

## CHEMICAL CONTAMINANTS IN THE ARCTIC ENVIRONMENT – ARE THEY A CONCERN FOR WILDLIFE?

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ABSTRACT.—Environmental contaminants are a global problem, and their presence in the Arctic reflects the way in which the Arctic interacts with the rest of the world. Most contaminants are transported to the North on air and ocean currents from more southerly agricultural and industrial sources. Upon reaching the Arctic environment, many persistent contaminants bioaccumulate and biomagnify in the food web, making those species feeding at high trophic positions more vulnerable to contaminant exposure via their diet. By examining contaminant levels in wildlife, we can look for the arrival of new contaminants in the Arctic, as well as determine whether existing chemical contaminants of concern are increasing or decreasing. Historically, contaminants of concern included compounds such as the polychlorinated biphenyls (PCBs), and organochlorine pesticides such as dichlorodiphenyltrichloroethane (DDT). During the 1950s to 1970s, bioaccumulation of organochlorine compounds such as DDT and its degradation product, dichlorodiphenyl-dichloroethane (DDE), were associated with eggshell thinning and reduced reproduction rates in top predatory species such as the Peregrine Falcon (*Falco peregrinus*). The majority of these legacy persistent organic pollutants (POPs) have significantly declined in Arctic biota over the last several decades. However, more recently, newer compounds such as brominated flame retardants (BFRs) and perfluorinated compounds (PFCs) have been detected in a wide variety of biota including Arctic wildlife. Certain metals are also contaminating the Arctic environment. Elemental mercury ( $\text{Hg}^0$ ) is highly volatile, and gaseous Hg partitions readily into the atmosphere where it can undergo long-range atmospheric transport to the polar regions which are global sinks for Hg. Although Hg occurs naturally in the environment, anthropogenic sources have been postulated to contribute more significantly to the occurrence of Hg in the Arctic than natural emissions, resulting in increasing Hg concentrations in a variety of Arctic biota, particularly in the Canadian Arctic and western Greenland. Recent warming ocean conditions and longer ice-free periods have also altered prey availability in some areas of the Arctic, affecting nutrition and chemical contaminant profiles. These changes in environmental conditions and contaminant exposure all contribute to the complexity of interpreting the contaminant profiles found in Arctic biota. *Received 1 March 2011, accepted 19 April 2011.*

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BIOTA AT NORTHERN LATITUDES are exposed to a variety of chemical residues that are, for the most part, transported there by the prevailing winds, ocean currents and rivers (AMAP 2009). Unlike metals, such as mercury, persistent organic pollutants are generated entirely from anthropogenic sources (AMAP 2004). In addition to the legacy organochlorine contaminants, such as the PCBs and DDT, other classes of chemical contaminants have been found more recently in the Arctic environment. These include the brominated flame retardants and perfluorinated compounds.

*Legacy Persistent Organic Pollutants.*—Persistent organic pollutants (POPs), which have been banned or restricted in use, are generally referred to as legacy POPs. These include such compounds as the polychlorinated biphenyls (PCBs), and organochlorine pesticides such as dichlorodiphenyltrichloroethane (DDT), its break-down product dichlorodiphenyldichloroethane (DDE), chlordane, hexabromobenzene (HCB), hexachlorocyclohexane (HCH), and mirex. In response to regulatory actions to reduce emissions (e.g. Stockholm Convention on POPs, UN ECE Convention on Long-range Transboundary Air Pollution (LRTAP) POPs Protocol), most of the legacy POPs have declined in the Arctic environment as reflected in time trends for air (Hung et al. 2010) and biota (Rigét et al. 2010). However, because of their chemical characteristics, POPs will remain in the environment for many decades to come.

*Brominated Flame Retardants.*—Brominated flame retardants (BFRs) are chemicals used in materials to make them more fire-resistant. Examples include polyurethane foam, plastics used in electronic equipment, circuit boards, expanded and extruded plastic (e.g. Styrofoam), and textile coatings (de Wit et al. 2010). Most BFRs are additives mixed into polymers and are not chemically bound to the materials, while others react chemically with the material. Those BFRs which are additives may separate/leach from surface of product into

environment, while others enter the environment through incomplete chemical reactions and destruction of the material. BFRs have many chemical characteristics which make them behave in ways that are similar to POPs. Brominated organic compounds, such as polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDs) and polybrominated biphenyls (PBBs), have been detected in a wide variety of biota including Arctic wildlife (de Wit et al. 2006). The United States is currently the only Arctic country producing BFRs (de Wit et al. 2010). Of the three technical PBDE products (Penta-BDE, Octa-BDE, Deca-BDE), Penta- and Octa-BDE were banned in the European Union and in Norway in 2004, and the sole manufacturer of these two products in the United States voluntarily discontinued production in 2005 (de Wit et al. 2010). Production of PBBs has also ceased. As of 2008, however, there were no restrictions on the production and use of technical HBCD.

*Perfluorinated Compounds.*—Poly- and perfluorinated organic compounds (PFCs) are ubiquitous in the Arctic environment. Although PFCs were commercialized over 40 years ago, they received little attention until they were reported as contaminants in wildlife in the Arctic and elsewhere by Giesy and Kannan (2001). Since then, PFCs have been shown to be widespread in the environment and in wildlife (Houde et al. 2006, Lau et al. 2007, Butt et al. 2010). The chemical and thermal stability of PFCs has led to the manufacture of a range of fluoropolymers used as lubricants, adhesives, stain and soil repellants, paper coatings, non-stick surfaces on cookware, and fire-fighting foams (Kissa 2001). The major fluorinated compounds that have been measured in the environment are the perfluorinated sulfonates (PFSA), of which perfluorooctane sulfonate (PFOS) is best known, and the perfluorinated carboxylates (PFCAs) which include perfluorooctanoate (PFOA) (Butt et al. 2010). Regulation of PFCs is being discussed at both international and national levels, and PFOS has recently been nominated as a candi-

date POP for the UN ECE LRTAP POPs Protocol (AMAP 2009) and, in 2009, was listed as a POP to be regulated for restricted use under the Stockholm Convention (Stockholm Convention on POPs 2010).

