

*An Assessment of Historical PCB
Contamination in Arctic Mammals*

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The Arctic Region (AMAP, 2004)

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1. Introduction

In recent years, climate change has been recognized as a significant threat throughout the world and especially in the Arctic, which is already experiencing disproportionately greater effects related to a warming climate than much of the rest of the world. While this issue is certainly deserving of our attention, many other environmental threats to the Arctic region are often overlooked, in particular environmental contamination from heavy metals, persistent organic pollutants (POPs), radionuclides, and hydrocarbons.

These contaminants are highly toxic, and severely threaten the viability of both human and animal populations in the Arctic. Due to global air and water circulation patterns, many of these contaminants emitted from industrialized areas throughout the Northern hemisphere end up in the Arctic, a phenomenon often referred to as the “Arctic sink”. Once there, they are taken up by and concentrate within individual Arctic organisms over time (*bioconcentration*). Because organisms in higher trophic levels accumulate all the contaminants stored in their prey, contaminant concentrations in organisms increase the higher in the food web they are (*biomagnification*).

Studying historical trends of these contaminants in different organisms could reveal significant information about whether and when the health of these organisms have been compromised by contaminant exposure, about the processes of bioconcentration and biomagnification, and could enable projections about future trends. Furthermore, because much of the Arctic region is far removed from industrial sources of contaminants, biota and physical media from this area could be used to establish global “background” levels of contaminants unaffected by local sources. Because transport of contaminants to the Arctic and their incorporation in Arctic ecosystems takes a long time, it is unlikely that these organisms can serve as the “canary in the coal mine” for contaminants, an immediate indicator when global burdens have been exceeded. Rather, they can potentially serve as “recorders” of past global production and emission trends of these pollutants (MacDonald *et al.*, 2005), which can be otherwise difficult to model (e.g. Breivik *et al.*, 2002).

This study focuses on polychlorinated biphenyls (PCBs), a class of POPs. PCBs are considered to be “legacy” POPs because much is known about these environmental contaminants while their use and presumably environmental burdens have decreased dramatically in recent decades (Braune *et al.*, 2005). A wealth of scientific literature already exists pertaining to levels and physiological effects of PCBs from individual species and human communities in various locations throughout the Arctic at different times. However, relatively few attempts have been made to compile this data altogether, and most of these attempts have rather limited scopes and lack comprehensiveness and any historical data or analysis. This study attempts to build a comprehensive review of PCB data from Arctic mammals, with a particular focus on investigating and evaluating historical trends from throughout the circumpolar Arctic. Additionally, by comparing to historical PCB production data, it attempts to assess how long it takes PCBs to be transported to and incorporate into the Arctic ecosystem, and whether significant landmarks in the use of PCBs (i.e. legislation) are evident in the historical levels in Arctic mammals.

Section 2 provides background information on PCBs, their transport to the Arctic, and their interactions with Arctic ecosystems. Section 3 outlines the methodology used in this review. The results are presented in section 4 and evaluated in section 5. Finally, possible improvements to this study are presented in section 6, and section 7 is a conclusion.

2. Background

2.1. PCBs

2.1.1. Chemistry

Polychlorinated biphenyls (PCBs) are a class of 209 different congeners, each containing the structural formula illustrated in Figure 2.1 below. Individual PCB congeners differ in how many chlorine atoms are bound to the biphenyl backbone (ranging from 1 to 10) and at which positions they are bound. The physical and environmental properties of PCBs are largely determined by their chlorine substitution pattern. In general, lower chlorinated congeners have greater volatility and are therefore less persistent than higher chlorinated congeners, though this also depends on which positions the chlorines are bound to (Erickson, 1997).

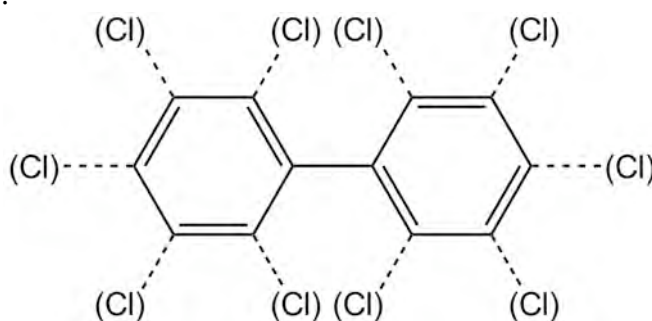


Figure 2.1. Generalized structure of PCBs, illustrating the biphenyl skeleton and the 10 different positions where chlorine atoms may be substituted.

The chlorination pattern of individual PCB congeners also determines their environmental toxicity by affecting their conformation, or the shape they take in 3-D space. The two phenyl components may rotate around the single connecting bond between them, and generally adopt the torsion angle that confers the least intramolecular interactions and strain in the molecule. For congeners that have one or more chlorines substituted at the *ortho* position, that is, closest to the single connecting bond, steric interactions force the molecules into conformations with greater torsion angles ($68-79^\circ$), with the two phenyl components in planes that are near perpendicular to each other. In contrast, non-*ortho* substituted PCBs adopt near coplanar conformations, with the two phenyl components almost in the same plane (torsion angle of 48°). These coplanar PCBs are the most toxic of all the PCB congeners, and are often referred to as dioxin-like congeners, as they have similar structural and toxicological properties to that class of carcinogens (Robertson and Hansen, 2001).

PCBs have low solubility in water, preferring a nonpolar phase. As a result, PCBs tend to partition away from water onto sediment, organic detritus or other particulate matter. This allows them to enter marine and aquatic ecosystems as these particulates are taken up by small organisms, which are in turn predated on by larger and larger organisms moving up through the food web. PCBs can also be absorbed directly by certain organisms. Due to their high lipophilicity (propensity to partition into fats, oils, and other nonpolar organic phases), PCBs within organisms tend to accumulate in tissues with high lipid content, such as blubber and liver (Robertson and Hansen, 2001).

2.1.2 History and Use

PCBs are not known to occur in nature, and were first synthesized and produced commercially in 1929. The Monsanto Chemical Corporation was the largest American and global producer of PCBs, and did so from 1935 until 1977, when its PCB manufacturing plants were shut down. PCBs were not produced as individual congeners, but rather as various mixtures under the trademarked name “Aroclor”. Many different varieties of these commercial mixtures existed, differing in the chlorination patterns of their components. Aroclor 1260 and Aroclor 1254, the most widely used mixtures before 1950, also contained the greatest composition of highly chlorinated congeners. Many other countries soon began producing commercial PCB mixtures, including Japan, Germany, France, Italy, Poland, and Czechoslovakia. Though most of these countries ceased production in the 1970s, Sovol, a commercial PCB mixture resembling Aroclor 1254, was produced by the former USSR through the 1990s (Erickson, 1997).

The highly persistent and stable nature of PCBs was what made them attractive for industrial uses, as well as what makes them an environmental threat and difficult to dispose of and degrade. In particular, PCBs have high thermostability, low flammability (making disposal by incineration difficult), low chemical reactivity, and high electrical insulative properties. These properties led to the widespread use of PCBs in a number of industrial applications (Robertson and Hansen, 2001), including uses as heat transfer fluids, hydraulic fluids, lubricating and cutting oils, and as additives in pesticides, paints, copy paper, adhesives, sealants, and plastics. However, their most common use were as dielectric fluids in capacitors and transformers, which accounted for about 70% of U.S. sales from 1930-1975 (Durfee *et al.*, 1976).

2.1.3 Regulation

The toxic effects of PCBs have been known as early as 1936, when workers involved PCB production started developing “chloracne”, an acne-like skin condition associated with high exposure to halogenated aromatic compounds. However, legislation regulating PCBs was not enacted until 1976, with the Toxic Substances Control Act (TSCA). The TSCA was passed as part of a wave of groundbreaking environmental legislation passed in the 1970s starting during the Nixon administration, including the Clean Air Act, Clean Water Act, National Environment Policy Act, and Endangered Species Act. The TSCA was passed in response to growing public awareness and support for environmental issues, and more immediately to the “Yusho” incident in 1968, wherein over 1600 adults and children in Japan developed symptoms including growth defects, anemia, and reduced neural capacity in response to consuming rice contaminated with PCBs (Erickson, 1997).

The TSCA mandated a complete ban on the manufacture, processing, distribution, or use of PCBs effective January 1, 1979, unless these activities were carried out in a “totally enclosed manner”. Further, the Environmental Protection Agency (EPA) was given the authority to implement the TSCA provisions, and specifically to determine which uses were considered “totally enclosed”, devise regulations concerning the marking and disposal of PCBs, and to enforce the ban. The EPA initially restricted the ban to activities involving greater than 500 ppm PCBs, restricting it further to 50 ppm a year later (in 1980), and finally in 1984 imposing the ban on activities listed involving “any detectable PCBs”. The

EPA has also mandated strict recordkeeping, disposal, and cleanup requirements related to current and former PCB activities. Nonetheless, PCB use in the United States still continues for “totally enclosed” uses, including in certain capacitors and transformers. Other pieces of legislation, including the Occupational Safety and Health Act (OSHA), Clean Water Act, Comprehensive Environmental Response, Compensation, and Liability Act (“Superfund”), Resource Conservation and Recovery Act, Hazardous Materials Transportation Act, Food, Drug, and Cosmetic Act, Safe Drinking Water Act, Clean Air Act, and state and local laws, also contain specific provisions pertaining to PCBs (Erickson, 1997).

Many other Western countries developed their own legislation regarding PCBs, leading to the ban of their use in Canada (1980), East Germany (1984), the Netherlands (1985), and Sweden (1973), and “restriction” on their use in Great Britain (1980) and West Germany (1978) (Barrie *et al.*, 1992). Continued observations of the environmental impacts of PCBs, as well as their continued use outside of North America and Europe throughout the 1980s and 90s revealed the growing need for a global international agreement regulating PCBs. The Stockholm Convention on Persistent Organic Pollutants in 2001, currently ratified by 152 countries, established a plan for cooperative management and protection against unintentional releases of PCBs in the short term, and a goal to decommission all equipment containing PCBs and eliminate all PCB waste sites by 2028 (Stockholm Convention website, <http://chm.pops.int/Default.aspx>). Despite these regulations, an estimated 31% of all PCBs produced (equivalent to 410,481 metric tons (905 million pounds) from 1930 through 1993) are thought to have been released into the environment (Barrie *et al.*, 1992; Breivik *et al.*, 2002), in part due to improper storage and disposal (Dewailly *et al.*, 1993), imparting toxic effects on ecosystems both near the source of their release, and in more remote areas like the Arctic through various transport mechanisms.

2.2. Transport Pathways of PCBs into the Arctic

While some local and regional emissions sources of PCBs in the Arctic exist (primarily from military facilities, harbors, and landfills), emissions from these sources alone are unable to account for the high levels of PCB contamination in the Arctic (AMAP, 2004; AMAP, 1998). Indeed, the vast majority of PCBs arrive in the Arctic by long-range transport via atmospheric circulation and precipitation, oceanic circulation, riverine input, ice pack drift, and biotic migrations. Figure 2.2 illustrates a generalized schematic of some of these transport pathways.

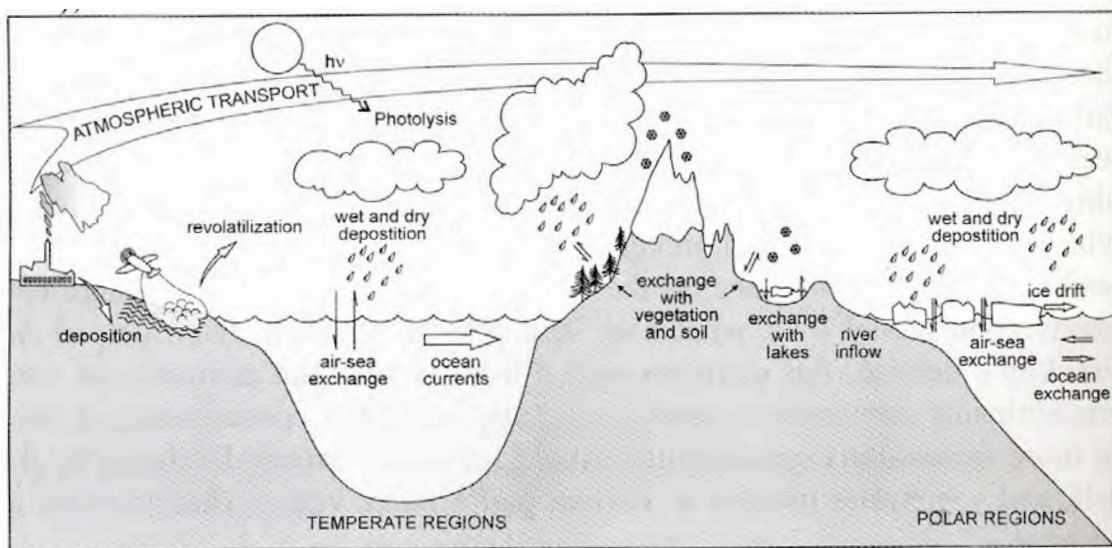


Figure 2.2. Some generalized long-range transport pathways of organic pollutants from temperate sources to the Arctic region. Taken from Figure 17.1 of (MacDonald, 2007).

2.2.1. *Atmospheric Transport*

2.2.1.1. Volatilization and Global Fractionation

The physical and chemical properties (such as solubility, vapor pressure, and molecular weight) of various organic pollutants affect their volatility into the atmosphere and ability to be transported to the Arctic region. Pollutants with high volatility directly enter the gaseous phase and can be immediately transported to the Arctic, whereas those that are less volatile are more difficult to sustain in the gaseous phase and are deposited closer to the emission source either through a direct phase change or by adhering to particulate matter that is subsequently deposited. However, under favorable meteorological conditions (typically warm weather events), less volatile pollutants may revolatilize and undergo further transport, a process colloquially referred to as the “grasshopper effect” (AMAP, 2004; AMAP, 1998). The different patterns of latitudinal atmospheric transport owing to differences in volatility is known as “global fractionation”, and is illustrated in Figure 2.3.

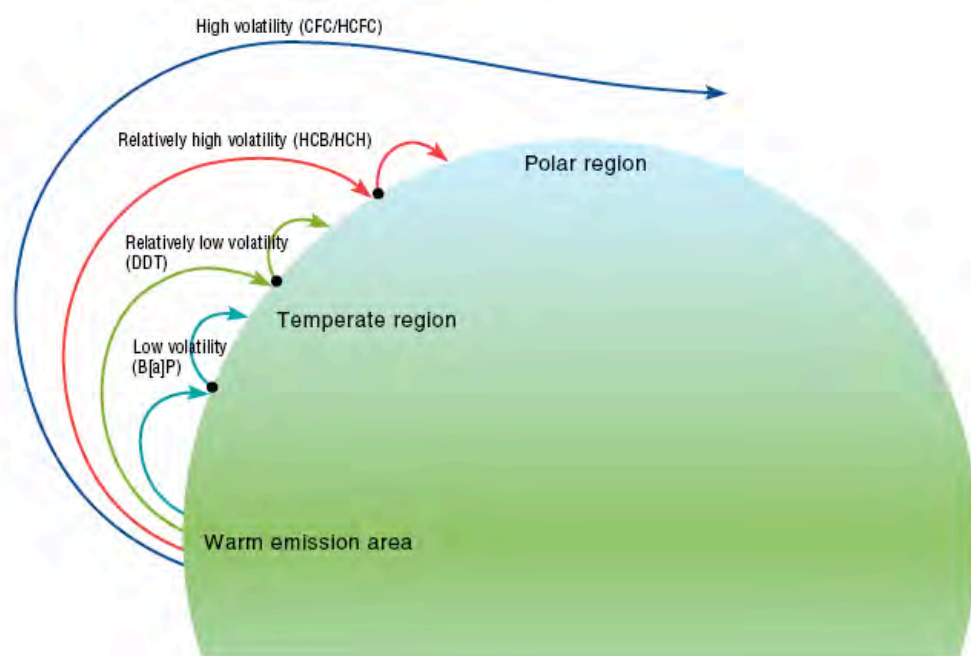


Figure 2.3. Global fractionation of selected organic pollutants. Highly volatile pollutants may enter the Arctic region directly, while low volatile pollutants require several revolatilization events to get there. Taken from Figure 2.2 of (AMAP, 2004; AMAP, 1998).

PCBs display a broad range of volatility, determined by their level of chlorination. Lower chlorinated congeners are more volatile, and therefore can enter the Arctic region more directly. Higher chlorinated congeners, in contrast, are first deposited onto water, plant, or soil surfaces somewhere between the emissions source and the Arctic, and depend on revolitalization to move further northward via the “grasshopper effect”.

2.2.1.2. Atmospheric Retention and Deposition Processes

Once they have entered the atmosphere through volatilization, most PCBs are retained in the atmosphere for a period of a few to many weeks, allowing sufficient time for them to be transported to the Arctic by atmospheric currents, which generally takes 20-30 days (Barrie *et al.*, 1992). PCBs partition strongly onto particulate matter in the atmosphere due to their low solubility in water and water vapor, though all congeners exist at a gas-particulate dynamic equilibrium (Barrie *et al.*, 1992; MacDonald, 2007). Given their relatively large size and high density, particulate matter is more likely to be deposited to the Earth’s surface than gaseous molecules.

PCBs are deposited into the Arctic either by “dry” (i.e. independent of precipitation) or “wet” (i.e. dependent on precipitation) methods. Particulate matter is continuously deposited and re-introduced into the atmosphere independently of precipitation events. Dry deposition can also occur over bodies of water through air-water exchange of either particulate-bound or gaseous PCBs at the surface (MacDonald, 2007). These dry methods contribute a significant portion of PCB deposition especially in the high Arctic, where annual precipitation is at desert-like levels (AMAP, 2004; AMAP, 1998).

Precipitation removes PCBs both in the gaseous phase (through gas exchange into rain or snow) and the particulate-bound phase from the atmosphere (MacDonald, 2007). In

the Arctic, the dominant form of precipitation is snow, due very low annual temperatures. Falling snow is extremely efficient at scavenging both gaseous and particulate-bound PCBs, and depositing them to the Earth's surface. The large surface area and convoluted, crystalline surface of snow crystals make them much better adsorbers of PCBs than rain droplets. Furthermore, the surface of snow crystals erodes as the snow ages following deposition. This provides a mechanism for snow-bound PCBs to be quickly released to other media on the Earth's surface (AMAP, 2004; AMAP, 1998).

At colder temperatures, PCBs enter the liquid, supercooled liquid, or solid phase, and are more likely to be either directly deposited to the surface, or become bound to particulate matter. At Arctic temperatures, most PCBs are in a solid form (MacDonald *et al.*, 2000). These denser phases increase the likelihood of deposition either directly or through particulate-bound intermediates. Therefore, atmospheric PCBs are more likely to be deposited the closer they are to the Arctic region, and in fact, less than about 5% of atmospheric PCBs are estimated to remain airborne at Arctic temperatures (MacDonald *et al.*, 2000). Figure 2.4 shows the propensity of the congener PCB-28 to favor deposition in Arctic and mountainous areas rather than at more temperate or tropical temperatures.

