this was indicative of a slowing of the environmental depuration process.

Stout (1986) reviewed various monitoring programs in the United States conducted throughout the 1970s and 1980s. They noted that PCB levels in Dover sole from the Southern California Bight declined by more than an order of magnitude between 1972 and 1981. Similarly, PCB concentrations in mussels near the Los Angeles County sewer outfall decreased 10 fold between 1971 and 1978, but then increased again in 1979.

Over a 15 year time period (1977 to1992) PCB concentrations in mussels collected under the California State Mussel Watch program decreased at one quarter of the stations sampled. In general, the stations with high PCB concentrations initially were more likely to show a decreasing trend than were the stations that had relatively low concentrations to start. The PCB concentrations at stations with relatively low concentrations in the 1970s tended to remain relatively constant over time (Stephenson *et al.*, 1995). Between 1980 and 1996, PCB concentrations decreased in transplanted mussels in the San Francisco Estuary. Significant declines were noted between 1980 and 1983, however, the concentrations have not continued to decline into the 1990s (Gunther *et al.*, 1999).

Laurenstein (1995) compared mussels and oysters from archived samples collected under the Mussel Watch program in the 1970s with samples collected more recently under the NOAA National Status and Trends (NST) program. They reported that PCB concentrations were higher in 1986 and 1987 than in the 1970s, but levels had declined between 1986 and 1992. Similar results were reported by O'Connor (1996), who found that PCB concentrations had declined significantly in oysters and mussels between 1986 and 1993, and by Baliaeff *et al.* (1997) who reported that PCB concentrations in shellfish had decreased between 1986 and 1994 at all but two sites on the California coast.

In Buzzard's Bay, Massachusetts PCB concentrations decreased in common terns between 1971 and 1981 (Nisbet and Reynolds, 1984) but not in brown pelican from South Carolina between 1969 and 1977 (Blus, 1982). Data for various species of birds from Texas collected between 1965 and 1985 showed that PCB concentrations in most species of birds had decreased over this time period (Mora, 1995).

PCB concentrations in mussels from the French Mediterranean declined by nearly an order of magnitude between 1972 and 1975, however, concentrations increased in mussels collected in 1980 and 1981 (Fowler, 1990). Sole *et al.* (1994) reported that PCB concentrations in mussels from the western Mediterranean showed no significant decline between 1980 and 1992. However, a relative decrease in the lower chlorinated congeners was noted and tentatively attributed to environmental decay of the less persistent congeners combined with a lack of recent inputs. Tolosa *et al.* (1997) reported that, although decreases in coastal areas of the Mediterranean have been noted since regulations on PCB use have been introduced, declines in PCB concentrations in the open ocean were not as apparent.

The reduction of PCB loading in Finland also resulted in decreasing environmental concentrations of PCBs. Monitoring of northern pike in large lakes in Finland over a 25 year period showed significant decreases in this species since the 1970s. Recent concentrations were approximately 25% of those detected in pike 25 years ago (Korhonen *et al.*, 1997).

Moss collected from remote sites in Norway showed declines in PCB concentrations at all sites between 1977 and 1990. Temporal changes in congener patterns were also noted. In southern Norway, the higher chlorinated hexa- and hepta-congeners decreased to a greater extent than in the north. The authors noted that this was likely a result of differences in recycling of the various congeners through the environment according to their volatility, and may support the theory of global fractionation. It was suggested that the lower chlorinated congeners were revolatilizing and being transported to still colder areas (Lead *et al.*, 1996).

At some locations in the Baltic Sea, declining PCB concentrations were observed although environmental levels of PCBs were still considered to be unacceptably high. PCB concentrations in white-tailed eagles from the Baltic Sea declined significantly between the 1970s and 1980s (Korhonen *et al.*, 1997), as did the PCB concentrations in the eggs of guillemot between 1969 and 1980 (Bignert *et al.*, 1995). A Swedish national monitoring program demonstrated an annual decrease in PCB concentrations in herring in the Baltic Proper and the Bothnian Sea by approximately 5% and by approximately 10% in the Bothnian Bay (Bignert *et al.*, 1995). PCB concentrations in both marine and freshwater species of fish in Swedish lakes and the Baltic area were significantly lower in 1995 than in the late 1970s and early 1980s (Bignert *et al.*, 1998). Haahti and Perttila (1988) reported that PCB concentrations in cod and herring decreased between 1979 and 1986 at all sampling locations in the Finnish Sea areas of the Baltic Sea.

Marine mammals from several areas of the world have exhibited decreased PCB concentrations since the 1970s. However, blubber samples obtained from male striped dolphin off the Pacific coast of Japan showed no change in PCB concentrations between 1978-1979 and 1986. The authors noted that temporal trends were occurring more slowly in the remote ocean areas than in coastal areas (Loganathan *et al.*, 1990).

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