*Mercury.*—Although mercury (Hg) occurs naturally in the environment, emissions from human activities, such as fossil fuel combustion, non-ferrous metal production, and waste incineration, have been postulated to contribute more significantly to the occurrence of Hg in the Arctic than natural emissions (Pacyna 2005). The quantities of mercury released from human activities have been increasing over the past 200 years, i.e. since the Industrial Revolution (Nriagu and Pacyna 1988, Pacyna et al. 2006). This has resulted in significant increases in biotic Hg levels over that time period, even in regions that are remote from most anthropogenic sources, such as the Arctic (Dietz et al. 2006, 2009). Although there is not yet a global policy to reduce emissions of mercury, many countries are already taking steps to lower their emissions. In 2003, a Protocol on Heavy Metals was adopted by the UN ECE LRTAP Convention aimed at limiting emissions of mercury and other metals from Europe and North America.

#### HOW DO CHEMICAL CONTAMINANTS REACH THE ARCTIC?

Winds, ocean currents and rivers are all potential transport pathways for chemical contaminants to reach the Arctic (AMAP 2009). Chemical characteristics, such as volatility and water solubility, determine the potential of an organic chemical to become an Arctic contaminant (Wania 2003, 2006). Air is the most important transport route to the Arctic for volatile and semi-volatile contaminants including Hg (AMAP 2002). Many chlorinated organics are present as gases even at low temperatures and are absorbed from the gas phase by water, snow, soil and plant surfaces (Braune et al. 2005a). More recently, however, studies

have found that many non-volatile but highly stable compounds such as the highly brominated compounds as well as the PFCAs and PFSA are present in the Arctic (Muir and de Wit 2010). Their presence may be due to atmospheric transport on particles, or to degradation of volatile precursors. For the more water-soluble, less volatile contaminants, transport via ocean currents may be more important than the atmospheric route (Li et al. 2002, 2004). Unlike the atmospheric transport pathway, which can deliver contaminants from mid-latitudes to the Arctic within a few days or weeks, ocean transport is relatively slow and it may take decades before contaminants reach the Arctic (AMAP 2002). Waterborne contaminants may also enter Arctic marine ecosystems via northward-flowing rivers draining into the Arctic Ocean (Braune et al. 2005a).

Unlike other POPs, the two main classes of PFCs found in the environment are not volatile and their transport pathways to the Arctic are complex. Two major potential pathways have been proposed. One pathway focuses on atmospheric transport of particles, or degradation of volatile precursors to PFCAs in the atmosphere, and the second pathway involves the transport of directly-emitted PFCAs and PFSA via ocean currents to the Arctic marine environment (Armitage et al. 2006, Wania 2007).

Metals are naturally-occurring elements but are also generated through human activities. Elemental Hg ( $\text{Hg}^0$ ) is highly volatile, and gaseous  $\text{Hg}^0$  partitions readily into the atmosphere where it can undergo long-range atmospheric transport. Due to a complex set of factors (Macdonald et al. 2005), polar regions have become global sinks for Hg.

Relatively low levels of contaminants are found in the terrestrial environment compared with the marine environment, especially in the high Arctic, due to snow cover in the winter and subsequent transfer of deposited contaminants to freshwater systems during the rapid

spring snow melt (Semkin 1996). The Canadian Arctic terrestrial environment, for example, has few significant contaminants issues that can be attributed to long-range transport and deposition of POPs and heavy metals, particularly when compared with the freshwater and marine environments (Gamberg et al. 2005). The major contaminant concerns for terrestrial animals are from non-essential elements such as cadmium (Cd) and Hg (Gamberg et al. 2005).

#### CONTAMINANTS AND WILDLIFE

Persistent chlorinated and brominated organocontaminants, as well as Hg, bioaccumulate in wildlife (Braune et al. 2005a). For example, concentrations of PCBs and Hg increase with age in Gyrfalcons (*Falco rusticolus*) (Ólafsdóttir et al. 1995, Dietz et al. 2006). These compounds also biomagnify up the food chain (Braune et al. 2005a) making those species feeding at high trophic positions more vulnerable to contaminant exposure via their diet (Hop et al. 2002, Borgå et al. 2004, Vorkamp et al. 2004, Wolkers et al. 2004). Food web studies in marine ecosystems suggest that PFCs can also biomagnify but there have been few studies to date, and there have been no studies carried out for terrestrial or freshwater food webs (Butt et al. 2010).

Seabirds feed at relatively high trophic positions in Arctic marine food webs (Hobson et al. 2002, Hop et al. 2002) and, therefore, seabirds, and in particular their eggs, have been used to monitor contamination of the marine environment in the Canadian Arctic since 1975 (Braune 2007). At the time of egg formation, the lipophilic halogenated compounds are transferred along with fat to the eggs thus reflecting the contaminant burden in the female at the time of laying (Verreault et al. 2006). Unlike other halogenated organic contaminants (e.g., PCBs, PBDEs), PFCs appear to bind to proteins rather than partition into lipid (Butt et al. 2010) but are found in eggs, as well (Houde et al. 2006, Verreault et al.

2007). Mercury is also readily transferred to the eggs (Wiener et al. 2003). Contaminant burdens in the egg reflect residues assimilated over a long time period by the female and, particularly in migratory species, may integrate exposure from a number of different locations (Hebert 1998, Monteiro et al. 1999).