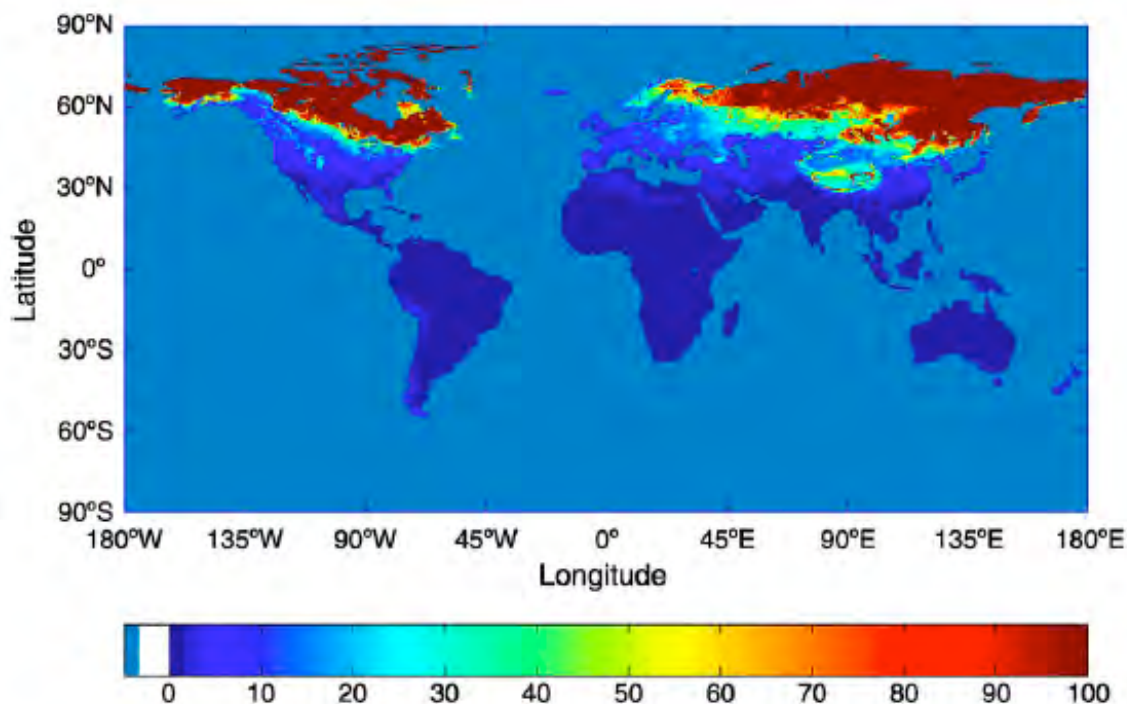


Figure 2.4. Global pattern of PCB-28 soil deposition in January (modeled). Darker and redder areas represent those with a higher ratio of PCB-28 retention in the soil compared to atmospheric PCB-28. Taken from Figure 2 of (Valle *et al.*, 2005).

2.2.1.3. Atmospheric Circulation Patterns

Atmospheric PCBs are dependent on air currents to transport them to the Arctic region. Figure 2.5 illustrates representative air circulation patterns in the Northern hemisphere for January and July.

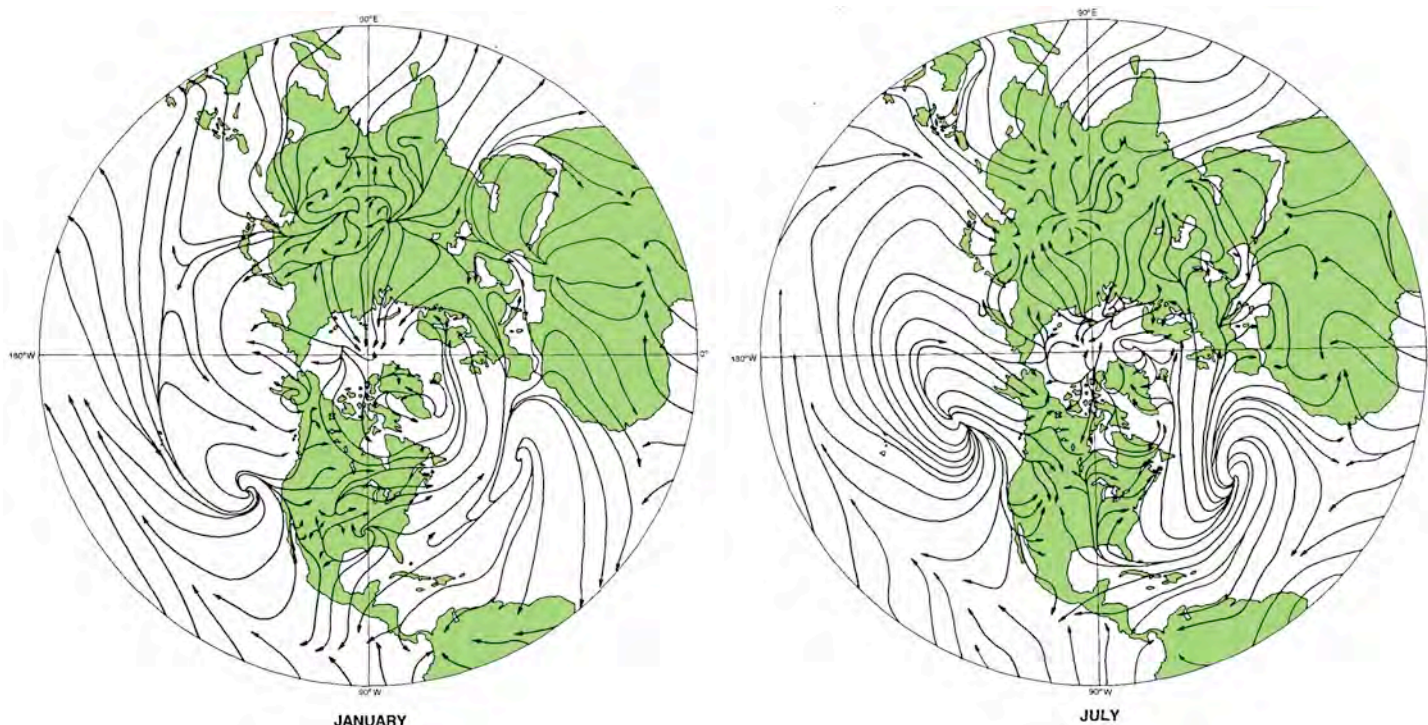


Figure 2.5. Northern hemisphere atmospheric circulation trends, mean monthly averages for January and July. Modified from Figures 6 and 7 of (Barrie *et al.*, 1992).

As can be seen from Figure 2.5, atmospheric circulation patterns are highly seasonally variable. These air streams flow from anticyclones, areas of high pressure where air masses are subsiding, to cyclones, areas of low pressure where air masses are rising. In the winter, the Siberian anticyclone drives strong currents from Eurasia into the Arctic, and then over northern Canada and Greenland (Barrie *et al.*, 1992). The Icelandic cyclone is responsible for drawing air masses from eastern North America and Europe into the Arctic, and the Aleutian cyclone does the same for air masses from Asia. Together, these three atmospheric formations account for 80% of south to north air transport (AMAP, 2004; AMAP, 1998).

In the summer, the Siberian anticyclone dissipates and is replaced by a much weaker north to south transport pattern (Barrie *et al.*, 1992). Likewise, the Icelandic and particularly the Aleutian cyclones also become weaker (MacDonald *et al.*, 2000). As a result, atmospheric transport to the Arctic overall is much weaker in the summer, accounting for only 20% of annual south to north transport, and this is reflected in seasonal differences of PCB soil deposition (Figure 2.6).

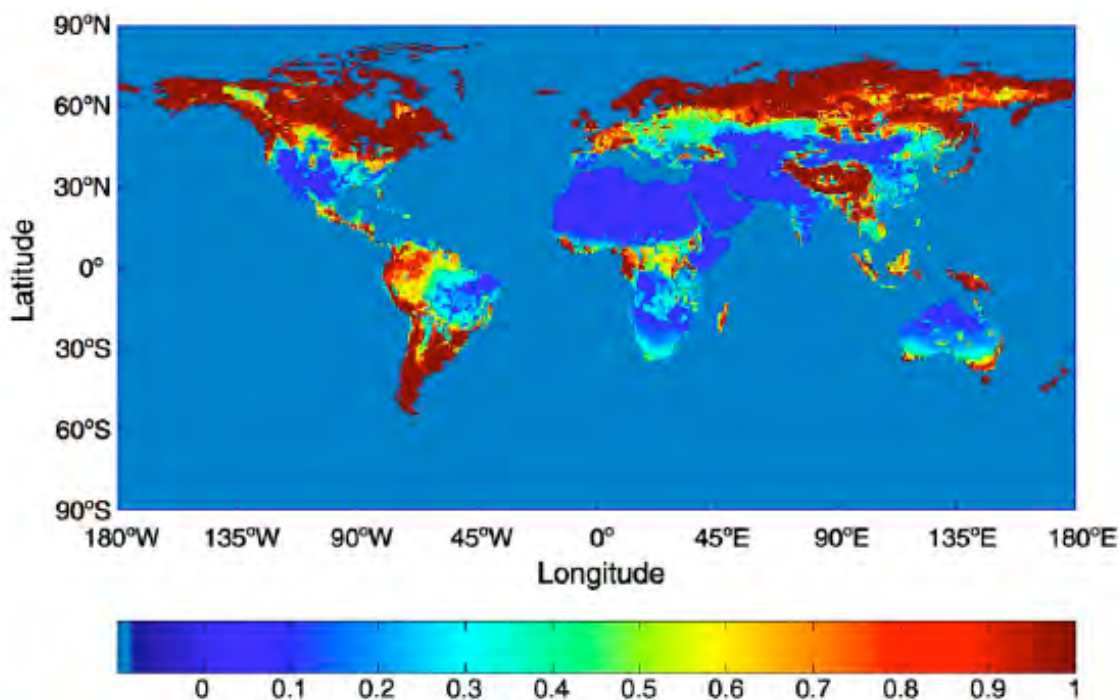


Figure 2.6. Global pattern of PCB-28 soil deposition in July (modeled). Compare to Figure 2.4, noting the different scale. Taken from Figure 2 of (Valle *et al.*, 2005).

2.2.2. Oceanic Transport

In contrast to the atmosphere, the ocean takes much longer to accumulate and transport PCBs, on the order of years or decades instead of days. However, once a significant concentration of PCBs in the ocean is attained, it can prove a potent pathway for contaminant transport, and ensure continued release of PCBs into the Arctic long after they were initially emitted. Furthermore, deposition to the ocean and subsequent circulation is a mechanism for even highly chlorinated congeners – which favor deposition at more temperate temperatures – to still undergo long range transport into the Arctic (AMAP, 1998; MacDonald *et al.*, 2000; AMAP, 2004).

The West Spitzbergen current on the western coast of Svalbard that cuts through the Fram Strait (see Figure 2.7) is a branch of the North Atlantic Drift derived from the American Gulf Stream, and accounts for about 42% of annual oceanic water flowing into the Arctic Ocean (MacDonald *et al.*, 2000). In the Arctic Ocean itself, two main currents dominate – the Beaufort gyre and the transpolar drift. The Beaufort gyre cycles water from the Beaufort and Chukchi seas, making one full rotation about every 10 years. The transpolar drift transports water from eastern Siberia across the Arctic and along the East Greenland coast. Arctic Ocean water also exits along West Greenland and the Canadian archipelago, though exchange can also take place in the other direction, depending on tidal currents.



Figure 2.7. General patterns of oceanic circulation in the Arctic region. © Woods Hole Oceanographic Institute.

2.2.3. Riverine Input

Total annual discharge of water from rivers into the Arctic Ocean amounts to only about 1% of that from the West Spitzbergen current alone (Barrie *et al.*, 1992). Nonetheless, rivers may transport substantial amounts of PCBs and other contaminants in the Arctic. PCB discharge from the Yenisey River (see Figure 2.8) into the Arctic has been estimated to be as much 1250 kg per year (Carroll *et al.*, 2008) while that from Canadian rivers is estimated to be 394 kg per year (MacDonald *et al.*, 2000).



Figure 2.8. Discharge of rivers into the Arctic Ocean. Taken from Figure 2.15 of (AMAP, 1998).

Arctic rivers are frozen in winter and only discharge water into the Arctic Ocean in summer. As they flow north, they accumulate PCBs from local sources. The Russian rivers in particular collect significant loads of PCBs from the industrial areas of Siberia, and also have the greatest discharge into the Arctic Ocean of all Arctic rivers (Figure 2.8). Therefore, while atmospheric transport of PCBs into the Arctic is minimal in summer, the melting of Arctic-bound rivers unleashes a substantial contaminant load at this time, when biological productivity in the Arctic is at its highest (Barrie *et al.*, 1992).

2.2.4. Ice pack drift

The extent of sea ice coverage in the Northern Hemisphere was estimated in the early 1990s to seasonally range from 9 – 16 million km², representing 64 – 114% of the surface area of the Arctic Ocean (extending into the northern Atlantic and Pacific Oceans in winter) (AMAP, 1998). In more recent years, this estimate has decreased to 5 – 13 million km², and some projections for the near future predict an Arctic Ocean that will eventually be completely free of ice in the summer (MacDonald *et al.*, 2005).

Sea ice provides another transport mechanism for PCBs and other contaminants into the Arctic. The ice follows the same current patterns as ocean water (Figure 2.7), but unlike water, the ice medium does not instantaneously inundate Arctic ecosystems with contaminants. Instead, like river ice, PCBs deposited onto the sea ice become trapped and accumulate through the winter, and are only released in large discharges when the ice melts in spring and summer (Barrie *et al.*, 1992). Because the ice may drift throughout different Arctic regions during this time, and even to more industrial areas in the North Atlantic and Pacific, it can accumulate a significant contaminant load.

2.2.5. Biotic migrations

A final transport mechanism of PCBs into the Arctic is through seasonal wildlife migrations. As described later in Section 3, relatively few species in the Arctic reside there year-round, as many only migrate there in spring and summer. These migrants often cover large distances and pass through industrial and agricultural areas. Certain whales and seabirds in particular are thought to be important carriers of contaminants into the Arctic. In the spring, the Eastern Pacific stock of grey whales migrates from Mexico and southern California to the Bering and Chukchi seas. They carry with them an estimated 20 to 150 kg of PCBs into the Arctic annually (Wania, 1998), though this contaminant load is only released if the whales die or are eaten there. While these whales are too big to be an important food source for most species, they are hunted by some indigenous groups, and dead grey whales are scavenged by polar bears and arctic foxes (AMAP, 2004).



Figure 2.9. The major migratory routes of bird species to the Arctic in summer. Taken from Figure 9.1 of (Blix, 2005).

While only four bird species are found in the Arctic year-round, over 100 different species migrate to the Arctic in summer. Many of these are seabirds that gather on cliffs or on the coastal tundra in colonies of up to 20 million (Blix, 2005). As these birds migrate from many diverse areas throughout the world (see Figure 2.9), they can transport significant amounts of PCBs into Arctic ecosystems (Wania, 1998). One recent study on a high Arctic lake located near a seabird cliff colony estimated that 14% of the PCBs in the lake's catchment area and 80% of the PCBs in the lake itself were transmitted there through the guano of the nearby bird colony (Evenset *et al.*, 2007).

2.3 PCBs and the Arctic Environment

2.2.1 *The Marine Ecosystem*

The Arctic marine ecosystem is host to a number of different mammal species. Only three cetacean (whale) species are resident in the Arctic during the whole year – the beluga, the narwhal, and the bowhead whale. In contrast, many pinniped (seal) species live in the Arctic year-round – including the ringed seal, harp seal, bearded seal, spotted seal, ribbon seal, and walrus. For most of the year, these pinnipeds live a pelagic lifestyle out at sea, but “haul out” onto the ice in large congregations three times during the year – to give birth, breed, and moult. The pinnipeds comprise the preferred and greatest proportion of the diet of polar bears, another year-round Arctic resident. While most bear species are thought of as terrestrial mammals, polar bears are uniquely adapted to living on and hunting from sea ice, making them ecologically wholly part of and the apex predator in the marine Arctic ecosystem. In winter, polar bear kills provide an important source of food for arctic foxes. In summer, the foxes prey on seabird adults, eggs, and chicks. The entire marine ecosystem in spring and summer is dependent on the phytoplankton bloom. When the sun finally rises above the horizon in spring, the phytoplankton bloom begins. In turn, zooplankton and small copepods increase in number, attracting an immense migration of fish, bird, and mammal species to the Arctic for the short window of extraordinary biological productivity in summer. The resident mammal species time the birth of their young to take advantage of this great source of food (Blix, 2005).

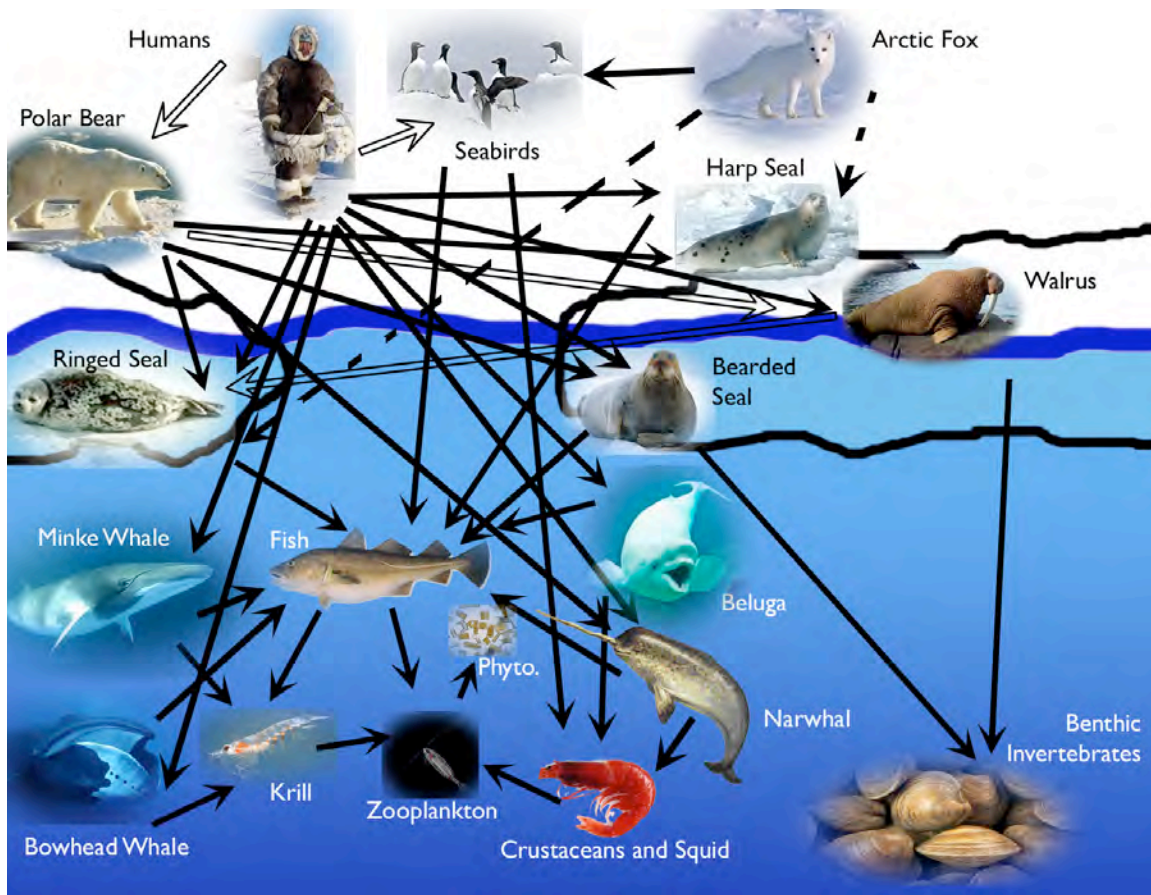


Figure 2.10. “Simplified” food web model of the marine Arctic ecosystem, illustrating many of the species covered by this study. Solid arrows represent frequent predatory relationships, open arrows represent occasional or rare predatory relationships, and dashed arrows represent scavenging. “Phyto.” = phytoplankton. In this schematic, seabirds, fish, and zooplankton are shown to comprise single unified groups. In reality, these groups display a great deal of trophic diversity, with distinct sets of prey and predators, and often members within these groups (especially seabirds and fish) will even predate each other. Self-made from information in (AMAP, 2004) and (Blix, 2005), among others.

A “simplified” schematic of the Arctic marine food web is presented in Figure 2.10. As seen from this figure, the marine Arctic food web is exceedingly complex, involving many different steps from the primary producer (phytoplankton) to the apex consumers (polar bears and humans). These long, complex food chains are one of the reasons why PCBs and other organic contaminants are thought to accumulate to a much greater degree in the marine ecosystem than in the terrestrial ecosystem, as there are many more opportunities for them to biomagnify between different trophic levels (Hoffman *et al.*, 2002). A second reason is that while all Arctic animals (both terrestrial and marine) possess significant fat reserves, marine mammals possess especially thick layers of blubber to keep warm, whereas terrestrial mammals rely on insulating fur in addition to their smaller fat reserves. One study of ringed seals on Svalbard revealed average blubber concentrations of up to 60% body weight (Blix, 2005). Finally, as discussed in section 2.2 above, oceanic, sea ice, and riverine transport of PCBs contribute significantly to the total contaminants loads of the marine ecosystem, whereas terrestrial ecosystems rely solely on atmospheric transport, precipitation, or biotic migrations.

2.2.2 The Terrestrial Ecosystem

Figure 2.11 illustrates a food web model of the Arctic terrestrial ecosystem. As evidenced from the figure, the terrestrial ecosystem is much simpler than its marine counterpart, with generally only two steps between primary producer (lichens and dwarf shrubs) and apex consumers (arctic wolf and humans). As discussed above, the shorter food chains in the terrestrial ecosystem is one reason for the generally lower concentrations of PCBs in terrestrial animals. Still, lichens are extremely efficient at scavenging contaminants from the atmosphere, because they have wide surface areas, and lack roots or vasculature and so must absorb nutrients (and contaminants) from the atmosphere (Gamberg *et al.*, 2005). It is for this reason, perhaps, that PCB levels are greater in caribou, whose diet consists significantly of lichens, than in muskoxen (see section 4), which solely eat vascular shrubs and sedges (Blix, 2005). Further, a number of ecological links exist between the terrestrial and marine ecosystems. Arctic foxes and humans span both the terrestrial and marine food webs. In one study, a gradient in PCB levels were observed in Arctic foxes correlating with how coastal or inland their ranges were, with coastal foxes that rely predominately on marine species displaying higher PCB concentrations than those that lived inland and depended more on terrestrial species (Klobes *et al.*, 1998). Finally, seabirds often concentrate in colonies on coastal cliffs. Their guano is not only an efficient purveyor of marine contaminants to the terrestrial ecosystem, but is also as a rich fertilizer, supporting microhabitats under seabird cliffs that display much greater biological productivity than the surrounding terrestrial environment (Evenset *et al.*, 2007).

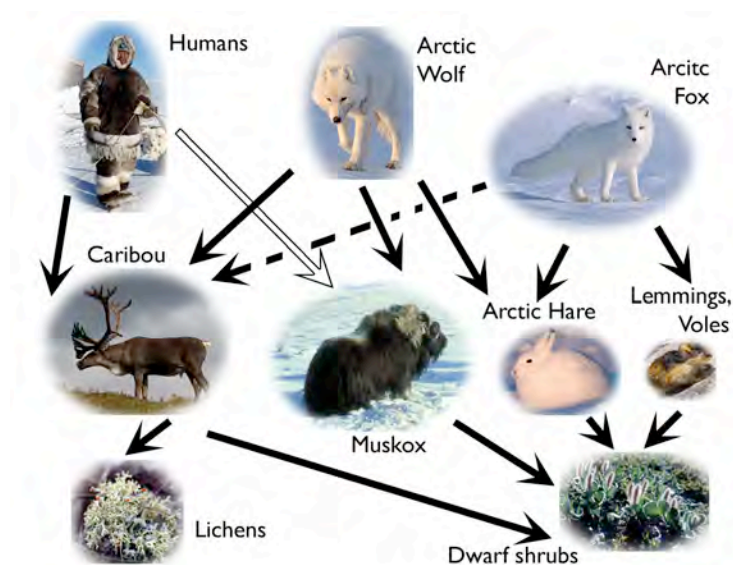


Figure 2.11. Simplified food web model of the terrestrial Arctic ecosystem, illustrating many of the species covered by this study. Solid arrows represent frequent predatory relationships, open arrows represent occasional or rare predatory relationships, and dashed arrows represent scavenging. Note that terrestrial bird species (e.g. ptarmigan, snowy owl) are not shown. Self-made from information in (AMAP, 2004) and (Blix, 2005), among others.

2.2.3 Physiological Effects of PCBs on Arctic Mammals

It is difficult to make broad generalizations about the effects of PCB exposure on all wildlife, because every species has its own unique biology and metabolic pathways, and thus different responses to and capabilities to deal with environmental contaminants. Nevertheless, certain categories of effects appear to be shared by many different species. In particular, PCBs seem to target the reproductive, endocrine, and immune systems, three biological systems that are extremely vital not only for the survival of individual organisms, but also for the continuation of entire species. This section aims to provide a brief review on some of the putative effects of PCB exposure observed in Arctic mammals. Note that where correlations are found, direct causations are not necessarily implied, and in several studies the effects of PCBs displayed complex interactions with those from other organic contaminants that organisms were exposed to (Letcher *et al.*, *in press*).

2.2.3.1 Polar Bears

As the apex predator and the species with the highest levels of PCBs in the Arctic (see section 4), physiological effects of PCB exposure have been best studied in polar bears more than any other Arctic mammal. Immunoglobulin G (IgG) proteins are a class of antibodies that accumulate in organisms as they age, ensuring proper immune responses to microorganisms throughout life. In a study from Svalbard, a significant ($p = .029$) negative correlation was observed between PCB concentrations and IgG levels in the blood of polar bears (Bernhoft *et al.*, 2000). Furthermore, correlations between PCB levels and delayed or impaired immune response were observed in polar bears inoculated with various viruses and antigens (Lie *et al.*, 2005; Lie *et al.*, 2004). Similarly, negative correlations were found between PCBs and levels of thyroid hormones required for proper behavioral, neurological, and psychological programming, and for ensuring sufficient oxygen uptake and maintaining proper body temperature (Braathen *et al.*, 2004). Negative correlations were also found between levels of PCBs and retinol (vitamin A) (Skaare *et al.*, 2001), a vitamin that serves as an antioxidant and ensures proper development (Letcher *et al.*, *in press*), and cortisol, an adrenal hormone that regulates energy metabolism, the maintenance of growth and development, and stress responses (Oskam *et al.*, 2004).

Significant correlations are also found between PCB exposure and reproductive phenomena such as the extremely skewed sex ratios found in some polar bear populations (Derocher *et al.*, 2003), reduction in the size of sexual organs and the rate of mating success (Sonne *et al.*, 2009a; Sonne *et al.*, 2006c), reduced testosterone levels in males (Oskam *et al.*, 2003), increased progesterone levels in females (Haave *et al.*, 2003), reduced sperm count and quality in males and increased occurrence of uterine lesions and endometriosis in females (Letcher *et al.*, *in press*), pseudohermaphroditism (enlarged clitoris) in females, and low cub survival (Wiig *et al.*, 1998). Finally, high PCB exposure may also be implicated in the development of tissue pathologies in kidney (Sonne *et al.*, 2006b) and liver (Sonne *et al.*, 2005a), as well as in reduced bone mineral density (Sonne *et al.*, 2004) and other skeletal deformities (Bechshoft *et al.*, 2009; Sonne *et al.*, 2005b).

2.2.3.2 Arctic Foxes

Like polar bears, arctic foxes also display strong correlations between PCB exposure and incidences of kidney and liver lesions (Sonne *et al.*, 2008c) and reduced bone mineral density (Sonne *et al.*, 2009c), in addition to thyroid effects (Sonne *et al.*, 2009b). Furthermore, domestic sledge dogs in Greenland, in experiments as analogs for arctic foxes, demonstrated impairment of liver and kidney function (Sonne *et al.*, 2008b) and immune capability (Sonne *et al.*, 2006a; Sonne *et al.*, 2007) in addition to sexual organ deformities (Sonne *et al.*, 2008a), in response to PCB exposure.

2.2.3.3 Humans

Significant physiological effects of PCB exposure are also observed in humans living in the Arctic. In one study of Inuit men from Greenland, increased concentrations of PCB-153 were associated with low sperm count and decreased sperm motility, though overall fertility was not affected and no direct evidence for endocrine hormone disruption was observed (Bonde *et al.*, 2008; Toft *et al.*, 2006). Also in Greenland, elevated concentrations of PCBs were associated with reduced insulin stimulation (Jorgensen *et al.*, 2008) and blood levels of high-density lipoprotein, responsible for binding “good cholesterol” (Deutch *et al.*, 2005). In Nunavik (northern Quebec), associations have been observed between high prenatal (*in utero*) exposure to PCBs and incidences of ear, respiratory, and gastrointestinal infections (Dallaire *et al.*, 2004; Dewailly *et al.*, 2000) and impaired visual acuity in infants (Saint-Amour *et al.*, 2006). In the Nunavik adults, negative correlations between PCBs and thyroid hormone levels have been observed (Dallaire *et al.*, 2009). Furthermore, high levels of PCBs have been demonstrated to affect the genomic architecture, resulting in reduced global DNA methylation (Rusiecki *et al.*, 2008) and the formation of DNA adducts, damaged DNA complexes implicated in cancer progression (Lagueux *et al.*, 1999). Finally, PCB exposure may also be implicated in the skewed sex ratios (Tiido *et al.*, 2006) and high incidence of Parkinson’s disease (Tanner *et al.*, 2006) observed in Inuit populations.

2.2.3.4 Cetaceans

Relatively little information exists on the physiological effects of PCB exposure on wild cetaceans in the Arctic or elsewhere. Nevertheless, some studies have revealed tentative associations between PCB exposure and the development of lesions and other histopathological effects in bowhead whales and belugas, and immune system deficiencies in belugas (Letcher *et al.*, *in press*).

2.2.3.5 Pinnipeds

Studies on the physiological effects of PCB exposure in seal species have predominately focused on populations in temperate, rather than Arctic areas. In temperate European populations of ringed, grey, and harbour seals, high levels of PCBs are thought to be associated with lesions, immune dysfunction, and reproductive and endocrine disruption. Though PCB concentrations in Arctic populations are generally lower than those in temperate ones (because they are further from local sources), it is likely that PCB

overexposure could present similar effects to populations of seals living in the Arctic (Letcher *et al.*, *in press*).

3 Methodology

3.1 Search Strategy

Two databases were used to find relevant articles for this literature survey – Web of Science (Thomson Reuters) and Environment Abstracts (CSA). Early searches used relatively stringent parameters (e.g. requiring the explicit inclusion of the term “Arctic”). The revelation of a cache of relevant articles that could not be identified by these parameters led to progressively more general searches, eventually including the names of all regions and countries in the Arctic and all possible references to PCBs (e.g. “PCBs”, “polychlorinated biphenyls”, “organochlorine contaminants”). Over 2000 hits were identified from these searches in the two databases combined, of which almost 600 were deemed relevant and obtained via Williams College Libraries or Interlibrary Loan. This list was then narrowed down based on the criteria described in section 3.2 below. In addition to database searches, many relevant articles were identified from citations in other articles that had been found from these searches. Often, it was difficult or impossible to find these cited articles (especially for institutional reports), and in these cases the author relied on the data presented for them in the article that cited them.

3.2 Scope

This literature survey focused exclusively on studies of PCB levels in Arctic species. However, the Arctic region cannot simply be defined as the area north of the Arctic Circle (about 66° 33' N), which is the southernmost extent of areas that experience the polar night (24 hours of darkness in winter) and midnight sun (24 hours of sunlight in summer). Rather, many other physical parameters besides solar irradiation, including atmospheric and oceanic circulation patterns (see sections 2.2.1.3 and 2.2.2), temperature, permafrost extent, and tree density, are also important in defining the extent of the Arctic region from a climatological and ecological perspective. Figures 3.1 and 3.2 illustrate some physical and political definitions of the Arctic region, respectfully.

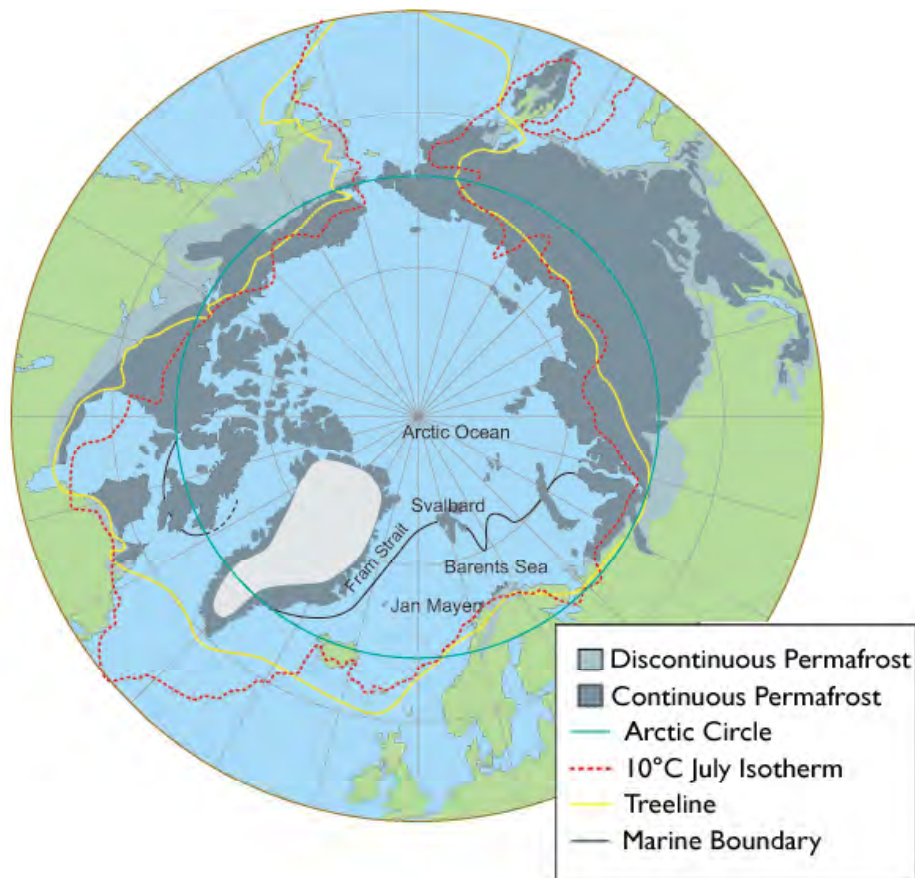


Figure 3.1. Physical definitions of the Arctic, including discontinuous (light grey) and continuous (dark grey) permafrost zones, the Arctic Circle (blue), 10°C July isotherm (red), treeline (yellow), and marine boundary (oceanic front between Arctic and Atlantic water masses, black). © Norwegian Polar Institute.

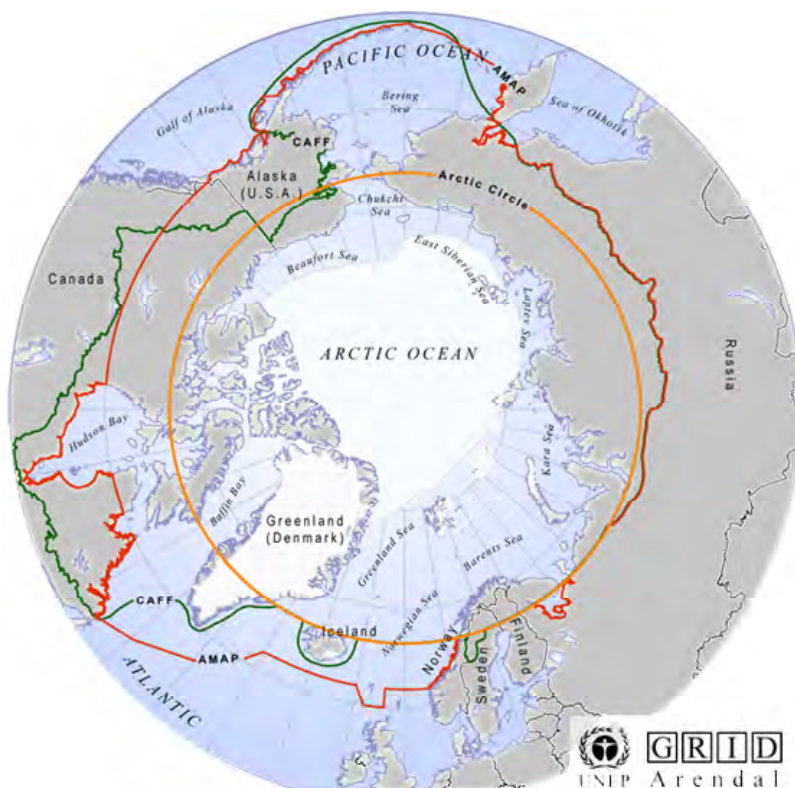


Figure 3.2. Political definitions of the Arctic, including international boundaries and those of the jurisdictions of the Arctic Monitoring and Assessment Programme (AMAP, red), the Arctic Council's working group for the Conservation of Arctic Flora and Fauna (CAFF, green), and the Arctic Circle (yellow). © UNEP/GRID–Arendal

It was deemed that the physical definitions of the Arctic would be more pertinent to the purposes of this study than the political definitions, which also incorporate human demography, and inter- and intranational boundaries. Specifically, the area above and including the 10°C July isotherm – that is, the region where mean temperatures for the hottest month of the year (July) are equal to or less than 10°C – was chosen as the study area. Only articles pertaining to locations within this region, or to those subjectively deemed by the author to be sufficiently close to its border (e.g. Tromsø, Norway), were included in this study. Many articles included samples from multiple areas, often including locations from both inside and outside the study region. For these articles, only data from locations within the study region were considered. Furthermore, many articles related to locations near local sources of PCBs, especially near Cold War-era radar stations along the Distance Early Warning (DEW) or Mid-Canada lines, built to provide sufficient warning time in case of an incoming Soviet attack from across the Arctic Ocean. As discussed in section 1 (the introduction), one of the benefits of studying PCB concentrations in the Arctic, and one of the goals of this study, is to establish baseline levels of PCB contamination in places far removed from local sources. Therefore, these articles were excluded from the data presented in section 4.

One of the original purposes of this study was to study historical trends of PCB levels in various physical media (e.g. atmosphere, seawater, soil, ice, snow) and throughout the entire trophic web (e.g. plants, zooplankton, invertebrates, fish, seabirds, mammals). However, due to the daunting number of articles obtained and out of the interest of time,

the scope of this study was narrowed exclusively to mammals, and only data from articles pertaining to mammals in the Arctic were analyzed.

Other organisms in the Arctic generally occupy lower trophic positions and have shorter lifespans than mammals, and therefore trends of PCB contamination in these organisms may reflect more instantaneous changes in global PCB emission and contaminations because they are closer to the point of emission. However, because there are more biomagnification steps to the mammal trophic levels, only they generally exhibit PCB levels that are high enough to be accompanied by physiological and toxicological effects. Furthermore, a major focus of this study is on humans living in the Arctic, and they, along with the sources of food they predominantly depend on, are mammals.

While an early analysis was performed on Arctic atmospheric data, these data were not included in the final analysis because they appeared incomplete (only covering the years 1992-2005), were highly regionally variable, and were at concentrations several orders of magnitude below those from mammals. Additionally, without data from other physical media or biota (e.g. zooplankton) that link the physical media to higher organisms, the atmospheric data was deemed to be uninformative.

Finally, only concentrations representing the sum of 5 or more PCB congeners were included in the data. Several articles reported concentrations only for a distinct functional category of PCBs, such as coplanar PCBs, that generally only comprised 3 congeners. Because these congeners were much less represented in Arctic mammals than other congeners, the concentrations were at orders of magnitude lower than concentrations reported for greater numbers of and types of congeners and so were insufficient measures of Σ PCB (the sum of all PCB congeners). Differences in concentrations of PCB-153 have been shown to mimic differences in concentrations of Σ PCB, and so several studies reported only concentrations of PCB-153 as a proxy to assess correlations between Σ PCB exposure and some physiological variable (e.g. (Tiido *et al.*, 2006)). However, while PCB-153 may be a substitute for relative Σ PCB concentrations, it does not provide information on absolute Σ PCB concentrations needed for this cross-study survey, and so these data were also excluded.

3.3 Data compilation and analysis

Each unique Σ PCB concentration reported in the data was recorded as a distinct datapoint, along with information regarding the year and location of sampling, the tissue and species analyzed, the gender of the specimen, the units (i.e. ng/g lipid or wet weight) and statistic (i.e. arithmetic mean, geometric mean, or median), whether Σ PCB was determined by calibration with Aroclor or individual congeners, how many and which congeners the Σ PCB value represents, and the variability of the data (reported as either standard deviations, confidence intervals, or ranges) if the concentration value represented multiple specimens. The articles varied greatly with respect to all these parameters.