Contaminant concentrations can vary considerably among species depending on their feeding and migration strategies (Braune et al. 2002, Buckman et al. 2004). For example, in Greenland, Peregrine Falcons (*Falco peregrinus*) had higher levels of DDE, PCBs and other organochlorine contaminants in their plasma compared with Gyrfalcons (Jarman et al. 1994). The authors attributed the difference in contaminant levels to the fact that Peregrines, which themselves are migratory, consumed contaminated, migratory prey, both on their wintering and breeding grounds, whereas the non-migratory Gyrfalcons consumed low trophic-level, non-migratory prey (e.g. ptarmigan, hare). Interpretation of contaminant concentrations in biota may also be confounded if populations vary their diet over trophic levels through time (e.g. see Hebert et al. 1997, 2000).

*Spatial Patterns.*—Compared with other circumpolar countries, concentrations of many organochlorine contaminants in Canadian Arctic biota are generally lower than in the European Arctic and eastern Greenland but are higher than in Alaska, whereas Hg concentrations are substantially higher in Canada and western Greenland than elsewhere (Braune et al. 2005a). Spatial patterns of PBDEs in seabirds and marine mammals are similar to those seen for PCBs, with the highest concentrations found in organisms from East Greenland and Svalbard in the European Arctic, lower concentrations in the Canadian Arctic (except Hudson Bay), and the lowest concentrations in Alaska (de Wit et al. 2010). For example, PBDE concentrations in eggs of alcids and/or Northern Fulmars (*Fulmarus glacialis*) are generally lower in the Bering

Sea, intermediate in Canada, western Greenland, Bjørnøya in the Barents Sea, northern Norway and the Faroe Islands, and highest on Iceland, east Greenland and Svalbard in the European high Arctic (de Wit et al. 2010). A similar pattern is seen for gulls (de Wit et al. 2010).

Little is known about the BFR contamination of the terrestrial ecosystem in the Arctic; however, concentrations of terrestrial animals at lower trophic levels were found to be low (de Wit et al. 2006, 2010), whereas they were much higher in terrestrial birds of prey, particularly in Peregrine Falcons (de Wit et al. 2006). For example, high BFR concentrations have been found in eggs of predatory birds feeding on terrestrial mammals and birds, particularly Peregrine Falcons in northern Sweden (Lindberg et al. 2004) and Norway (Herzke et al. 2001), and more recently at several sites in South Greenland (Vorkamp et al. 2005).

Although the marine ecosystem has been well-studied, there is a general lack of information at the present time to define any circumpolar trends for PFCs (Butt et al. 2010). There are few reports of PFCs in terrestrial wildlife but PFC levels have been measured in Common Loons (*Gavia immer*), Mink (*Mustela vison*), Arctic Fox (*Alopex lagopus*) and Caribou (*Rangifer tarandus*) from the Canadian Arctic (Martin et al. 2004, Tittlemier et al. 2005). In general, PFOS and perfluorooctane sulfonamide (PFOSA), as well as some of the PFCAs, were commonly detected (Butt et al. 2010).

Large-scale spatial patterns of element concentrations in tissues of terrestrial and freshwater animals may reflect both geochemical environments as well as anthropogenic inputs, as is the case for Cd in Canadian Caribou, for example, which have higher Cd concentrations in some Yukon herds where Cd is naturally higher in the soils (Braune et al. 1999). A circumpolar survey of Cd in Willow Ptarmigan (*Lagopus lagopus*) also showed considerable variation in

Cd levels among regions but the median concentration for Canada was much higher than in other countries, likely due to natural Cd exposure, possibly through a high intake of willow (*Salix sp.*) which is known to contain higher amounts of Cd than other food plants (Pedersen et al. 2006). In the marine environment, the highest Cd levels in seabirds were observed in northeastern Siberia and the lowest, in the Barents Sea, with intermediate levels in Arctic Canada and Greenland (Savinov et al. 2003). For Hg, concentrations in seabirds from the Barents Sea were also lower than in Greenland, Canada, and northeast Siberia (Savinov et al. 2003). Although there are some spatial differences in Hg in the freshwater and terrestrial environments, most levels are low (AMAP 2002). It should be noted that spatial differences in contaminant levels may also be affected by differences in available food items.

*Temporal Trends.*—The majority of legacy POPs (e.g., PCBs, DDT) have significantly declined in Arctic biota over the last several decades (Rigét et al. 2010) whereas Hg has increased in a number of Arctic species (Rigét et al. 2011). The declines in the legacy POPs are a consequence of past national and regional bans and restrictions on uses and emissions in circumpolar and neighbouring countries which began in the 1970s for chlorinated pesticides and PCBs (Muir and de Wit 2010). Seabird eggs from the Canadian high Arctic reflect the documented decline in legacy POPs (Braune 2007, Braune et al. 2007) as do eggs of Alaskan Peregrine Falcons (Ambrose et al. 2000). However, concentrations of Hg have been increasing in seabird eggs from the Canadian high Arctic (Braune 2007, Braune et al. 2006). In fact, Braune et al. (2006) found that Hg levels in eggs of the Ivory Gull (*Pagophila eburnea*) from the Canadian high Arctic, a species recently listed as “endangered” in Canada (COSEWIC 2010), were among the highest ever reported for seabird eggs from the Arctic marine environment. The pattern of increasing Hg concentrations in Canadian Arctic seabirds in recent decades supports the

west-to-east circumpolar gradient in the occurrence of recently increasing Hg trends reflected in Arctic biota. A recent analysis of Hg time series for Arctic biota showed that there is a higher proportion of temporal trend datasets, particularly for marine biota, in the Canadian and Greenland region of the Arctic showing significant Hg increases than in the North Atlantic Arctic (Rigét et al. 2011). The reasons for this are complex but likely involve anthropogenic and natural emissions coupled with environmental and biological (e.g. food-web) processes which may also be affected by climate change. In contrast, Hg in terrestrial biota and freshwater biota, for the most part, showed either no change over time or a decreasing trend (Rigét et al. 2011). Likewise, Cd levels in biota and in the abiotic environment are, for the most part, either stable or declining (Braune et al. 2005b).

Temporal trend studies for BFRs in Arctic wildlife have generated differing results with some PBDEs showing increasing concentrations and others showing a tendency to level off or decline in response to reductions in use and emissions (de Wit et al. 2010). However, no uniform pattern in temporal trends emerges for the Arctic. Concentrations of PBDEs increased from 1986 to 2003 in Peregrine Falcon eggs from South Greenland (Vorkamp et al. 2005). In the Canadian Arctic, PBDEs were first detected in seabird liver and eggs by Braune and Simon (2004). Subsequent retrospective analyses and continued monitoring showed that concentrations of total PBDE ( $\Sigma$ PBDE) concentrations in eggs of Thick-billed Murres (*Uria lomvia*) and Northern Fulmars from the Canadian high Arctic steadily increased between 1975 and 2003 after which time, levels appear to have started to decline (Braune 2008). An increase in  $\Sigma$ PBDE concentrations between 1976 and 2004 is also reflected in eggs of the Ivory Gull in the Canadian high Arctic (Braune et al. 2007). North America accounted for most of the global demand for the commercial Penta-BDE product used in polyurethane foam (Hale et al.

2003), and the declining  $\Sigma$ PBDE concentrations observed in the seabird eggs after 2003 may reflect the phasing out of Penta-BDE product usage in North America after 2005 (de Wit et al. 2010).

As with the BFRs, there are inconsistencies in the PFC temporal trends, most of which are from the North American Arctic and Greenland. Martin et al. (2004) screened livers of Northern Fulmars and Black Guillemots (*Cephus grylle*) collected from the Canadian high Arctic in 1993 for the presence of PFCs and found relatively low concentrations of both PFOS and the PFCAs in both species. Subsequent retrospective analyses showed that overall PFC concentrations in livers of Thick-billed Murres and Northern Fulmars had increased significantly from 1975 to 2003–2004 (Butt et al. 2007). A recent review by Butt et al. (2010) generally showed increasing trends from the 1970s in Arctic wildlife, although some studies from the Canadian Arctic showed recent declines in PFOS levels. Temporal trends of PFCs in eggs of Swedish Peregrine Falcons also showed increasing concentrations of PFCAs whereas concentrations of PFOS levelled off after the mid-1980s (Holmström et al. 2010). In contrast, Ringed Seals (*Phoca hispida*) and Polar Bears (*Ursus maritimus*) from Greenland continue to show increasing PFOS concentrations. The inconsistency in the PFC temporal trends between regions may represent differences in emissions from source regions (Butt et al. 2010).

*Effects.*—Given the complex and dynamic environment in which animals live, it is difficult to establish a cause-effect relationship for any one stressor. A recent assessment on the effects of POPs on Arctic wildlife and fish concluded that there still remains minimal evidence that POPs are having widespread effects on the health of Arctic organisms, with the possible exception of Polar Bears in East Greenland and Svalbard, as well as Svalbard Glaucous Gulls (*Larus hyperboreus*) where

studies have demonstrated sub-lethal physiological effects (Letcher et al. 2010).

Effect studies on wild terrestrial birds in the Arctic have been limited to Peregrine Falcons, although laboratory studies have been carried out using the American Kestrel (*Falco sparverius*). Population declines of Peregrine Falcons at a number of North American locations during the mid-part of the last century were associated with DDE-induced eggshell thinning (Peakall et al. 1990). With the significant decrease in most of the legacy POPs including *p,p'*-DDE, there has been an increase in eggshell thickness in Alaskan Peregrines and no apparent exceedance of toxicity thresholds for the legacy POPs for eggs sampled between 1988 to 1995 (Ambrose et al. 2000). Peregrine Falcon eggs sampled from South Greenland between 1986 and 2003 also showed a decreasing trend for the legacy POPs (Vorkamp et al. 2009), and Falk et al. (2006) demonstrated a significant long-term decrease in eggshell thinning in Greenlandic Peregrines. However, Johnstone et al. (1996) found no improvement in eggshell thickness in eggs sampled in the Canadian tundra during 1982–1986 and 1991–1994, although *p,p'*-DDE residue levels did decline. A recent study on Peregrines from Sweden showed that the average brood size decreased with increasing  $\Sigma$ PBDE concentrations, suggesting that PBDEs could influence reproduction in this species (Johansson et al. 2009). However, Nordlöf et al. (2010) found no correlation between levels of BFRs and reproductive success in White-tailed Eagles (*Haliaeetus albicilla*) in four regions of Sweden. Letcher et al. (2010) concluded that there is currently a low risk from  $\Sigma$ PBDE and HBCD exposure in the eggs of Arctic birds with respect to the reproductive and developmental effects reported for captive Kestrels by Fernie et al. (2009). The effects of PFCs on wildlife are not well known, in particular for Arctic biota (Butt et al. 2010). However, a recent study showed a relationship between blood plasma clinical-chemical parameters and organohalogen compounds

including PFCs in chicks of three raptor species in northern Norway which suggested potential effects on liver, kidney, bone, endocrinology and metabolism (Sonne et al. 2010).