Many studies conducted by the same authors or labs reported on the same specimens in different articles. When the concentration data were identical, only one set of data was included. However, datasets for the same specimens that were reported using different parameters (e.g. lipid or wet weight, the number of congeners included in Σ PCB, which tissues were analyzed) were considered unique, and each set was included in the final data. Though giving more weight to certain samples because they were reported multiple ways

could introduce some bias into an analysis of the entire final dataset, the benefits of including different parameters to provide better comparison with studies that only looked at one kind of parameter (e.g. only lipid or wet weight) were thought to outweigh the potential bias that could be introduced.

In all animals, gender differences exist with regard to concentration of contaminants. This is because females are able to shunt contaminants to their offspring, resulting in generally lower levels in females than in males (e.g. (Verreault *et al.*, 2006)). In mammals, this can occur both before birth (*in utero*) and after birth (through lactational transfer). Because of this phenomenon, PCB levels in males and females are considered separately. Likewise, juveniles are also considered separately. Because they have small body masses while receiving significant loads of PCBs at a stage in life when the viability of developmental, neurological, and endocrine pathways is critical, the levels and toxic effects of PCBs in juveniles are often severe.

“Lipid weight” refers to concentration values that have been adjusted to the amount of lipids in the sample analyzed. Because PCBs are extremely lipophilic and hydrophobic, they only concentrate in the lipid portions of various tissues. Therefore, “wet weight” (unadjusted) values in tissues that are rich in lipids (e.g. blubber or adipose tissue) are much greater than those in tissues that are poor in lipids (e.g. blood). Lipid weights, calculated by simply dividing the concentration value by the lipid percentage of the tissue being analyzed, eliminate this bias inherent in comparing results from different tissues, as well as the more subtle effects caused by differences in lipid percentages within the same kind of tissue from different individuals, enabling broad comparisons to be made. However, wet weight values are much more physiologically relevant, because they indicate the precise amount of PCBs in a given tissue, enabling determinations, for instance, of how much Σ PCB will be transferred to a caribou calf by drinking a certain amount of its mother’s milk, or to a polar bear by eating a certain amount of ringed seal blubber (Loughlin *et al.*, 2002). Furthermore, differences in PCB levels between tissues could still be preserved even when lipid-adjusted, depending on the method of PCB intake and how different species deal with PCB contamination (how they are metabolized and where they are shunted to). Because many articles reported lipid percentages for the tissues they analyzed, regardless of whether they reported concentrations in lipid or wet weight, it could have been possible to standardize the data by calculating all the concentration values in lipid or wet weight. However, not all articles included lipid percentages, and given the advantages of both lipid and wet weights, it was decided that concentration data would be recorded as whichever a given article used.

In order to quantify PCB concentrations from spectral data, the peak sizes need to be calibrated to a standard amount of PCB of a known concentration. For many early studies and some later studies, Aroclor mixtures (either Aroclor 1254 or Aroclor 1260) were used to calibrate PCB concentrations. While much less time-consuming than using many different congeners to calibrate the data and then presenting PCB concentrations as the sum of individual congeners, the use of Aroclor standards for calibration tends to result in overestimation of Σ PCB concentrations (Oehme *et al.*, 1995b). Furthermore, as methods for determining PCB concentrations had not yet been fully standardized and perfected, the results from these early studies tend to be somewhat suspect ((Holden, 1973); Jay Thoman, *personal communication*). Nonetheless, differences between Aroclor or individual congener calibration alone were shown to be insufficient to explain differences in PCB concentrations

from different samples (Muir *et al.*, 1988). The results from these early studies were included for the sake of completeness, with the understanding that they may lack accuracy.

For many articles, a number of manipulations and calculations needed to be made by the author on the data presented. For instance, some articles only presented concentration data graphically, with no mention of exact values. In these cases, the images of the graphs (usually bar graphs) were imported into Adobe Photoshop CS3 v. 10.0. Photoshop's digital ruler tool was used, calibrated to the scale presented on the y-axis, to measure the height of the various bars, and thereby obtain rough concentration values. Many articles also reported data that had not been pooled for various parameters such as gender, species, or location, either presenting each sample individually, or pooling samples based on subdivisions outside the scope of this report (e.g. dividing male samples into different age groups). For these data, the author himself calculated averages and standard deviations for pooled values. Many studies (particularly the Canadian and Greenland studies) presented samples on a highly geographically localized basis. In an effort to obtain a sufficient number of data points to compare different historical trends between somewhat larger areas, the author designated 16 distinct regions (shown in figure 3.3) in which all data would fit into. In a later analysis (figure 5.2) some of these regions were in turn pooled into larger regions. Finally, to calculate 3-year running averages, geometric means were first calculated for all datapoints in a given year (to buffer against very high or very low values) before the arithmetic mean was taken for each 3-year period of these geometric means.

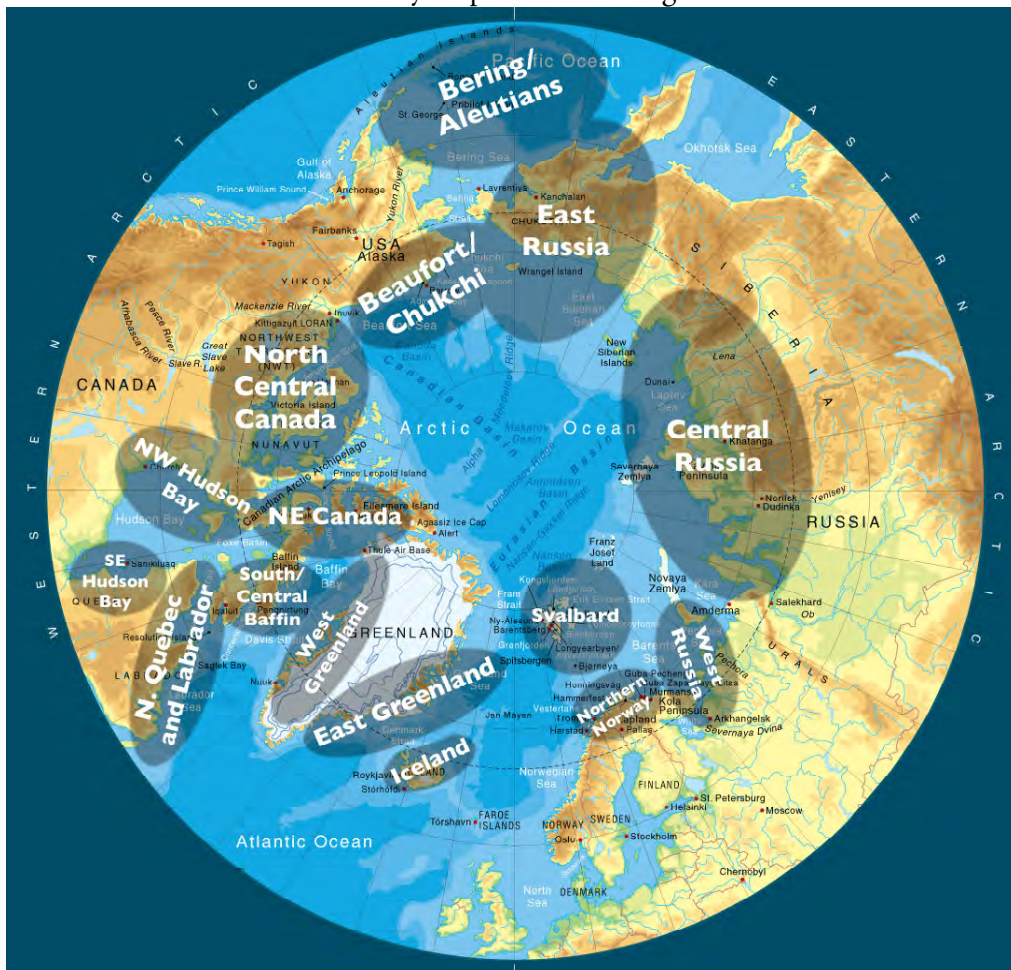


Figure 3.3. Regions designated and analyzed in this study. Modified from (AMAP, 2004).

3.4 Establishing threshold levels for physiological effects

While field studies have revealed numerous putative associations between PCB exposure and physiological impairments in Arctic wildlife as described section 2.3.3 above, they are incapable of establishing precise threshold levels of PCB concentrations at which physiological effects are seen, as these determinations can only be made using dose response experiments in laboratory settings. It is useful in studies such as this to compare observed PCB concentration data to threshold levels that may at least tentatively identify when (or where) high PCB exposure could be accompanied by physiological effects in certain organisms. Therefore, the data presented in figure 4.2 are presented alongside experimentally derived PCB threshold levels that have been cited for comparison in previous reviews of PCB contamination in Arctic mammals (e.g. (AMAP, 1998; Fisk *et al.*, 2005)). These thresholds are those at which neurological (1,000 ng/g lipid weight, loss of visual and short-term memory in human neonates, (Ahlborg *et al.*, 1992)), immune effects (21,000 ng/g lipid weight in rhesus monkeys, (Tryphonas, 1994)), and reproductive effects (25,000 ng/g lipid weight in captive harbour seals, (Boon *et al.*, 1987)) are observed.

4 Results

A total of 955 datapoints from 225 different reference articles were compiled in the final analysis. The breakdown of these data and articles by mammal group is shown in figure 4.1. A high degree of variance was observed in Σ PCB concentrations, particularly between different sampling locations, years, species, and tissues. Additionally, a great deal of variability was observed even between different individuals of the same species and gender sampled in the same location at the same time. For example, the Σ PCB concentration (sum of 23 congeners) in the breast milk of one woman from Monchegorsk, in the Kola peninsula (western Russian Arctic) was 13.5 times higher than the mean and 25 times higher than the minimum of the other 14 women sampled there at that time (Polder *et al.*, 1998). This variance is also evidenced from the quite large standard deviations, ranges, and 95% confidence intervals that were collected for the concentration data (where possible), weakening statistical confidence in individual values and the differences between datapoints, even where these differences appeared significant. Unfortunately, due to time constraints and technical issues with plotting these standard deviations, ranges, and confidence intervals, they are not shown in the following figures. Figure 4.2 shows all datapoints and 3-year moving averages broken down by mammal group. In figures 4.3 – 4.8, the data for pinnipeds and sea otters, cetaceans, polar bears, humans, terrestrial carnivores, and terrestrial herbivores, respectfully, are further broken down by the various parameters discussed in section 3.3. Finally, figure 4.9 lists the reference citations for each mammal group.

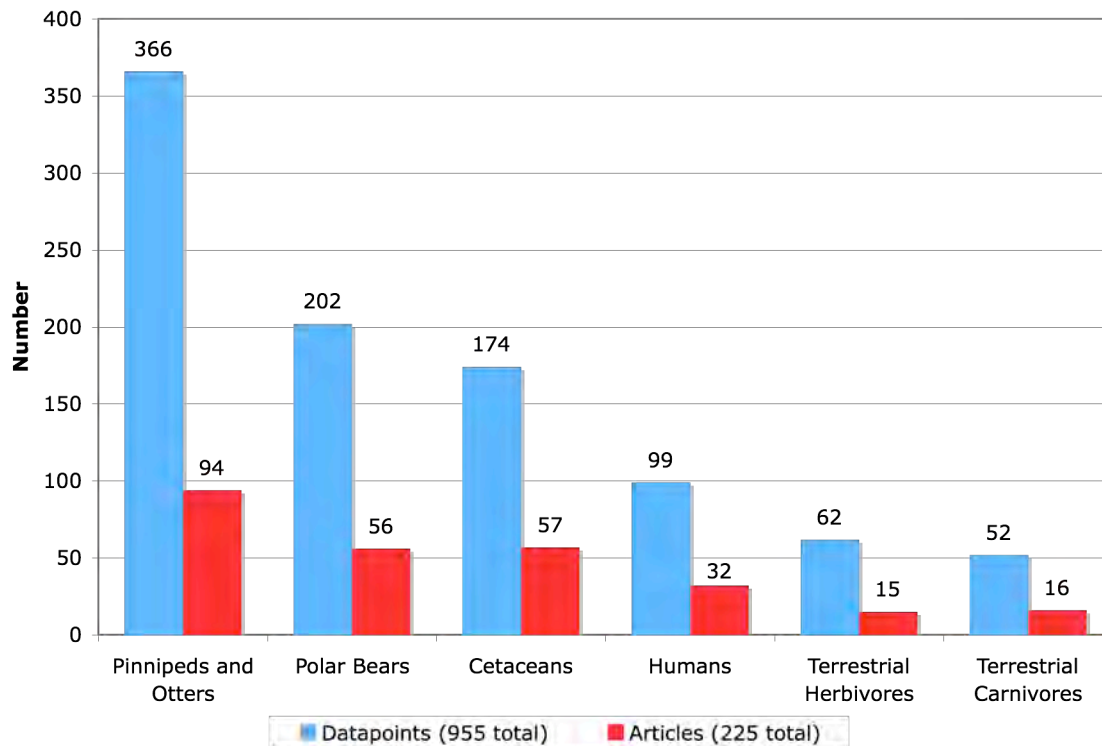


Figure 4.1. The number of datapoints and references used in the final analysis, broken down by mammal group. Note that the total number of references is less than the sum of the number of references for each mammal group because it counts references that contained data for multiple mammal groups only once.

Figure 4.2. Σ PCB concentration results for pinnipeds and sea otters, cetaceans, polar bears, humans, terrestrial carnivores, and terrestrial herbivores, shown with 3-year moving averages. Note that all y-axes are on a log scale, and that this scale is the same for all groups except terrestrial herbivores. Also shown are threshold Σ PCB levels for neurotoxic effects in human infants (dotted line, Ahlborg *et al.*, 1992), immune effects in rhesus monkeys (dashed line, Tryphonas *et al.*, 1994), and reproductive effects in captive harbour seals (solid line, Boon *et al.*, 1987).

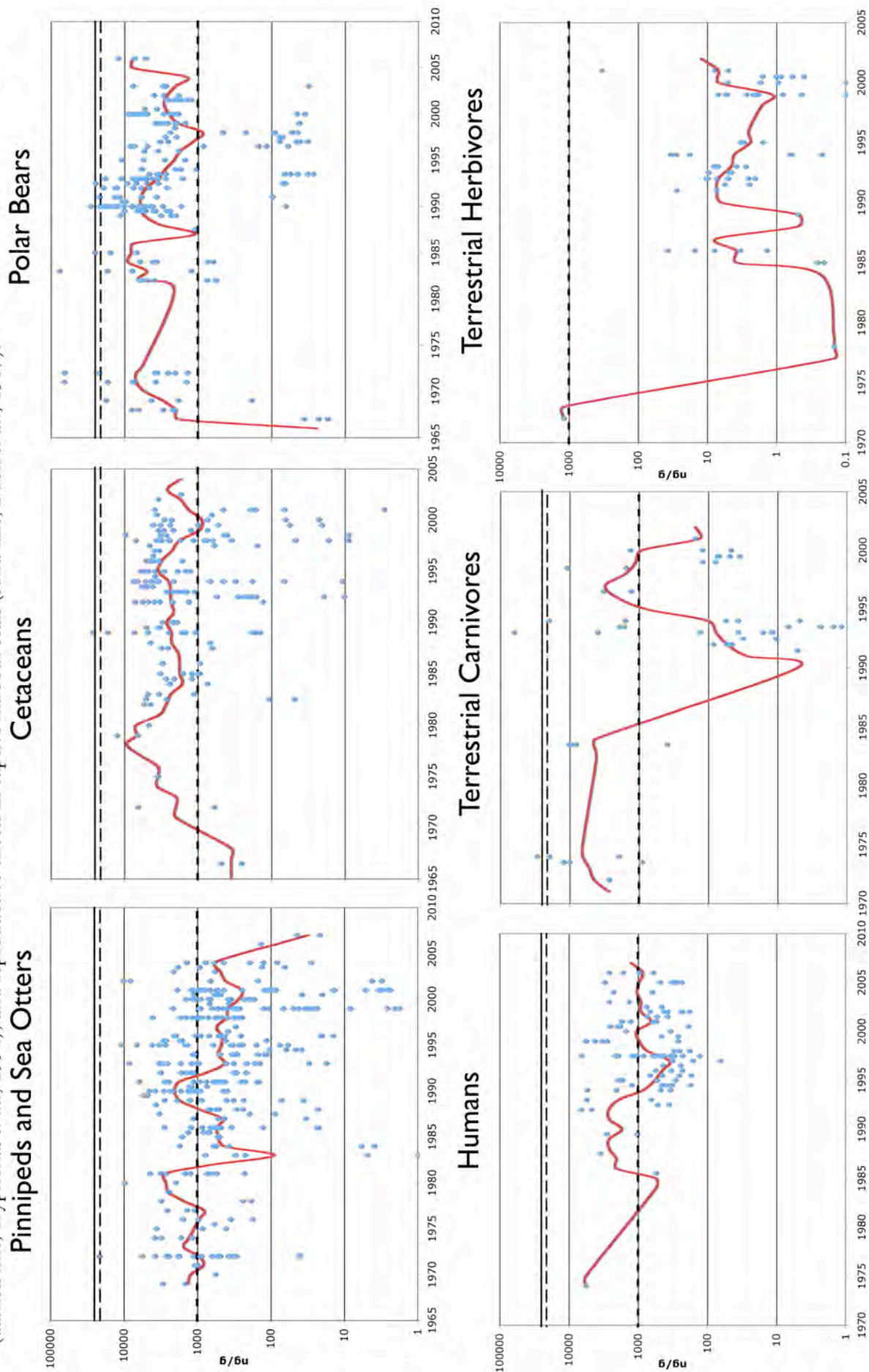


Figure 4.3. Σ PCB concentration results for pinnipeds and sea otters, grouped by tissue and weight measurement (top left), species and gender (top right), region (bottom left), and statistic and number of congeners (bottom right).

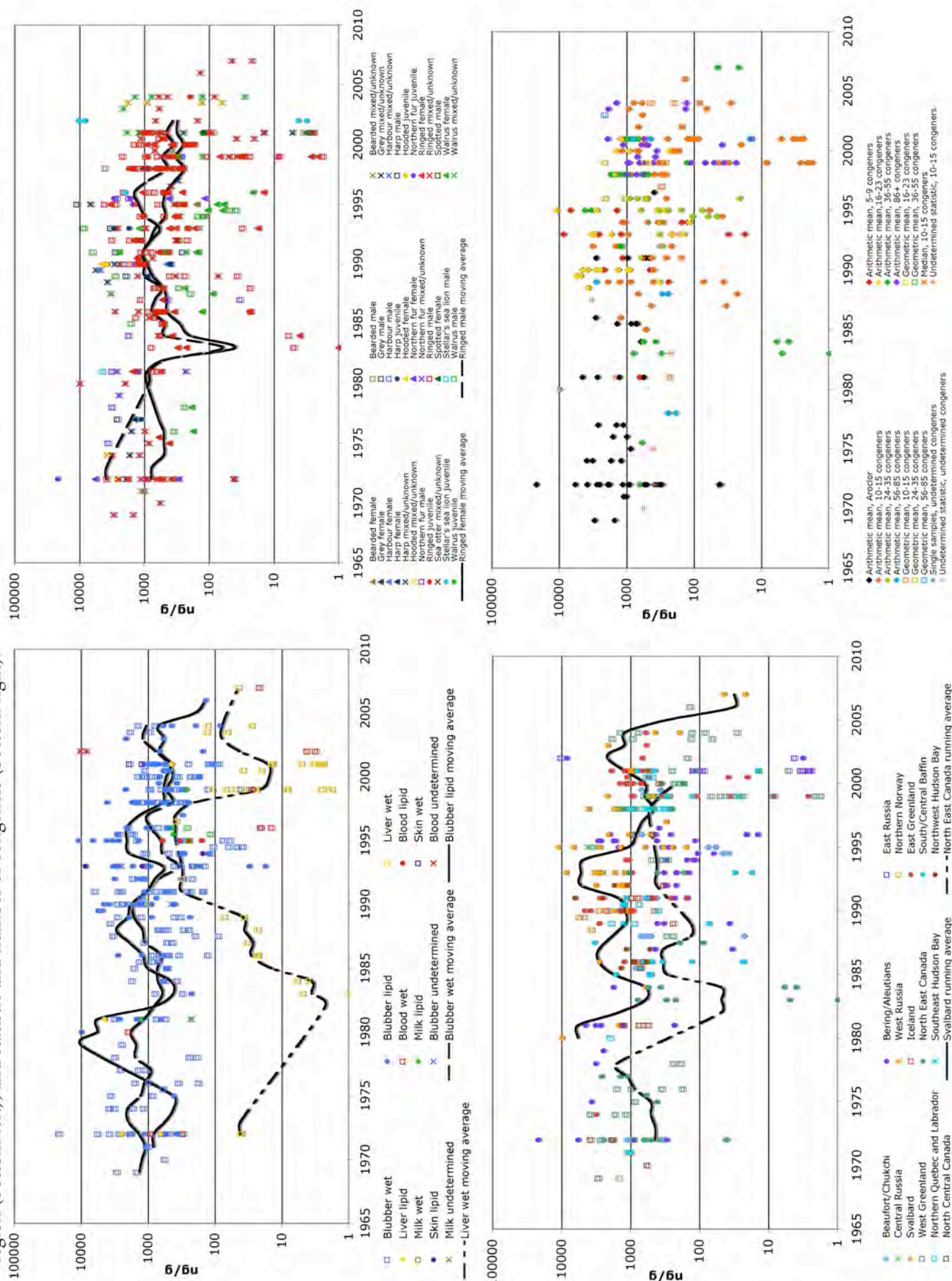


Figure 4.4. Σ PCB concentration results for cetaceans, grouped by tissue and weight measurement (top left), species and gender (top right), region (bottom left), and statistic and number of congeners (bottom right).