Many high trophic level predators associated with marine and other aquatic ecosystems are exposed to Hg primarily as methylmercury (MeHg) in their diet, and in some areas of the Arctic, Hg concentrations in marine food webs have significantly increased in recent decades (Rigét et al. 2011) suggesting that levels in some marine mammals, birds, and fish may reach the point where adverse biological effects might be expected (AMAP 2002). However, compared to the amount of contaminant data available for the Arctic region, knowledge of the effects of these contaminant loads are very limited and, therefore, most evaluations of potential toxic effects of contaminants in wildlife are based on comparisons of tissue residue levels with toxicity thresholds derived from laboratory-based studies reported in the literature. Exceedance of these toxicity thresholds, however, does not necessarily result in adverse biological effects or population-level impacts. For example, Hg levels in 30% of the eggs of Peregrine Falcons sampled between 1991 and 1995 in Alaska exceeded the critical threshold for reproductive effects (Ambrose et al. 2000) although no causal association with Hg was established. Likewise, Cd in some ptarmigan, Caribou and Moose (*Alces alces*) in the Yukon Territory, which has high levels of naturally-occurring Cd, are high enough to raise concern for kidney damage, but effects have not been documented (AMAP 2002). Since Hg concentrations are still increasing in some Arctic animals (Rigét et al. 2011), Arctic species (particularly top predators) are likely to be exposed to increasing concentrations of Hg for some time to come.

#### EFFECTS OF CLIMATE CHANGE

Many of the transfer, transformation and storage processes driving global contaminant

cycles are likely to be significantly affected by the impacts of climate change, especially as changes in river runoff and precipitation patterns occur, ice and snow cover reduces, and permafrost thaws in the Arctic (Macdonald et al. 2005). Not only could changes in seasonal ice cover and temperature result in more re-emissions of contaminants to the air, but ecological changes, such as diet shifts and nutritional changes, disease, and species invasion may also be mediated by climate change which could alter patterns of contaminant exposure in biota (Muir and de Wit 2010). For example, there is evidence that earlier ice break-up over the last 20 years in western Hudson Bay has caused a temporal shift in the diet of Polar Bears resulting in increased concentrations of PBDEs and legacy POPs in the bears (McKinney et al. 2009). Gaston et al. (2003) have also associated the general warming of Hudson Bay waters with a shift from Arctic Cod (*Boreogadus saida*) and benthic fish species to Capelin (*Mallotus villosus*) and Sandlance (*Ammodytes hexaptera*) in the diet of Thick-billed Murres in northern Hudson Bay which could also affect the exposure of those birds to contaminants. Similarly, Gaden et al. (2009) have postulated that the length of the ice-free season affects the prey composition available to Ringed Seals in the western Canadian Arctic, indirectly influencing Hg uptake in the seals. Carrie et al. (2010) have associated increasing concentrations of PCBs and, in particular, Hg in Mackenzie River Burbot (*Lota lota*) with increasing algal primary productivity driven by warming temperatures and reduced ice cover. They postulate that the increased productivity increased the scavenging rate of Hg from the water column, potentially enhancing MeHg production, and increasing contaminant bioavailability to the fish. Clearly, there is already evidence that climate change is affecting contaminant exposure in biota, but it will be some time before the complexities of these interactions are understood.

#### LITERATURE CITED

- AMAP. 2002. Arctic Pollution 2002. Arctic Monitoring and Assessment Programme, Oslo, Norway.
- AMAP. 2004. AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.
- AMAP. 2009. Arctic Pollution 2009. Arctic Monitoring and Assessment Programme, Oslo, Norway.
- AMBROSE, R. E., A. MATZ, T. SWEM, AND P. BENTE. 2000. Environmental contaminants in American and Arctic Peregrine Falcon eggs in Alaska, 1979–95. Technical Report NAES-TR-00-02, US Fish and Wildlife Service, Fairbanks, Alaska.
- ARMITAGE, J., I. T. COUSINS, R. C. BUCK, K. PREVEDOUROS, M. H. RUSSELL, M. MACLEOD, AND S. H. KORZENIOWSKI. 2006. Modelling global-scale fate and transport of perfluorooctanoate emitted from direct sources. *Environmental Science and Technology* 40:6969–6975.
- BORGÅ, K., A. T. FISK, P. F. HOEKSTRA, AND D. C. G. MUIR. 2004. Biological and chemical factors of importance in the bioaccumulation and trophic transfer of persistent organochlorine contaminants in Arctic marine food webs. *Environmental Toxicology and Chemistry* 23:2367–2385.
- BRAUNE, B. M. 2007. Temporal trends of organochlorines and mercury in seabird eggs from the Canadian Arctic, 1975 to 2003. *Environmental Pollution* 148:599–613.
- BRAUNE, B. 2008. Temporal trends of contaminants in Arctic seabird eggs: inter-year variability. Pages 76–82 in *Synopsis of Research Conducted Under the 2007–2008 Northern Contaminants Program*. Indian and Northern Affairs Canada, Ottawa, Ontario, Canada.
- BRAUNE, B. M., G. M. DONALDSON, AND K. A. HOBSON. 2002. Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable iso-