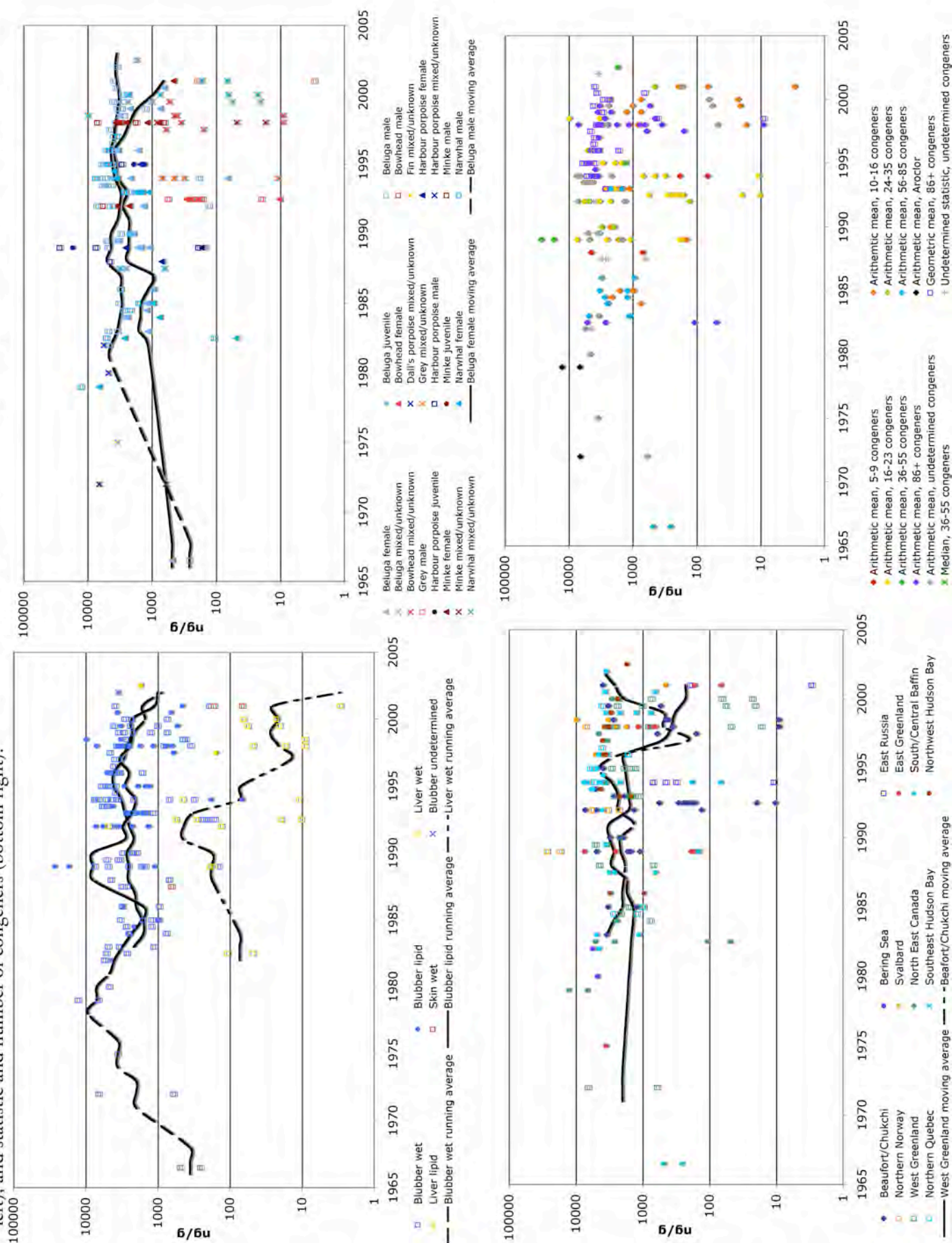


Figure 4.5. Σ PCB concentration results for polar bears, grouped by tissue and weight measurement (top left), gender (top right), region (bottom left), and statistic and number of congeners (bottom right).

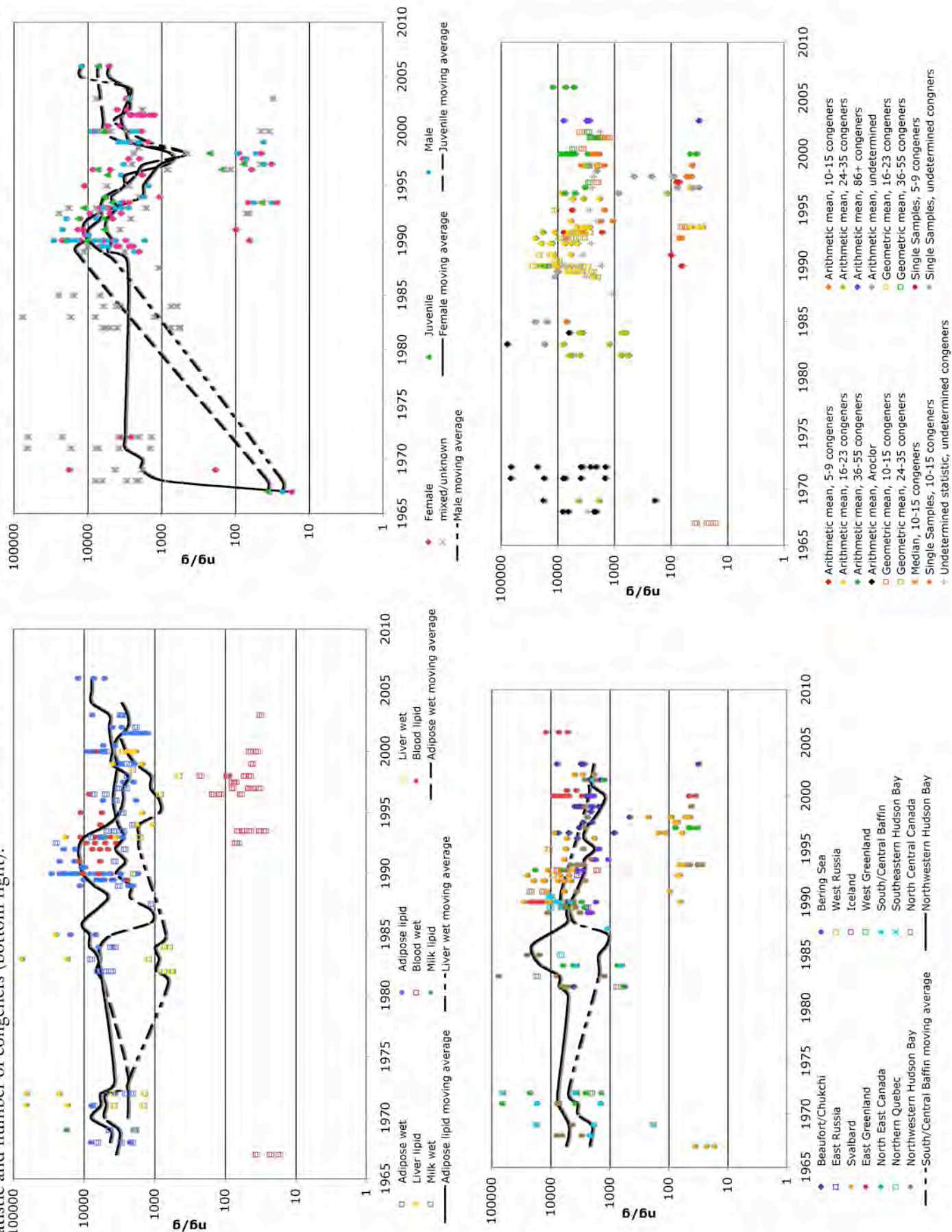


Figure 4.6. Σ PCB concentration results for humans, grouped by tissue and gender (top left), region (top right), and statistic and number of congeners (bottom). Note that all concentrations were reported as lipid-adjusted weights.

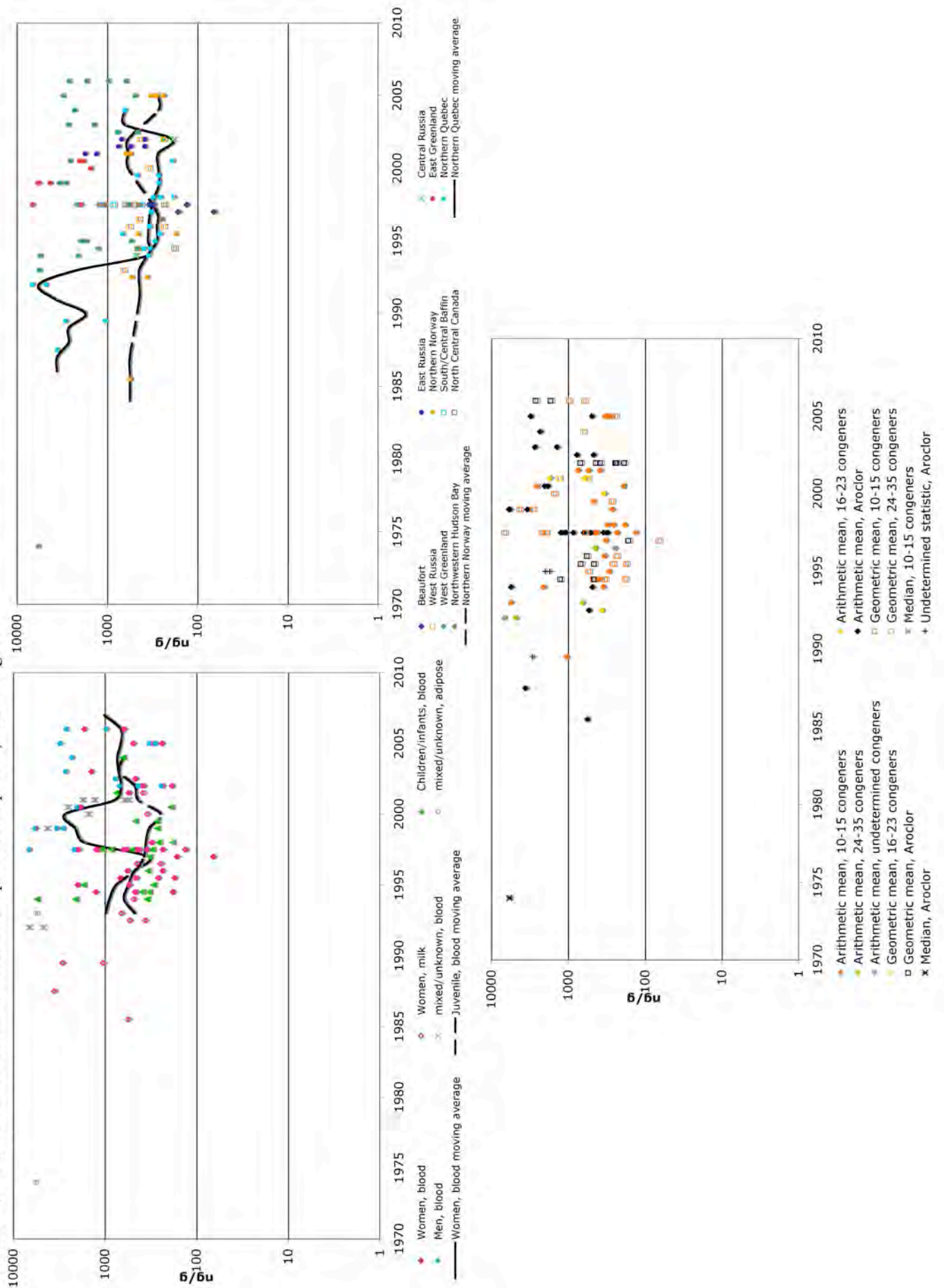


Figure 4.7. Σ PCB concentration results for terrestrial carnivores, grouped by tissue and weight measurement (top left), species and gender (top right), region (bottom left), and statistic and number of congeners (bottom right).

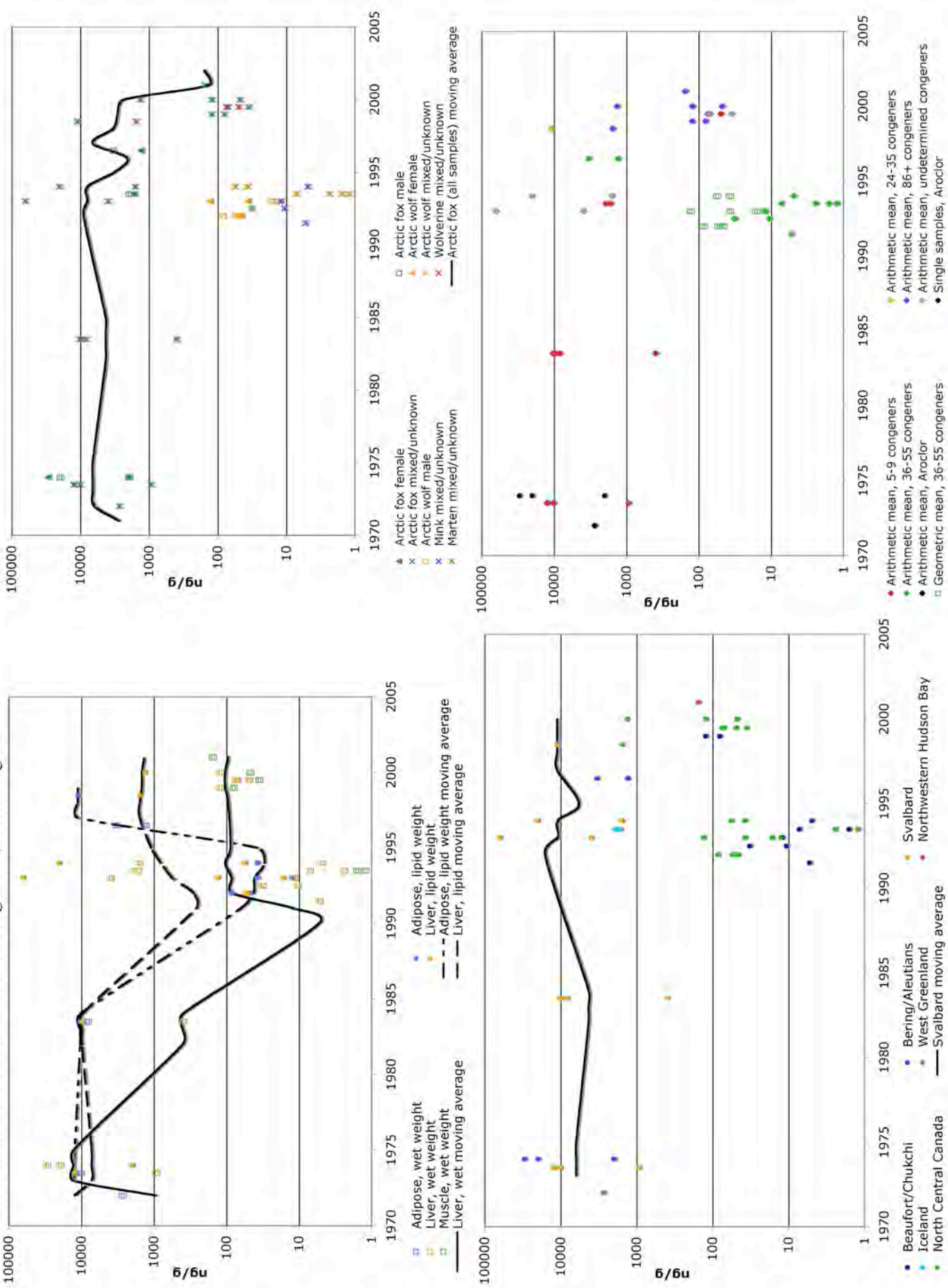
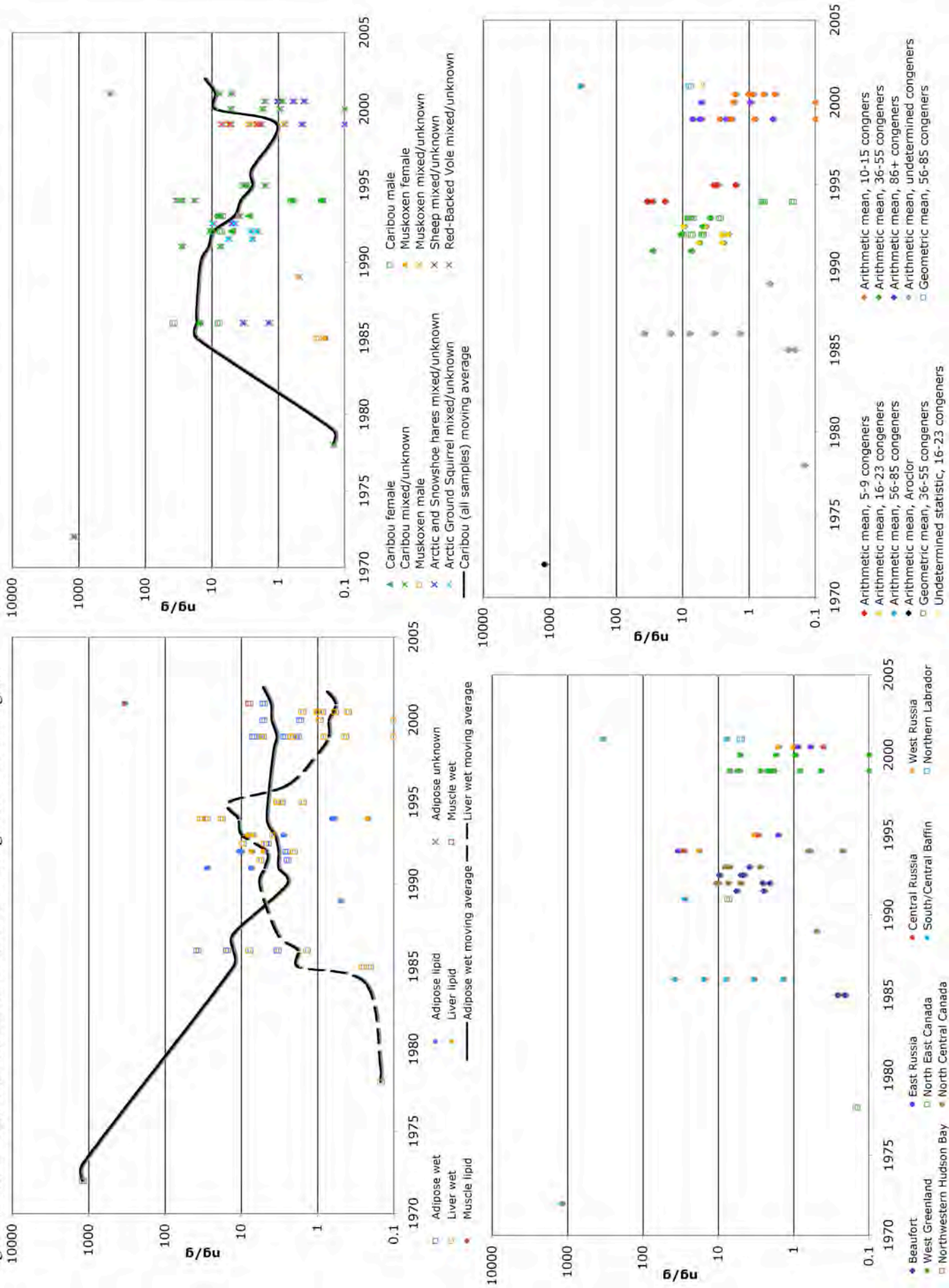


Figure 4.8. Σ PCB concentration results for terrestrial herbivores, grouped by tissue and weight measurement (top left), species and gender (top right), region (bottom left), and statistic and number of congeners (bottom right).