- topes for intercolony differences. *Environmental Pollution* 117:133–145.
- BRAUNE, B. M., M. L. MALLORY, AND H. G. GILCHRIST. 2006. Elevated mercury levels in a declining population of Ivory Gulls in the Canadian Arctic. *Marine Pollution Bulletin* 52:969–987.
- BRAUNE, B. M., M. L. MALLORY, H. G. GILCHRIST, R. J. LETCHER, AND K. G. DROUILLARD. 2007. Levels and trends of organochlorines and brominated flame retardants in Ivory Gull eggs from the Canadian Arctic, 1976 to 2004. *Science of the Total Environment* 378:403–417.
- BRAUNE, B., D. MUIR, B. DEMARCH, M. GAMBERG, K. POOLE, R. CURRIE, M. DODD, W. DUSCHENKO, J. EAMER, B. ELKIN, M. EVANS, S. GRUNDY, C. HEBERT, R. JOHNSTONE, K. KIDD, B. KOENIG, L. LOCKHART, H. MARSHALL, K. REIMER, J. SANDERSON, AND L. SHUTT. 1999. Spatial and temporal trends of contaminants in Canadian Arctic freshwater and terrestrial ecosystems: a review. *Science of the Total Environment* 230:145–207.
- BRAUNE, B. M., P. M. OUTRIDGE, A. T. FISK, D. C. G. MUIR, P. A. HELM, K. HOBBS, P. F. HOEKSTRA, Z. A. KUZYSK, M. KWAN, R. J. LETCHER, W. L. LOCKHART, R. J. NORSTROM, G. A. STERN, AND I. STIRLING. 2005a. Persistent organic pollutants and mercury in marine biota of the Canadian Arctic: An overview of spatial and temporal trends. *Science of the Total Environment* 351–352:4–56.
- BRAUNE, B., P. OUTRIDGE, S. WILSON, A. BIGNERT, AND F. RIGET. 2005b. Chapter 5. Temporal Trends. Pages 84–106 in *AMAP Assessment 2002: Heavy Metals in the Arctic*. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.
- BRAUNE, B. M., AND M. SIMON. 2004. Trace elements and halogenated organic compounds in Canadian Arctic seabirds. *Marine Pollution Bulletin* 48:986–992.
- BUCKMAN, A. H., R. J. NORSTROM, K. A. HOBSON, N. J. KARNOVSKY, J. DUFFE, AND A. T. FISK. 2004. Organochlorine contaminants in seven species of Arctic seabirds from northern Baffin Bay. *Environmental Pollution* 128:327–338.
- BUTT, C. M., U. BERGER, R. BOSSI, AND G. T. TOMY. 2010. Levels and trends of poly- and perfluorinated compounds in the arctic environment. *Science of the Total Environment* 408:2936–2965.
- BUTT, C. M., S. A. MABURY, D. C. G. MUIR, AND B. M. BRAUNE. 2007. Prevalence of long-chained perfluorinated carboxylates in seabirds from the Canadian Arctic between 1975 and 2004. *Environmental Science and Technology* 41:3521–3528.
- CARRIE, J., F. WANG, H. SANEI, R. MACDONALD, P. OUTRIDGE, AND G. STERN. 2010. Increasing contaminant burdens in an Arctic fish, Burbot (*Lota lota*), in a warming climate. *Environmental Science and Technology* 44:316–322.
- COSEWIC. 2010. Canadian Wildlife Species at Risk. Committee on the Status of Endangered Wildlife in Canada. [Online.] Available at [www.cosewic.gc.ca/eng/sct0/rpt/rpt\\_csar\\_e.cfm](http://www.cosewic.gc.ca/eng/sct0/rpt/rpt_csar_e.cfm)
- DE WIT, C. A., M. ALAEE, AND D. C. G. MUIR. 2006. Levels and trends of brominated flame retardants in the Arctic. *Chemosphere* 64:209–233.
- DE WIT, C. A., D. HERZKE, AND K. VORKAMP. 2010. Brominated flame retardants in the Arctic environment—trends and new candidates. *Science of the Total Environment* 408:2885–2918.
- DIETZ, R., P. M. OUTRIDGE, AND K. A. HOBSON. 2009. Anthropogenic contributions to mercury levels in present-day Arctic animals—a review. *Science of the Total Environment* 407:6120–6131.
- DIETZ, R., F. RIGÉT, D. BOERTMANN, C. SONNE, M. T. OLSEN, J. FJELDSÅ, K. FALK, M. KIRKEGAARD, C. EGEVANG, G. ASMUND, F. WILLE, AND S. MØLLER. 2006. Time trends of mercury in feathers of West Greenland birds of prey during 1851–2003. *Environmental Science and Technology* 40:5911–5916.

- FALK, K., S. MØLLER, AND W. G. MATTOX. 2006. A long-term increase in eggshell thickness of Greenlandic Peregrine Falcons *Falco peregrinus tundrius*. *Science of the Total Environment* 355:127–134.
- FERNIE, K. J., J. L. SHUTT, R. J. LETCHER, I. J. RITCHIE, AND D. M. BIRD. 2009. Environmentally relevant concentrations of DE-71 and HBCD alter eggshell thickness and reproductive success of American Kestrels. *Environmental Science and Technology* 43:2124–2130.
- GADEN, A., S. H. FERGUSON, L. HARWOOD, H. MELLING, AND G. A. STERN. 2009. Mercury trends in Ringed Seals (*Phoca hispida*) from the western Canadian Arctic since 1973: Associations with length of ice-free season. *Environmental Science and Technology* 43:3646–3651.
- GAMBERG, M., B. BRAUNE, E. DAVEY, B. ELKIN, P. HOEKSTRA, D. KENNEDY, C. MACDONALD, D. MUIR, A. NIRWAL, M. WAYLAND, AND B. ZEEB. 2005. Spatial and temporal trends of contaminants in terrestrial biota in the Canadian Arctic. *Science of the Total Environment* 351–352:148–164.
- GASTON, A. J., K. WOO, AND J. M. HIPFNER. 2003. Trends in forage fish populations in northern Hudson Bay since 1981, as determined from the diet of nestling Thick-billed Murres *Uria lomvia*. *Arctic* 56:227–233.
- GIESY, J. P., AND K. KANNAN. 2001. Distribution of perfluorooctane sulfonate in wildlife. *Environmental Science and Technology* 35:1339–1342.
- HALE, R. C., M. ALAEE, J. B. MANCHESTER-NEESVIG, H. M. STAPLETON, AND M. G. IKONOMOU. 2003. Polybrominated diphenyl ether flame retardants in the North American environment. *Environment International* 29:771–779.
- HEBERT, C. E. 1998. Winter severity affects migration and contaminant accumulation in northern Great Lakes Herring Gulls. *Ecological Applications* 8:669–679.
- HEBERT, C. E., K. A. HOBSON, AND J. L. SHUTT. 2000. Changes in food web structure affect rate of PCB decline in Herring Gull (*Larus argentatus*) eggs. *Environmental Science and Technology* 34:1609–1614.
- HEBERT, C. E., J. L. SHUTT, AND R. J. NORSTROM. 1997. Dietary changes cause temporal fluctuations in polychlorinated biphenyl levels in Herring Gull eggs from Lake Ontario. *Environmental Science and Technology* 31:1012–1017.
- HERZKE, D., R. KALLENBORN, T. NYGÅRD, AND T. SANDANGER. 2001. Species dependent distribution of polybrominated biphenyls and diphenylethers in eggs of Norwegian birds of prey. Pages 321–324 in *The Second International Workshop on Brominated Flame Retardants*, 14–16 May 2001. Stockholm, Sweden.
- HOBSON, K. A., A. FISK, N. KARNOVSKY, M. HOLST, J. M. GAGNON, AND M. FORTIER. 2002. A stable isotope ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ) model for the North Water food web: Implications for evaluating trophodynamics and the flow of energy and contaminants. *Deep-Sea Research II* 49:5131–5150.
- HOLMSTRÖM, K. E., A. K. JOHANSSON, A. BIGNERT, P. LINDBERG, AND U. BERGER. 2010. Temporal trends of perfluorinated surfactants in Swedish Peregrine Falcon eggs (*Falco peregrinus*), 1974–2007. *Environmental Science and Technology* 44:4083–4088.
- HOP, H., K. BORGÅ, G. W. GABRIELSON, L. KLEIVANE, AND J. U. SKAARE. 2002. Food web magnification of persistent organic pollutants in poikilotherms and homeotherms from the Barents Sea. *Environmental Science and Technology* 36:2589–2597.
- HOUDE, M., J. W. MARTIN, R. J. LETCHER, K. R. SOLOMON, AND D. C. G. MUIR. 2006. Biological monitoring of polyfluoroalkyl substances: A review. *Environmental Science and Technology* 40:3463–3473.
- HUNG, H., R. KALLENBORN, K. BREIVIK, Y. SU, E. BRORSTRÖM-LUNDÉN, K. OLAFSDOTTIR, J. M. THORLACIUS, S. LEPPÄNEN, R. BOSSI,