Mammal Group	References
Pinnipeds and Sea Otters	(Addison <i>et al.</i> , 2005; Addison and Smith, 1998; AMAP, 2004; Andersson <i>et al.</i> , 1988; Bacon <i>et al.</i> , 1992; Bacon <i>et al.</i> , 1999; Bang <i>et al.</i> , 2001; Barron <i>et al.</i> , 2003; Beck <i>et al.</i> , 1994; Becker <i>et al.</i> , 1989; Beckmen <i>et al.</i> , 1999; Born <i>et al.</i> , 1981; Bowes and Jonkel, 1975; Bowes and Lewis, 1974; Braune <i>et al.</i> , 2005; Calambokidis and Peard, 1985; Cameron and Weis, 1993; Cameron <i>et al.</i> , 1997; Clausen <i>et al.</i> , 1974; Cleemann <i>et al.</i> , 2000; Daelemans <i>et al.</i> , 1993; Dewailly <i>et al.</i> , 1993; Edelstam <i>et al.</i> , 1987; Espeland <i>et al.</i> , 1997; Fisk <i>et al.</i> , 2002; Helm <i>et al.</i> , 2002; Hoekstra <i>et al.</i> , 2003d; Holden, 1970; Hop <i>et al.</i> , 2002; Jansson <i>et al.</i> , 1993; Jarman <i>et al.</i> , 1992; Johansen <i>et al.</i> , 1980; Johansen <i>et al.</i> , 2003; Johansen <i>et al.</i> , 2004a; Johansen <i>et al.</i> , 2009; Kamrin and Ringer, 1994; Kannan <i>et al.</i> , 2008; Kelly <i>et al.</i> , 2007; Kleivane <i>et al.</i> , 1997; Kleivane <i>et al.</i> , 2000; Kostamo <i>et al.</i> , 2000; Krahn <i>et al.</i> , 1996; Kucklick <i>et al.</i> , 2005; Kucklick and Struntz, <i>unpublished</i> ; Kucklick <i>et al.</i> , 2002; Kuhnlein <i>et al.</i> , 1995; Kurtz, 1984; Kurtz, 1987; Kurtz and Kim, 1976; Lee <i>et al.</i> , 1996; Letcher <i>et al.</i> , 2009; Loughlin <i>et al.</i> , 2002; Muir, 1994; Muir, 1996; Muir <i>et al.</i> , 2000; Muir <i>et al.</i> , 2001; Muir <i>et al.</i> , 1992b; Muir and Kwan, <i>unpublished</i> ; Muir <i>et al.</i> , 1999c; Muir <i>et al.</i> , 1999b; Muir <i>et al.</i> , 1988; Muir <i>et al.</i> , 2003; Muir <i>et al.</i> , 1995; Muir <i>et al.</i> , 1992c; Myers <i>et al.</i> , 2008; Nakata <i>et al.</i> , 1998; Norstrom <i>et al.</i> , 1990; Oehme, 1990; Oehme <i>et al.</i> , 1995b; RAIPON <i>et al.</i> , 2001; Riget <i>et al.</i> , 2004; Riget <i>et al.</i> , 2003; Riget <i>et al.</i> , 2006; Ronald <i>et al.</i> , 1984; Routti <i>et al.</i> , 2008; Ruus <i>et al.</i> , 1999; Sandau <i>et al.</i> , 2000; Schantz <i>et al.</i> , 1993; Seagars and Garlich-Miller, 2001; Severinsen <i>et al.</i> , 2000; Skaare, 1995; Thomas and Hamilton, 1988; Vetter <i>et al.</i> , 1995; Vorkamp <i>et al.</i> , 2004; Vorkamp <i>et al.</i> , 2008; Wang-Andersen <i>et al.</i> , 1993; Wiig <i>et al.</i> , 2000; Wolkers <i>et al.</i> , 2008; Wolkers <i>et al.</i> , 2004; Wolkers <i>et al.</i> , 2006b; Wolkers <i>et al.</i> , 1998; Wolkers <i>et al.</i> , 1999; Zhu and Norstrom, 1993; Zhu <i>et al.</i> , 1995)
Cetaceans	(AMAP, 1998; AMAP, 2004; Andersen <i>et al.</i> , 2001a; Borrell <i>et al.</i> , 2004; Braune <i>et al.</i> , 2005; Bruhn <i>et al.</i> , 1999; Cameron and Weis, 1993; Clausen <i>et al.</i> , 1974; Dewailly <i>et al.</i> , 1993; Dietz <i>et al.</i> , 2004a; Granby and Kinze, 1991; Helm <i>et al.</i> , 2002; Hobbs <i>et al.</i> , 2002; Hobbs <i>et al.</i> , 2003; Hoekstra, <i>unpublished</i> ; Hoekstra <i>et al.</i> , 2003c; Hoekstra <i>et al.</i> , 2005; Hoekstra <i>et al.</i> , 2003d; Hoekstra <i>et al.</i> , 2002a; Hoekstra <i>et al.</i> , 2002b; Holden, 1975; Johansen <i>et al.</i> , 1980; Johansen <i>et al.</i> , 2004b; Kelly <i>et al.</i> , 2007; Kleivane <i>et al.</i> , 1995; Kleivane and Skaare, 1998; Kuhnlein <i>et al.</i> , 1995; Langlois and Langis, 1995; Letcher <i>et al.</i> , 2000; McKinney <i>et al.</i> , 2006; Metcalfe <i>et al.</i> , 1999; Muir, 1994; Muir, 1996; Muir <i>et al.</i> , 1999a; Muir <i>et al.</i> , 1992a; Muir <i>et al.</i> , 1990; Muir <i>et al.</i> , 1999b; Muir <i>et al.</i> , 1992c; O'Hara <i>et al.</i> , 1999; RAIPON <i>et al.</i> , 2001; Riget <i>et al.</i> , 2003; Sang <i>et al.</i> , 2000; Schantz <i>et al.</i> , 1993; Schantz <i>et al.</i> , 1996; Skaare, <i>unpublished</i> ; Stern, <i>unpublished</i> ; Stern and Ikonomou, 2003a; Stern, 1999; Stern and Ikonomou, 2003b; Stern <i>et al.</i> , 2005; Stern <i>et al.</i> , 1994; Tanabe <i>et al.</i> , 1983; Tilbury <i>et al.</i> , 2002; van Scheppingen <i>et al.</i> , 1996; Wade <i>et al.</i> , 1997; Wagemann and Muir, 1984; Wolkers <i>et al.</i> , 2006a)
Polar Bears	(AMAP, 2004; Andersen <i>et al.</i> , 2001b; Bandiera <i>et al.</i> , 1997; Bentzen <i>et al.</i> , 2008a; Bentzen <i>et al.</i> , 2008b; Bergman <i>et al.</i> , 1994; Bernhoft <i>et al.</i> , 2000; Bernhoft <i>et al.</i> , 1997; Bowes and Jonkel, 1975; Braathen <i>et al.</i> , 2004; Braune <i>et al.</i> , 2005; Clausen <i>et al.</i> , 1974; Corsolini <i>et al.</i> , 2002; Derocher <i>et al.</i> , 2003; Dewailly <i>et al.</i> , 1993; Dietz <i>et al.</i> , 2004b; Evans, 2001; Gebbink <i>et al.</i> , 2008; Haave <i>et al.</i> , 2003; Jarman <i>et al.</i> , 1992; Kannan <i>et al.</i> , 2005; Klobes <i>et al.</i> , 1998; Krahn <i>et al.</i> , <i>unpublished</i> ; Kucklick <i>et al.</i> , 2002; Kuhnlein <i>et al.</i> , 1995; Kumar <i>et al.</i> , 2002; Letcher <i>et al.</i> , 2003; Letcher <i>et al.</i> , 1995; Letcher <i>et al.</i> , 1996a; Letcher <i>et al.</i> , 1996b; Lie <i>et al.</i> , 2000; Lie <i>et al.</i> , 2005; Lie <i>et al.</i> , 2004; Norstrom, 2001; Norstrom <i>et al.</i> , 1998; Norstrom <i>et al.</i> , 1988; Oehme <i>et al.</i> , 1995a; Olsen <i>et al.</i> , 2003; Oskam <i>et al.</i> , 2003; Oskam <i>et al.</i> , 2004; Polischuk <i>et al.</i> , 2002; Riget, <i>unpublished</i> ; Sandala <i>et al.</i> , 2004; Sandau, 2000; Sandau <i>et al.</i> , 2000; Skaare, 1995; Skaare <i>et al.</i> , 2001; Sonne <i>et al.</i> , 2004; Sonne <i>et al.</i> , 2006b; Sonne <i>et al.</i> , 2005a; Sonne <i>et al.</i> , 2009a; Sonne <i>et al.</i> , 2006c; Verreault <i>et al.</i> , 2005; Wiig <i>et al.</i> , 1998; Zhu and Norstrom, 1993; Zhu <i>et al.</i> , 1995)
Humans	(AMAP, 2009; Anda <i>et al.</i> , 2007; Ayotte <i>et al.</i> , 1997; Becher <i>et al.</i> , 1995; Bjerregaard <i>et al.</i> , 2001; Bjerregaard and Hansen, 2000; Dallaire <i>et al.</i> , 2003; Dallaire <i>et al.</i> , 2004; Dallaire <i>et al.</i> , 2009; Despres <i>et al.</i> , 2005; Deutch <i>et al.</i> , 2006; Deutch and Hansen, 2000; Deutch <i>et al.</i> , 2007; Deutch <i>et al.</i> , 2004; Dewailly <i>et al.</i> , 1993; Dewailly <i>et al.</i> , 1999; Dewailly <i>et al.</i> , 1992; Dewailly <i>et al.</i> , 1989; Jensen and Clausen, 1979; Jorgensen <i>et al.</i> , 2008; Klopov <i>et al.</i> , 1998; Newsome and Ryan, 1999; Polder <i>et al.</i> , 1998; Polder <i>et al.</i> , 2008; Polder <i>et al.</i> , 2003; Rylander <i>et al.</i> , 2009; Sandanger <i>et al.</i> , 2003; Sandanger <i>et al.</i> , 2006; Van Oostdam <i>et al.</i> , 1999; Van Oostdam <i>et al.</i> , 2004; Walker <i>et al.</i> , 2003; Zhu <i>et al.</i> , 1995)
Terrestrial Carnivores	(AMAP, 1998; AMAP, 2004; Braune <i>et al.</i> , 2001; Elkin, <i>unpublished</i> ; Fuglei <i>et al.</i> , 2007; Hoekstra <i>et al.</i> , 2003a; Hoekstra <i>et al.</i> , 2003b; Kelly and Gobas, 2001; Klobes <i>et al.</i> , 1998; Krahn <i>et al.</i> , <i>unpublished</i> ; Norheim, 1978; Poole <i>et al.</i> , 1995; Poole <i>et al.</i> , 1998; Skaare, <i>unpublished</i> ; Wang-Andersen <i>et al.</i> , 1993; White and Risebrough, 1977)
Terrestrial Herbivores	(Allen-Gil <i>et al.</i> , 1997; AMAP, 1998; AMAP, 2004; Clausen <i>et al.</i> , 1974; Elkin and Bethke, 1995; Johansen <i>et al.</i> , 2004a; Mallory <i>et al.</i> , 2005; Melnikov <i>et al.</i> , 1995; Muir <i>et al.</i> , 1992c; Pollock <i>et al.</i> , 2009; Poole <i>et al.</i> , 1998; RAIPON <i>et al.</i> , 2001; Salisbury <i>et al.</i> , 1992; Thomas and Hamilton, 1988; Thomas <i>et al.</i> , 1992)

Figure 4.9. Reference citations for concentration data listed by mammal group.

5 Discussion

5.1 Pinnipeds and sea otters

As seen in figure 4.1, the most data by far were available for pinnipeds and sea otters, with nearly twice as many datapoints as the next largest group (cetaceans). This abundance of data points toward a storied role for seal species as subjects for environmental contaminant monitoring. Though the earliest samples in this survey do not come from pinnipeds or sea otters (the earliest samples were collected from beluga whales in 1966-1967, (Muir *et al.*, 1990)), the earliest article in this survey was written about them. In 1970, Holden *et al.* reported the first incidence of PCB contamination in an Arctic animal (ringed seals), 4 years before PCBs were reported in any other Arctic mammal. Though sea otters are phylogenetically more closely related to the mink, marten, and wolverine included in the terrestrial carnivore group (they are all in the mustelid family), they are ecologically much more similar to pinnipeds, living their entire life at sea (even giving birth in the water) and predating mostly benthic invertebrates and some fish (Kannan *et al.*, 2008). Therefore, the two sea otter datapoints were grouped with the pinniped data.

Given the physiological thresholds discussed in section 3.3, the data presented in figure 4.2 seem to indicate that many Arctic pinnipeds and sea otters experience at least some neurological effects related to PCB exposure, though the majority does not, and direct associations between PCBs and immune or reproductive effects are much less likely. When all data are considered, a strongly overall decreasing, if variable, historical trend of PCB levels appears to be present for pinnipeds and sea otters. However, when the data is controlled for tissue type (figure 4.3, top left), the support for this trend is weakened. When only blubber data are considered, a different trend emerges that is still decreasing slightly overall, but perhaps more interesting, appears to show cyclic changes in PCB levels. These cycles, with an almost constant period of about 8 years, are perhaps related to the periodicity of various climatological phenomena that influence transport of PCBs into the Arctic. Similar cyclic trends were observed independently in both lipid and wet weight measurements of blubber samples, confirming this possibility. Wet weight data from liver, the only other tissue with sufficient data to assess long-term historical trends, demonstrated an overall increasing trend since the early 1980s, bringing the trend seen in figure 4.2 into further debate. As expected, wet weight concentrations from blubber were greater than those of any other tissue, though this appeared to be less the case with lipid weight concentrations. Within the same tissue, the lipid weight values are higher than the wet weight values, as expected, in years where there is sufficient data for each.

The most represented pinniped species in the data by far was the ringed seal, with 230 datapoints, 63% of the total for pinnipeds and sea otters. Though it is the largest pinniped species covered in this review and the third largest pinniped in the world, the walrus displayed relatively low Σ PCB concentrations (figure 4.3, top right), owing to its predation almost exclusively on benthic invertebrates that are lower in the trophic web and have lower contaminant loads than fish (Blix, 2005; Muir *et al.*, 1995). Northern fur seals, in contrast, appeared to have rather high Σ PCB concentrations, especially from 1970 – 1985 (where the most data are available for them), perhaps because their more southerly ranges around the Bering Sea, Aleutians, and Alaska Peninsula are closer to temperate sources of

PCBs from the Pacific coast of North America and eastern Russia and Asia. Where sufficient data exist for both genders, Σ PCB concentrations in males appeared to be somewhat higher than those in females for the same species, as expected. The 3-year moving averages calculated for male and female ringed seals followed similar historical trends, independently confirming the overall historical trend for this species.

The bottom left graph of figure 4.3 shows historical trends of Σ PCB concentrations in pinnipeds and sea otters by region (refer to figure 3.3 for region designations). Regions that are relatively close to one another share the same color (but different shapes) in the graph. In early years, the Bering/Aleutian region appears to have supported the highest Σ PCB concentrations. However, Svalbard and East Greenland appear to have the highest levels in later years, when data for these regions becomes more available. When broken down by region in this manner, it appears that Σ PCB concentrations experienced an overall decline within each region, perhaps indicating a general declining trend across the circumpolar Arctic. Similar historical patterns are seen in regions as far apart as Svalbard and northeastern Canada, especially from about 1978 to 1992 (as revealed by their moving averages), lending support to this idea. The substantive differences in Σ PCB concentrations between regions such as East Greenland and West Greenland confirms the role oceanic circulation patterns play in PCB transport (though they share a land mass, East Greenland and West Greenland border different water masses and oceanic current systems). A few of the published studies analyzed pinniped samples from both Arctic and temperate locations. These studies revealed that temperate populations showed much greater levels of PCBs than Arctic populations of the same species, presumably because they are closer to industrial sources of PCBs (e.g. Routti *et al.*, 2008; Beck *et al.*, 1994).

Finally, it is difficult to assess long-term trends based on the number of congeners reported (figure 4.3, bottom right), because it appears that this parameter is largely dependent on when PCB analyses were conducted. The majority of early samples (1967 – 1982) were calibrated with Aroclor, and the number of congeners analyzed appears to increase with time. Samples analyzed for greater than 86 congeners were reported only from about 1997 on. Nevertheless, where sufficient data exist, Σ PCB concentrations that include greater numbers of congeners are higher than those including lower numbers, though it is difficult to determine how this impacts the overall trends observed for this group of mammals. When they coexist, Aroclor measurements appear to be much higher than those calibrated from individual congeners, but due to relatively few data from the latter for comparison, it is difficult to assess whether this is symptomatic of an overestimation bias with Aroclor (see discussion in section 3.2). As a statistical rule, geometric means are always equal to or less than their corresponding arithmetic means. However, it is unclear whether reporting data in geometric means and medians significantly changes the observed trends from what would be seen if arithmetic means for these data were reported instead.

5.2 Cetaceans

Unlike pinnipeds and sea otters, the cetacean data in figure 4.2 seem to indicate that the majority of whales in the Arctic, particularly odontocetes (toothed whales, see top right graph of figure 4.4), likely experience some neurological effects of PCB exposure based on the physiological thresholds, though incidence of immune and reproductive effects directly related to PCB exposure is minimal. The overall historical trend for PCB contamination in

cetacean species from the Arctic (figure 4.2) appears to be slightly increasing, if highly variable. When the data are grouped by tissue (figure 4.4, top left), a somewhat subtler trend is revealed, indicating an increase followed by a decrease for all series where there is sufficient data for a long-term historical analysis. The upturn seen in the trend for all data from about 1999 – 2003 appears to be predominantly attributed to a single liver lipid weight datapoint, indicating the importance of controlling for biases from the type of tissue analyzed. As with pinnipeds, the wet weight blubber samples displayed much higher PCB loads than those for other tissues. The number of wet weight measurements far exceeded lipid weight measurements, making comparisons between lipid-adjusted values for different tissues difficult. Overall, little difference between wet and lipid weight measurements within the same tissue was observed, when sufficient data existed for both.

The top right graph of figure 4.4 groups the data by species and reveals a relatively strong partition in PCB levels between the odontocetes (toothed whales, shades of blue) and mysticetes (baleen whales, shades of red and orange), two groups that are quite different anatomically, phylogenetically, and ecologically. Though some of this distinction may be attributed to the overrepresentation of mysticetes in tissues with lower PCB levels (especially liver, compare to the top left graph), most of the lower values reported for blubber also come from mysticetes (particularly bowhead and grey whales). This distinction, if real, is due to the lower trophic level of mysticete species that predate mostly krill and small amphipods, as opposed to the odontocetes that predate fish and larger invertebrates like squid. The data from mysticete species also appeared to group into small, discrete time intervals and are largely only available from the 1990s, making long-term historical analyses difficult. The levels of PCBs in beluga alone appeared to be quite stable historically (excluding the two isolated pre-1980 datapoints from 1966-1967), indicating the absence of any increasing or decreasing trend for this species. While a moving average is not shown from narwhal, a very closely related species (shown in a slightly darker shade of blue), the data from this species appears to match the beluga data quite well. The lack of long-term data for the other odontocete species (Dall's and harbour porpoises) makes it difficult to establish historical trends. As with pinnipeds, males tended to display greater contaminant loads than females for all species, and the historical trends for beluga appeared to be relatively similar between the two genders, though PCB levels in females appear to have declined significantly since 1995 while those in males were more stable.

In contrast to the data for pinnipeds and sea otters, the regional trends (figure 4.4, bottom left) for cetacean data appear less conclusive overall, due to a lack of sufficient historical data for many regions and an overrepresentation of mysticetes in certain regions, particularly Beaufort/Chukchi. Still, the data from South/Central Baffin region (light blue circles, moving average not shown), which was relatively plentiful and represented odontocete species only, indicated a slight increase to about 1994 followed by a slight decrease. Note that the relatively low values for this region from 1994 are most likely attributed to the fact that these concentrations were reported as the sum of only 5-9 congeners (bottom right graph). Furthermore, the moving averages for West Greenland and Beaufort/Chukchi, two regions that are relatively distant from one another and contained substantial mysticete data, appeared to show at least some correlation in historical trends, particularly from about 1984 to 1998. Finally, similar to the results for pinnipeds, studies that analyzed cetacean samples from both Arctic and temperate locations revealed that temperate populations displayed greater levels of PCBs than Arctic populations of the same

species, likely due to their closer proximity to sources of PCBs in temperate areas (e.g. McKinney *et al.*, 2006; Bruhn *et al.*, 1999).

Like pinnipeds, the number of congeners (figure 4.4, bottom right) reported appears to be largely dependent on the year of sampling or publishing, making long-term assessments difficult. Nonetheless, where multiple measurements are reported (e.g. 1984-1985 and 1999-2001), values are greater for measurements calibrated with greater numbers of congeners, as anticipated, though these relatively few instances would not appear to significantly alter the other trends observed. Only 3 datapoints were calibrated with Aroclor, and so do not warrant further analysis. For data reported as the sum of greater than 86 congeners, there did not appear to be any substantive difference between values reported as geometric versus arithmetic means.

5.3 Polar bears

Polar bears fill a unique ecological niche, due to their role as an apex predator, keystone species, and the only marine mammal completely adapted to and dependent on sea ice, as opposed to water. This unique role, as well as the abundance of data for this species (second only to ringed seals) and its recent popularity as a “charismatic megafauna” and symbol of global climate change, led it to be considered separately in this analysis from other mammal groups.

Based on the complete dataset for polar bears (in figure 4.2) and the established physiological thresholds, it appears that the vast majority of polar bears likely experience some neurological effects of PCB exposure, and unlike pinnipeds or cetaceans, many also likely experience immune and reproductive effects directly related to PCB exposure. That a more significant incidence of immune and reproductive effects is seen in polar bears in the field (see discussion in section 2.2.3.1) can be explained in that these observed effects are due to a number of different contaminants and factors in combination with PCBs, that the thresholds were established in laboratory experiments designed to assess the exclusive and direct effects of PCBs, and that the data considered here are averages that do not convey the frequency of more significant effects in individuals with greater than average concentrations of Σ PCBs.

The moving average calculated from all polar bear data (in figure 4.2) indicates that historical levels of PCBs have fluctuated quite dramatically, while no overall increasing or decreasing trend is observed since 1968. Complicating all historical trend analyses for polar bears in the absence of data from the period 1972-1982. When the data are grouped by tissue (figure 4.5, top left), it appears that PCB levels have roughly stayed the same or slightly increased in all tissues, at least since 1995. Like pinnipeds and cetaceans, wet weight measurements in adipose (fat) tissue were significantly greater than those in liver and blood. However, in contrast to pinnipeds and cetaceans, lipid weight measurements in liver were much greater than those in adipose (at least from 1970-1985). Polar bears, like terrestrial mammals, are much more dependent on the insulative property of their fur rather than large fat reserves to keep them warm (as used by pinnipeds and cetaceans). Therefore, because their adipose layer is much less significant, perhaps polar bears preferentially shunt PCBs to the liver instead. Finally, lipid-adjusted values were significantly greater than wet weight values within the same tissue, and this is particularly striking for blood, which has a relatively low lipid percentage.

The top right graph of figure 4.5 shows temporal trends of Σ PCB concentrations based on gender (and age). The historical patterns for males, females, and juveniles (of mixed gender) are extremely well correlated, especially considering that the apparent divergence from 1969 – 1989 is solely because data from females only exists for this time period. That these three independent groups display a similar pattern is confirmation of a possible general historical trend seen for all polar bears. As expected, concentrations from males and juveniles were greater than those from females, except for a short period from 1995-1999.

As in pinnipeds, all regions appear to demonstrate an overall decreasing trend of PCB levels in polar bears, with the possible exceptions of East Greenland and Beaufort/Chukchi, both of which appear to decrease and then increase (figure 4.5, bottom left). As confirmation of this general trend, the moving averages for South/Central Baffin and Northwestern Hudson Bay showed similar decreases from 1990 onward. Svalbard and East Greenland appear to support much greater levels of PCBs than any other region when the wet weight blood data in the lower portion of the graph are disregarded, for which these regions are overrepresented. This is not surprising, given that the majority of physiological effects of PCB exposure on polar bears (see section 2.2.3.1) were observed predominantly in populations from these regions, and that pinnipeds, which comprise the majority of the polar bear's diet, also displayed the highest levels of PCBs in these two regions (see section 5.1).

Finally, like pinnipeds and cetaceans, it is difficult to determine trends and relationships for data grouped by the number of congeners and statistic reported (figure 4.5, bottom right) for the reasons discussed. The Aroclor-calibrated values were greater than those calibrated with individual congeners, though the lack of a significant time overlap in the data still precludes a firm conclusion that Aroclor calibration leads to overestimation.

5.4 Humans

Indigenous human communities exist throughout all regions of the circumpolar Arctic, with the exception of Svalbard. Though accessibility to “market foods” imported from temperate industrial regions has greatly increased in recent decades, many of these communities are still dependent on traditional food sources, particularly marine mammals and caribou (depending on the region), to ensure adequate nutrition and cultural survival, and because they are often still cheaper than imported food (Theriault *et al.*, 2005). Therefore, because these communities accumulate PCBs from their food sources, long-range transport of PCBs to the Arctic is not just a wildlife toxicology issue, but also a global human health issue.

The level of exposure to PCBs in these communities is often significant. Some studies showed that daily intake of PCBs exceeded the World Health Organization's “tolerable daily intake” in communities in Greenland (Johansen *et al.*, 2004b), Nunavik (northern Quebec, (Dewailly *et al.*, 1993)), and Baffin Island (Kuhnlein *et al.*, 1995). Intake of PCBs was even greater in earlier years when a greater proportion of the diet of indigenous people in the Arctic came from traditional food sources (and possibly also when PCB levels in those food sources were greater). One study showed that daily intake of PCBs (measured as Aroclor 1260) in a West Greenland community in 1976 was over 3 times greater than that of 2004 (Deutch *et al.*, 2006). PCB concentrations in blood samples from communities throughout the circumpolar Arctic from 1997-2007 were consistently greater than the

Canadian public health department's "level of concern", with sometimes as much as 90% of those sampled exceeding this value, and some samples from one community in northwest Greenland exceeded the department's "level of action" (AMAP, 2009).

When average PCB concentration data from humans in the Arctic are compared to established physiological thresholds (figure 4.2), it appears likely that many individuals in the communities sampled experience at least some neurological effects, though there may be no direct correlations with immune or reproductive effects. As with the polar bear data, this does not mean that associations between PCB levels and these effects do not exist, and evidence of these effects in humans have been found (see section 2.2.3.3). The historical trend for PCB levels in humans appears to have decreased overall but has been slowly increasing since 1997. Because all concentration values from humans were lipid-adjusted, the tissue and gender parameters were considered together (figure 4.6, top left). Note that the children/infant blood data includes blood samples collected from umbilical cords at birth. Like the overall data, the historical trends for each gender and tissue group also appear to have decreased and then increased, though they reached their minima in different years. Several of the blood data were taken from children 4-6 years after their birth, whose mothers had also been sampled when they were pregnant with these children (Despres *et al.*, 2005). Interestingly, this time lag is seen in the moving averages for blood data, with the trend for children reaching its minimum several years after the minimum for women, revealing the PCB load these children received from their mothers *in utero*.

When grouped by region (figure 4.6, top right), West and East Greenland, and earlier, Nunavik, emerge as the regions supporting the highest levels of PCBs in indigenous human communities. The values from these regions are significantly higher than those from anywhere else in the circumpolar Arctic. Within regions, PCB levels appear to have declined overall, though limited long-term data for regions such as Beaufort and East Russia make it difficult to assess whether these trends can be extended to the Arctic region at large. Unlike pinnipeds and cetaceans, people living in the Arctic often display higher levels of PCBs than those living in temperate regions, an observation most likely attributed to their consumption of foods with greater concentrations of PCBs. For example, Inuit from communities throughout Nunavik had levels of PCBs that were 32 times greater than those seen in people from the urban centers of Quebec City and Montreal (Ayotte *et al.*, 1997).

Unlike studies of other mammals in the Arctic, PCB studies on humans have continued to use Aroclor for calibration of samples, even through the 2000s. When compared to values obtained from the sum of concentrations of individual congeners (figure 4.6, bottom), the corresponding Aroclor values are much greater. However, because the individual congener data only represent relatively few (generally 10-15) congeners, it is difficult to say whether the Aroclor values are inflated.

5.5 Terrestrial carnivores

As discussed in section 2.2.2, terrestrial species generally show lower concentrations of PCBs than marine species. The exceptions to this generalization are carnivorous species like the arctic fox and wolverine that exploit food sources from both the terrestrial and marine Arctic ecosystem (Hoekstra *et al.*, 2003b). The lack of sufficient PCB data for most terrestrial carnivore species required that they be grouped and analyzed together. It should be noted however, that this group contains species that are diverse ecologically and

physiologically, though only one phylogenetic order (carnivora) and two phylogenetic families (canids and mustelids) are represented.

Most PCB concentrations from Arctic terrestrial carnivores (figure 4.2) are below any established physiological thresholds. However, several concentrations do exceed the threshold for neurological effects, and some exceed those for immune and reproductive effects as well. All data exceeding the neurological threshold come from either arctic foxes or wolverines (figure 4.7, top right), and data exceeding the immune and reproductive thresholds come from arctic foxes only, as expected from the mixed marine and terrestrial diets of these species. The historical trend from all data appears to fluctuate quite dramatically over time. The patterns for different tissues and weight measurements (figure 4.7, top left) seem to correlate well with each other and the trend for all data. Like the polar bear data, the liver appears to be a more significant reservoir for PCBs than adipose in these terrestrial carnivores, as the liver data was consistently greater than most adipose data, even when measured as wet weight.

When controlled for species, it is evident that the sharp decrease seen in the previous two graphs in the early 1990s is largely due to the inclusion of data from other species (arctic wolf, mink, marten) whose diets are more strictly terrestrial, and therefore have lower levels of PCBs. However, a dip is still seen in 1995 in the arctic fox, the only species with sufficient long-term data to calculate a historical moving average. Overall, the concentrations in arctic foxes appear to have increased then decreased beginning in 1997. However, this recent decrease is likely caused in part by an abundance of wet weight measurements from liver and muscle from 1999-2001 (top left graph). Most data for terrestrial carnivores represented mixed or unknown gender samples, making assessments of gender differences in PCB levels difficult.

Only the Svalbard region contained enough long-term data to permit a temporal analysis based on region (figure 4.7, bottom left). In Svalbard, the PCB levels appear to have steadily increased since 1983. The Svalbard and Bering/Aleutian regions supported concentrations of PCBs that were much higher than those from other regions, but this is solely because data from arctic foxes only were available for these regions, and most data from this species came from these two regions. Like pinnipeds and cetaceans, PCB levels in wolves and minks from Arctic locations were generally lower than those from temperate areas not included in the data (Elkin, *unpublished*; Gamberg and Braune, 1999; Poole *et al.*, 1995; Poole *et al.*, 1998), presumably because more temperate populations are closer to sources of PCBs.

As with other mammal groups, no category based on number of congeners (figure 4.7, bottom right) contained sufficient long-term data to make a historical assessment. Furthermore, biases related to which species and tissues were reported prevent comparisons in levels between these different categories.

5.6 Terrestrial herbivores

Like the terrestrial carnivores, the paucity of data for herbivores necessitated the grouping of very diverse species together. This terrestrial herbivore group includes representatives from the lagomorph (hares), rodent (arctic ground squirrel and red-backed vole) and artiodactylid (caribou, muskox, sheep) phylogenetic orders. Note that the terms “reindeer” and “caribou” are used to describe different subspecies of the same animal, but

this study will refer to all populations as “caribou”. While they are not native to the arctic, domestic sheep from Greenland were still included in this study because presumably they consume similar vegetation, and therefore possibly PCB loads, as the indigenous herbivore species. As expected, all PCB concentration values but one were very low and below all the physiological thresholds (figure 4.2). The only outlying sample was from a sheep that was calibrated with Aroclor in 1972 (figure 4.8).

The historical trend based on all data appears to fluctuate somewhat but remains at the same level overall since 1985 (figure 4.2). This trend is replicated in the wet weight adipose data (figure 4.8, top left). The wet weight liver data, however, indicates a decline since 1995. In contrast to the polar bear and terrestrial carnivores, adipose tissue overall appears to be a more significant reservoir for PCBs than the liver in terrestrial herbivores, as evidenced by both lipid and wet weight values for these tissues.

Most of the terrestrial herbivore data come from caribou, and it is only for this species that a moving average could be calculated. The trend for caribou indicates a steady decline in Σ PCB concentrations beginning in 1986, followed by a sharp increase in 1999. Though the data from snowshoe and arctic hares is less abundant, the PCB concentrations reported in these two closely related species seem to fit the caribou curve quite well, possibly confirming a general trend for these herbivores. PCB levels in muskox, in contrast, appear to have increased from 1985 to 1999. Except for the outlying sheep datapoint, caribou appear to have the highest concentrations of PCBs overall, though levels in arctic ground squirrel, red-backed vole, and (more recently) sheep are close. When data for the separate genders is available, males show higher levels of PCBs than females, as expected.

No region analyzed (figure 4.8, bottom left) contained sufficient data for a long-term historical assessment of PCB levels within different regions, with the possible exception of South/Central Baffin, which appears to show an increase since 1986. Furthermore, that each short time interval contains data only from one or two regions significantly weakens all the historical trends discussed previously in this section, because they could just be artifacts of the differences in PCB levels between regions. The only inter-regional comparison that can be made with any sort of confidence is that North Central Canada appears to support higher levels of PCB than the Beaufort Sea region to the west. Interestingly, unlike pinnipeds, cetaceans, and terrestrial carnivores, red-backed voles from Arctic locations had higher levels of PCBs than those from temperate locations (Poole *et al.*, 1998), though it is unclear why this would be so.

The sheep sample from 1972 was the only value obtained by Aroclor calibration, while the first sample known to be calibrated with individual congeners was not until 1991 (figure 4.8, bottom right). Like the other mammal groups, it is difficult to say how differences in the number of congeners included in Σ PCB, or in which statistic was reported, influenced the concentration values from terrestrial herbivores.

5.7 Trophic trends

Due to biomagnification, species that are higher in the trophic food web accumulate greater contaminant loads than those that are lower down. To appropriately assess if biomagnification is at work in Arctic ecosystems, it is beneficial to use a measure that quantifies “trophic position” in order to determine valid statistical correlations with PCB levels. The ratio of stable isotopic ^{15}N to ^{14}N (referred to as $\delta^{15}\text{N}$) has been used as such a

measure, because it is thought to increase between trophic levels at a near-constant rate of 2.4‰ in birds and 3.8‰ in most other organisms (Fisk *et al.*, 2001), though it should be used with caution because a number of different factors can undermine the fidelity of these numbers (Dehn *et al.*, 2006). Nevertheless, $\delta^{15}\text{N}$ as a measure of trophic position has been shown to correlate well exponentially with PCB concentrations in studies from individual Arctic ecosystems in the Barents Sea (Hop *et al.*, 2002), Beaufort/Chukchi Seas (Hoekstra *et al.*, 2003d), and Nunavik/Southeastern Hudson Bay (Kelly *et al.*, 2007). To see if this correlation would hold for the entire Arctic area and over a longer time period, average lipid weight PCB levels in various species from blubber or adipose (with the exception of blood from humans) from 1990-2007 were compared to the $\delta^{15}\text{N}$ values for these species, which were averages from several different locations in the Arctic when multiple data were available (Dehn *et al.*, 2006; Kelly *et al.*, 2007). The results are presented in figure 5.1.

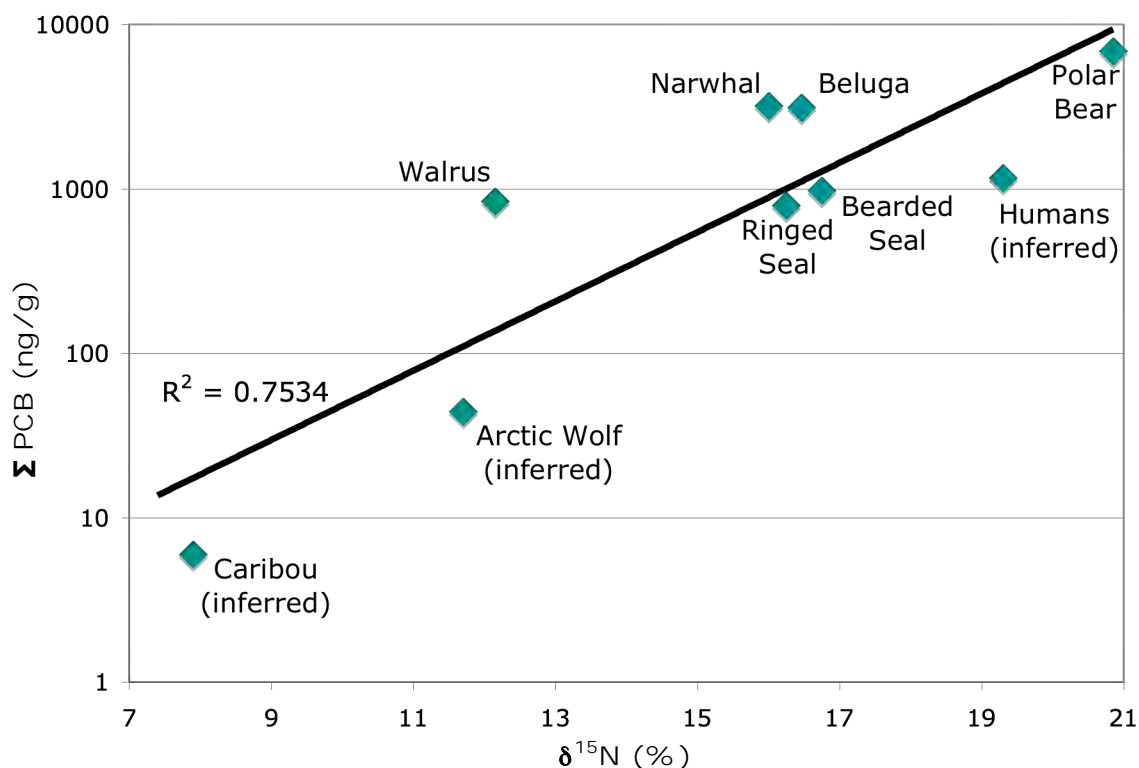


Figure 5.1. Correlation between trophic position (quantified as $\delta^{15}\text{N}$) and average ΣPCB concentrations from 1990-2007 for various species from throughout the circumpolar Arctic. $\delta^{15}\text{N}$ values for caribou, arctic wolf, and humans are inferred by back-calculation from “trophic level” values in (Kelly *et al.*, 2007), which is a regular function of $\delta^{15}\text{N}$. Note the log scale of the y-axis.