- H. SKOV, S. MANØ, G. W. PATTON, G. STERN, E. SVERKO, AND P. FELLIN. 2010. Atmospheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993–2006. *Science of the Total Environment* 408:2854–2873.
- JARMAN, W. M., S. A. BURNS, W. G. MATTOX, AND W. S. SEEGAR. 1994. Organochlorine compounds in the plasma of Peregrine Falcons and Gyrfalcons nesting in Greenland. *Arctic* 47:334–340.
- JOHANSSON, A-K., U. SELLSTRÖM, P. LINDBERG, A. BIGNERT, AND C. A. DE WIT. 2009. Polybrominated diphenyl ether congener patterns, hexabromocyclododecane, and brominated biphenyl 153 in eggs of Peregrine Falcons (*Falco peregrinus*) breeding in Sweden. *Environmental Toxicology and Chemistry* 28:9–17.
- JOHNSTONE, R. M., G. S. COURT, A. C. FESSER, D. M. BRADLEY, L. W. OLIPHANT, AND J. D. MACNEIL. 1996. Long-term trends and sources of organochlorine contamination in Canadian Tundra Peregrine Falcons, *Falco peregrinus tundrius*. *Environmental Pollution* 93:109–120.
- KISSA, E. 2001. *Fluorinated Surfactants and Repellants*, 2<sup>nd</sup> ed. Marcel Dekker, New York, New York, USA.
- LAU, C., K. ANITOLE, C. HODES, D. LAI, A. PFAHLES-HUTCHENS, AND J. SEED. 2007. Perfluoroalkyl acids: A review of monitoring and toxicological findings. *Toxicological Sciences* 99:366–394.
- LETCHER, R. J., J. O. BUSTNES, R. DIETZ, B. M. JENSSON, E. H. JØRGENSEN, C. SONNE, J. VERREAULT, M. M. VIJAYAN, AND G. W. GABRIELSEN. 2010. Exposure and effects assessment of persistent organohalogen contaminants in arctic wildlife and fish. *Science of the Total Environment* 408:2995–3043.
- LI, Y. F., R. W. MACDONALD, L. M. M. JAN-TUNEN, T. HARNER, T. F. BIDLEMAN, AND W. M. J. STRACHAN. 2002. The transport of  $\beta$ -hexachlorocyclohexane to the western Arctic Ocean: A contrast to  $\alpha$ -HCH. *Science of the Total Environment* 291:229–246.
- LI, Y. F., R. W. MACDONALD, J. M. MA, H. HUNG, AND S. VENKATESH. 2004. Historical  $\alpha$ -HCH budget in the Arctic Ocean: The Arctic Mass Balance Box Model (AMBBM). *Science of the Total Environment* 324:115–139.
- LINDBERG, P., U. SELLSTRÖM, L. HÄGGBERG, AND C. A. DE WIT. 2004. Higher brominated polybrominated diphenyl ethers and hexabromocyclododecane found in eggs of Peregrine Falcon (*Falco peregrinus*) breeding in Sweden. *Environmental Science and Technology* 38:3–96.
- MACDONALD, R. W., T. HARNER, AND J. FYFE. 2005. Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data. *Science of the Total Environment* 342:5–86.
- MARTIN, J. W., M. M. SMITHWICK, B. M. BRAUNE, P. F. HOEKSTRA, D. C. G. MUIR, AND S. A. MABURY. 2004. Identification of long-chain perfluorinated acids in biota from the Canadian Arctic. *Environmental Science and Technology* 38:373–380.
- MCKINNEY, M. A., E. PEACOCK, AND R. J. LETCHER. 2009. Sea ice-associated diet change increases the levels of chlorinated and brominated contaminants in Polar Bears. *Environmental Science and Technology* 43:4334–4339.
- MONTEIRO, L. R., J. P. GRANADEIRO, R. W. FURNESS, AND P. OLIVEIRA. 1999. Contemporary patterns of mercury contamination in the Portuguese Atlantic inferred from mercury concentrations in seabird tissues. *Marine Environmental Research* 47:137–156.
- MUIR, D. C. G., AND C. A. DE WIT. 2010. Trends of legacy and new persistent organic pollutants in the circumpolar Arctic: Overview, conclusions, and recommendations. *Science of the Total Environment* 408: 3044–3051.
- NORDLÖF, U., B. HELANDER, A. BIGNERT, AND L. ASPLUND. 2010. Levels of brominated