A striking and statistically supported exponential correlation (note the log scale) is observed between $\delta^{15}\text{N}$ and ΣPCB concentrations in figure 5.1. Furthermore, two of the more significant outliers in this correlation (walrus and humans) can be accounted for. Much of the walrus data came from a population thought to predate more on seals, and therefore occupy a higher trophic position, than the populations that were used to calculate the $\delta^{15}\text{N}$ for this species, resulting in ΣPCB values that appear too high for its calculated trophic position (Muir *et al.*, 1995). The ΣPCB values for humans came from blood lipids, which may support lower concentrations of PCBs than adipose tissue, depending on where

PCBs are preferentially shunted to in humans. Alternatively, the human populations where the Σ PCB values came from could depend less on traditional food sources, and therefore have lower levels of PCBs, than the population that was used to calculate the trophic position of humans in the Arctic. Still, this significant correlation is a good indication that PCBs are indeed biomagnifying in ecosystems throughout the circumpolar Arctic.

5.8 Regional trends

The discussion in sections 5.1 – 5.6 seemed to indicate that when regions are considered separately, the Σ PCB concentrations in each region have declined over time, and that trends for data from throughout the Arctic are obfuscated by regional differences and the different years when samples from different regions are reported. To further confirm this, it would be advantageous to control for the other parameters considered in this study. However, doing so severely reduces the amount of data available. Therefore, because it had the most data by far of any species, the ringed seal was chosen for further regional analysis. Only lipid weight blubber data from this species were included. PCB levels between male and female ringed seals did not appear to differ significantly, and followed similar trends when they did, so both genders and juveniles were included (see figure 4.3, top right). Likewise, data representing all numbers of congeners and Aroclor were included due to the difficulty in evaluating the significance of these parameters. Because data were still insufficient for certain regions, Svalbard, West Russia, Central Russia, and Northern Norway were combined into one region, as were East Russia and Bering/Aleutians, Beaufort/Chukchi and North Central Canada, Northern Quebec and South/Central Baffin, North East Canada and West Greenland, and Southeastern and Northwestern Hudson Bay. The data are also grouped into 5-year intervals to buffer against temporal variability on a small scale. The results of this analysis are shown in figure 5.2.

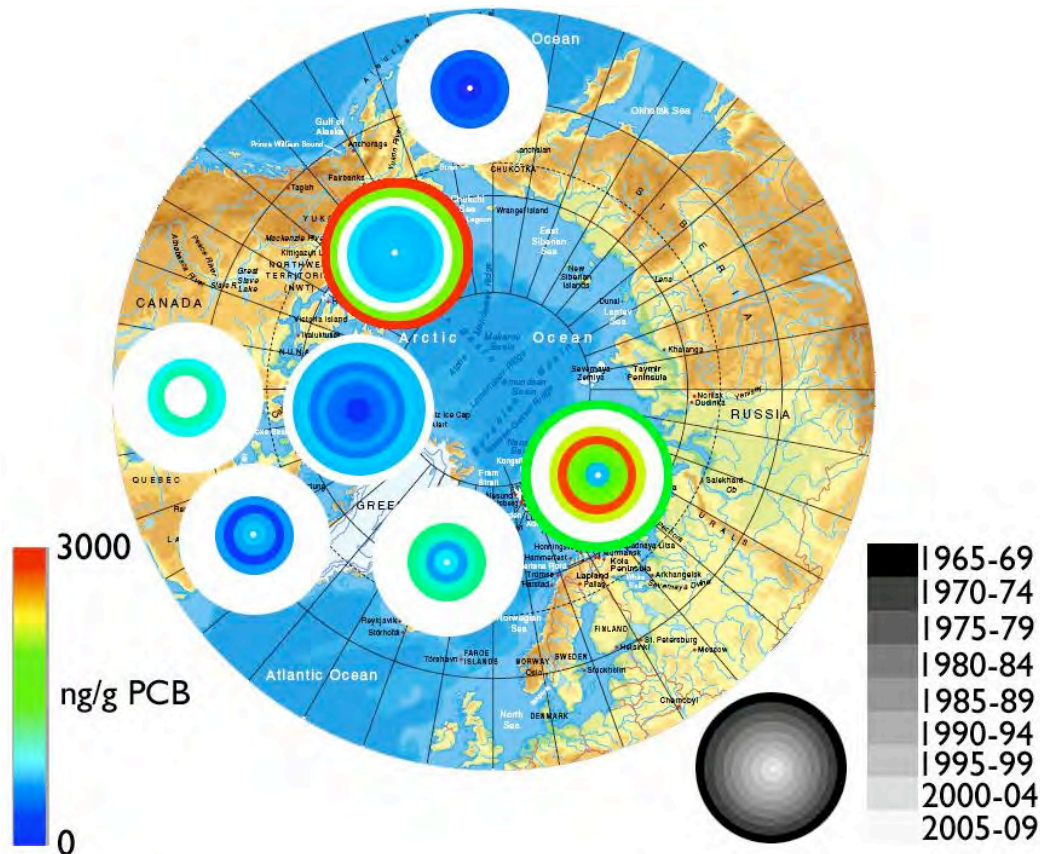


Figure 5.2. Historical trends of Σ PCB concentrations in ringed seal blubber, by region. Each concentric circle corresponds to a different time interval indicated the legend in the lower right. Circles that are white represent time intervals for which no data are available.

For four of the areas shown in Figure 5.2, PCB levels have steadily decreased since the mid 1980s through the present. However, levels in the areas from the southern Canadian Arctic and East Greenland have increased since the mid 1990s. One possible explanation could be that these areas are closer to sources of PCBs, and long-range transport to these areas thus takes a shorter amount of time. If this were the case, the trends from these areas could possibly reflect more real-time changes in PCB production and emission.

5.9 Temporal trends and comparison to global production data

Figure 5.3 below compares the historical patterns of PCB levels in various mammal groups to each other to assess the possibility of common temporal trends. The graphs are calibrated on a linear, rather than a log scale. To control for absolute differences in PCB levels between different mammal groups but reveal similar relative temporal differences between these groups, each graph is based on a different numerical y-axis scale. The data were chosen to eliminate biases introduced from various confounding parameters (e.g. only odontocete cetaceans are included to avoid the problems associated with the mysticete data as discussed in section 5.2). While the figure overall appears to be quite chaotic, certain similarities are seen between the historical trends of different mammal groups. For example, pinnipeds, odontocetes, humans, and arctic foxes all exhibit a dip in Σ PCB concentrations

during the early to mid 1980s, while polar bears show a similar dip in the late 1980s. Likewise, odontocetes, arctic foxes, and pinnipeds all show a peak in the early 1970s, caribou and humans both show a similar dip in levels in the mid to late 1990s, and the temporal pattern between pinnipeds and odontocetes appear to be quite similar from the early 1990s onward. That at least some correlation seems to exist between certain groups during certain times could provide justification for a general temporal trend in PCB levels for the entire Arctic ecosystem.

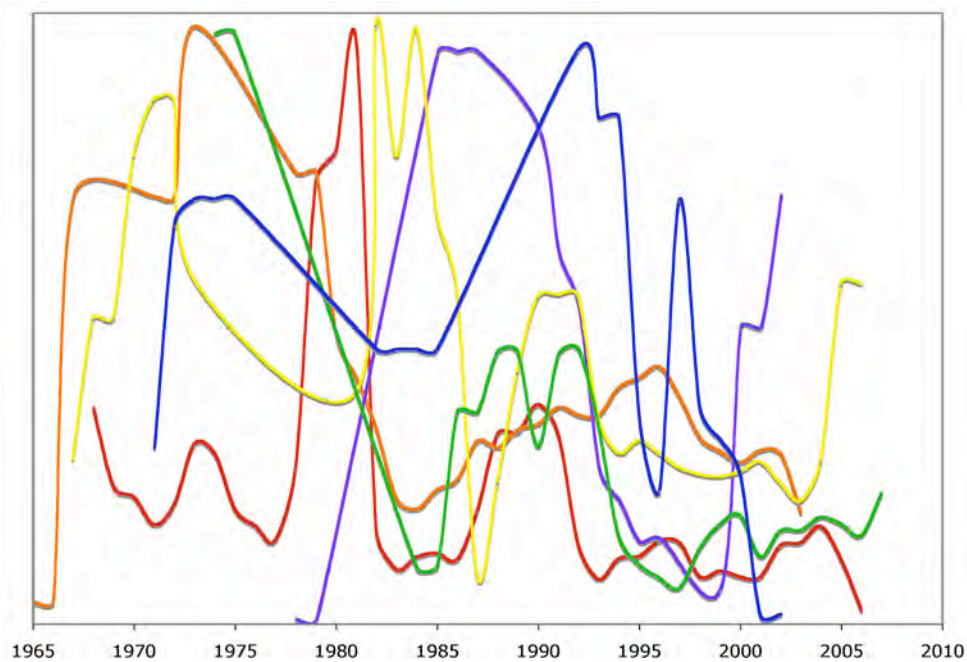


Figure 5.3. 3-year moving averages of Σ PCB concentrations (right y-axis) in pinnipeds and sea otters (red, all species), lipid weight data from all tissues plus wet weight data from blubber), cetaceans (orange, odontocete species only, lipid weight data from all tissues plus wet weight data from blubber), polar bears (yellow, lipid weight data from all tissues plus wet weight data from adipose), humans (green, lipid weight data from all tissues), arctic foxes (blue, all data), and caribou (purple, all data). Note that all y-axes are presented on a linear scale, though all numerical scales are different from each other.

To evaluate whether these trends are indicative of changes in the global production levels of PCBs, in figure 5.4 they are compared to the levels of Σ PCB (sum of 22 congeners) produced in North America and Europe reported from the “default scenario” model from (Breivik *et al.*, 2007). Interestingly, certain mammal groups, especially pinnipeds, show very similar trends to historical PCB production, except with a time lag. Particularly in pinnipeds, this time lag appears to be fairly consistent, and therefore is perhaps a useful estimation of the amount of time it takes PCBs to enter Arctic organisms from when they are produced. Figure 5.5 presents a summary of whether a correlation to production data is seen, and the length of the time lag, for different mammal groups. Interestingly, the termination of PCB production has clearly not corresponded to a cease in PCB contamination in Arctic mammals, an observation likely due to environmental release and re-emission, and the lengthy transport time to the Arctic.

With regard to legislation pertaining to PCBs, neither the 1979 American ban nor the 2001 Stockholm Convention seem to have significantly influenced the historical trends

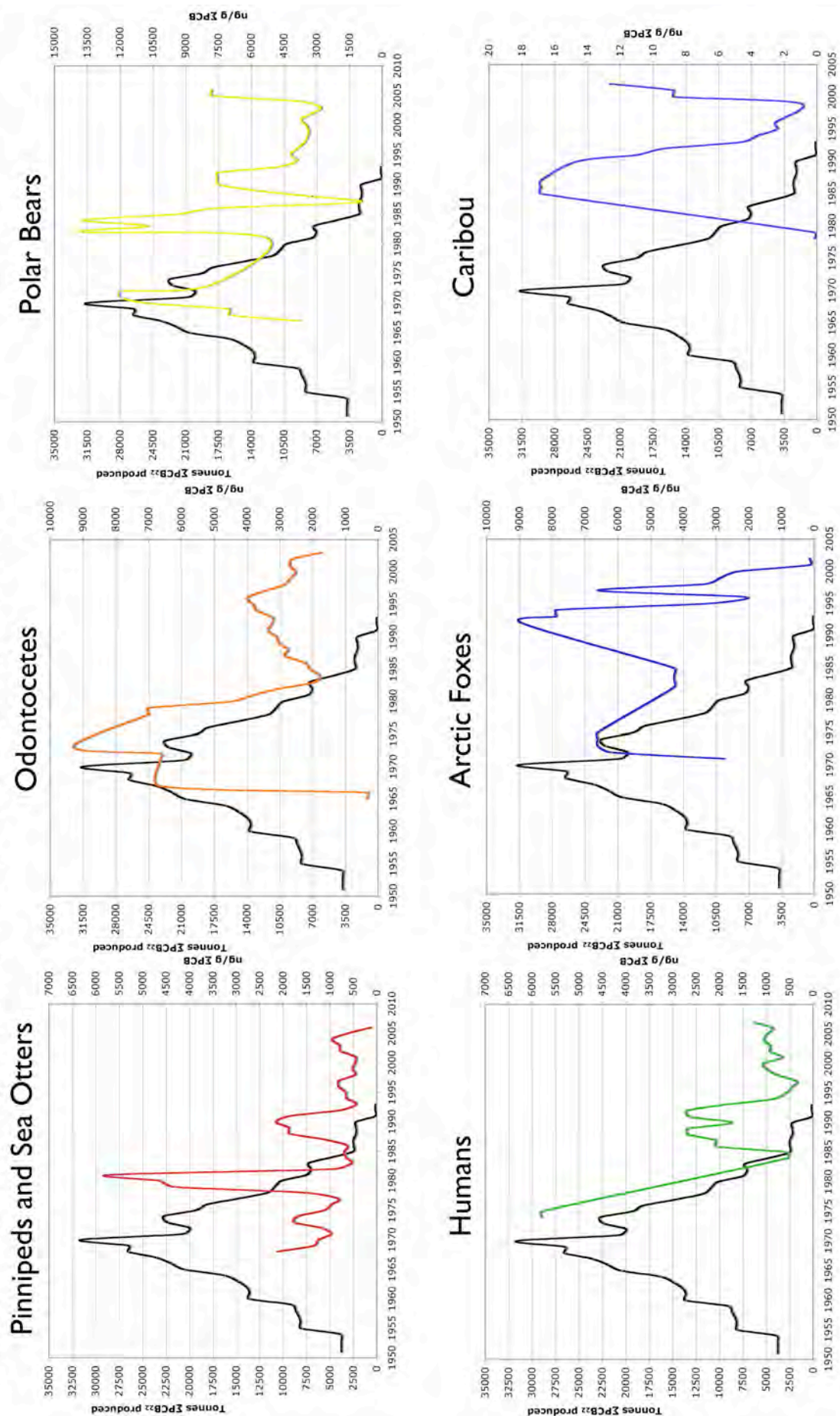
of PCBs either in production or in Arctic biota. Production has apparently been in decline since the early 1970s, well before the TSCA was passed in 1976 enacting the 1979 ban. However, growing public awareness of the toxic effects of PCBs to wildlife and humans, and of environmental issues at large, together with industrial and financial difficulties like the oil crisis and economic recession, may have initiated the decline at this time before legislation was passed.

Finally, when compared to the production curve, many of the historical trends from Arctic mammals appear to fluctuate in amplitude quite dramatically. Because many of these mammal species occupy relatively high trophic positions, and therefore accumulate a significant of PCBs through biomagnification, it is possible that they are sensitive to even small changes in environmental PCB levels that are compounded as they are transferred up the Arctic food web.

<u>Mammal Group</u>	<u>Correlation to Production?</u>	<u>Time Lag (approx.)</u>
Pinnipeds and Sea Otters	Possibly	10-15 years
Odontocetes	Unclear	–
Polar Bears	Unclear	–
Humans	Unclear	–
Arctic Foxes	Possibly	25 years
Caribou	Possibly	20-30 years

Figure 5.5. Summary of historical patterns seen in figure 5.4 (next page).

Figure 5.4. Comparison between modeled historical trend of Σ PCB production (black, left y-axis, sum of 22 congeners, “default scenario” from Breivik *et al.*, 2007)) and 3-year moving averages of Σ PCB concentrations (right y-axis) in pinnipeds and sea otters (red, all species, lipid weight data from all tissues plus wet weight data from blubber), cetaceans (orange, odontocete species only, lipid weight data from all tissues plus wet weight data from adipose), humans (green, lipid weight data from all tissues), polar bears (yellow, lipid weight data from all tissues), arctic foxes (blue, all data), and caribou (purple, all data). Note that all y-axes are presented on a linear scale.



6 Future Directions

While this study attempted to be as comprehensive as possible, a number of improvements could be made in the future to make it even more thorough and rigorous.

6.1 More species and physical data

Mammals comprise important components of the marine and especially terrestrial Arctic ecosystems, and are the most crucial species for indigenous human populations in the Arctic. However, species from other vertebrate and invertebrate groups are also critical and merit attention. Birds, for example, can shape Arctic ecosystems in profound ways, and a wealth of literature has been written about the levels and effects of PCB contamination in these species. Some birds occupy trophic positions at the top of the food web, such as the glaucous gull in the marine ecosystem and the snowy owl in the terrestrial ecosystem, and PCB levels in these species are expected to be on the order of some of the mammalian carnivores discussed in this study. Other groups that are lower in the food web, such as plants, plankton, marine invertebrates, and fish, serve as important links between the physical environment and animals in higher trophic levels, and would therefore be particularly prudent to study in the context of PCB contamination and biomagnification. Finally, while comparison between PCB levels in biota and global production levels potentially reveals significant correlations, a number of steps take place between the moment PCBs are produced and when they are found in Arctic species. Further assessments of historical trends of global PCB emission and consumption levels, as well as PCB levels in Arctic air, seawater, soil, sediment, ice, and/or snow, could prove a useful analysis to trace the path of PCBs as they enter the Arctic ecosystem.

6.2 Age

One parameter that was not included in this study but frequently reported on in the literature is the age of the animals that were sampled. For the purposes of this study, only a crude distinction was made between juveniles and adults. Through bioconcentration, individual organisms accrue greater levels of PCBs as they age. Therefore, the age of the animals studied could explain much of the variance seen both within and between datapoints. Furthermore, the age of animals compared to their PCB levels can be used as a proxy measure of when PCBs started entering the Arctic ecosystem.

6.3 Seasonality

Another parameter often reported in the literature but not accounted for in this review is the season in which samples were collected. Arctic animals undergo dramatic changes in body condition between different seasons due to changing availability and quality of food. For example, caribou can lose upwards of 30% of their total body mass in winter (Blix, 2005). Most of the mass lost is from fat reserves to provide energy, though muscle mass is also lost in leaner times. Given that blubber and adipose are major reservoirs for PCBs, the toxicity of PCBs may be more potent during fasting periods when these reserves are burned off and the PCBs they contained enter the bloodstream, where they gain access

to vital organs like the brain. Polischuk *et al.* (2002) demonstrated that while body burden (the total weight) of PCBs in polar bears did not change significantly between summer and autumn, the concentration of PCBs in the entire body and individual tissues did increase, owing to a decrease in body mass. Seasonal changes in PCB levels in Arctic animals have implications for the physiological toxicity of these compounds, and are yet another possible confounding variable when trying to compile and analyze historical PCB data.

6.4 Statistical rigor

Relying on subjective interpretations of graphically or visually presented data is a crude way at best to assess historical trends and the effect of various parameters on the variability seen in the data. Many of the studies included in this review employed rigorous statistical assessments, such as analysis of variance (ANOVA) and principal components analysis (PCA), to determine how much influence different parameters had on the variance seen in their own data. Other Williams College students studying historical environmental data have used techniques such as seasonal decomposition, generalized additive models, and the Mann-Kendall test for trend to build and evaluate the significance of temporal trends (Edwards, 1996). Due to a lack of time and experience with these techniques, they were not utilized in this review. Furthermore, the choice of a 3-year moving average was arbitrary and may not appropriately account for statistical seasonality. Together with the absence of standard deviations, confidence intervals, and ranges presented in the graphs, this lack of statistical rigor may weaken the associations or trends seen in this study.

7 Conclusion

Though their production and much of their use has ceased, PCBs remain a present and potent threat to Arctic wildlife, due to environmental release and re-emission, and delayed transport to the Arctic. While the existence of many confounding parameters related to the selection, analysis, and reporting of biological samples and the lack of statistical analysis precludes any firm conclusions from being made, it does appear that PCB levels overall have declined in many mammal species, at least within the past few decades. Furthermore, some species, like pinnipeds and the arctic fox, appear to be useful “recorders” of historical trends in PCB production and emission.

Other persistent organic pollutants (POPs) like polybrominated diphenyl ethers (PBDEs) and other brominated flame retardants (BFRs) are expected to soon overtake PCBs as the major contaminants of Arctic wildlife. Additionally, global climate change may induce a complex set of changes in the distribution, levels, and toxicity of POPs found in Arctic wildlife (MacDonald *et al.*, 2005). The historical record of PCBs established here could serve scientists studying these future phenomena as a useful model of the responses Arctic wildlife to various contaminants throughout their historical production cycle, and as a basis of comparison of toxicological processes of PCBs in the Arctic before and after future effects of climate change.

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