- flame retardants and methoxylated polybrominated diphenyl ethers in eggs of White-tailed Sea Eagles breeding in different regions of Sweden. *Science of the Total Environment* 409:238–246.
- NRIAGU, J. O., AND J. M. PACYNA. 1988. Quantitative assessment of worldwide contamination of air, water and soils by trace metals. *Nature* 333:134–9.
- ÓLAFSDÓTTIR, K., Æ. PETERSEN, S. THØRDARDÓTTIR, AND T. JÓHANNESSON. 1995. Organochlorine residues in Gyrfalcons (*Falco rusticolus*) in Iceland. *Bulletin of Environmental Contamination and Toxicology* 55:382–389.
- PACYNA, J. 2005. Sources and emissions. Pages 5–10 in *AMAP Assessment 2002: Heavy Metals in the Arctic*. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.
- PACYNA, E. G., J. M. PACYNA, F. STEENHUISEN, AND S. WILSON. 2006. Global anthropogenic mercury emission inventory for 2000. *Atmospheric Environment* 40:4048–4063.
- PEAKALL, D. B., D. G. NOBLE, J. E. ELLIOTT, J. D. SOMERS, AND G. ERICKSON. 1990. Environmental contaminants in Canadian Peregrine Falcons, *Falco peregrinus*: A toxicological assessment. *Canadian Field-Naturalist* 104:244–254.
- PEDERSEN, H. C., F. FOSSØY, J. A. KÅLÅS, AND S. LIERHAGEN. 2006. Accumulation of heavy metals in circumpolar Willow Ptarmigan (*Lagopus l. lagopus*) populations. *Science of the Total Environment* 371:176–189.
- RIGÉT, F., A. BIGNERT, B. BRAUNE, J. STOW, AND S. WILSON. 2010. Temporal trends of legacy POPs in Arctic biota, an update. *Science of the Total Environment* 408:2874–2884.
- THIS REFERENCE SHOULD BE CHANGED TO:  
RIGÉT, F., B. BRAUNE, A. BIGNERT, S. WILSON, J. AARS, E. BORN, M. DAM, R. DIETZ, M. EVANS, T. EVANS, M. GAMBERG, N. GANTNER, N. GREEN, H. GUNNLAUGSDÓTTIR, K. KANNAN, R. LETCHER, D. MUIR, P. ROACH, C. SONNE, G. STERN, AND Ø. WIIG. 2011. Temporal trends of Hg in Arctic biota, an update. *Science of the Total Environment* 409:3520–3526.
- SAVINOV, V. M., G. W. GABRIELSEN, AND T. N. SAVINOVA. 2003. Cadmium, zinc, copper, arsenic, selenium and mercury in seabirds from the Barents Sea: Levels, inter-specific and geographical differences. *Science of the Total Environment* 306:133–159.
- SEMKIN, R. 1996. Processes and fluxes of contaminants in aquatic systems. Pages 105–118 in J. L. Murray and R. G. Shearer (Eds.). *Synopsis of Research Conducted Under the 1994/95 Northern Contaminants Program*. Environmental Studies no. 73. Indian and Northern Affairs Canada, Ottawa, Ontario, Canada.
- SONNE, C., J. O. BUSTNES, D. HERZKE, V. L. B. JASPERS, A. COVACI, D. J. HALLEY, T. MOUM, I. EULAERS, M. EENS, R. A. IMS, S. A. HANSSON, K. E. ERIKSTAD, T. JOHNSEN, L. SCHNUG, F. F. RIGÉT, AND A. L. JENSEN. 2010. Relationships between organohalogen contaminants and blood plasma clinical-chemical parameters in chicks of three raptor species from Northern Norway. *Ecotoxicology and Environmental Safety* 73:7–17.
- STOCKHOLM CONVENTION ON POPs. 2010. Stockholm Convention on Persistent Organic Pollutants (POPs) website. [Online.] Available at: <http://www.chm.pops.int/default.aspx>
- TITTMER, S. A., K. PEPPER, G. TOMY, AND L. CHAN. 2005. Examination of dietary exposure to polyfluorinated compounds via consumption of traditional foods. Poster ANA041, FLUOROS symposium, Toronto, Ontario, Canada. [Online.] Available at <http://www.chem.utoronto.ca/symposium/fluoros/pdfs/ANA041Tittlemier.pdf>
- VERREAULT, J., U. BERGER, AND G. W. GABRIELSEN. 2007. Trends of perfluorinated alkyl substances in Herring Gull eggs from two coastal colonies in northern Nor-

- way: 1983–2003. *Environmental Science and Technology* 41:6671–6677.
- VERREAULT, J., R. A. VILLA, G. W. GABRIELSEN, J. U. SKAARE, AND R. J. LETCHER. 2006. Maternal transfer of organohalogen contaminants and metabolites to eggs of Arctic-breeding Glaucous Gulls. *Environmental Pollution* 144:1053–1060.
- VORKAMP, K., J. H. CHRISTENSEN, AND F. RIGET. 2004. Polybrominated diphenyl ethers and organochlorine compounds in biota from the marine environment of East Greenland. *Science of the Total Environment* 331:143–155.
- VORKAMP, K., M. THOMSEN, K. FALK, H. LESLIE, S. MØLLER, AND P. B. SØRENSEN. 2005. Temporal development of brominated flame retardants in Peregrine Falcon (*Falco peregrinus*) eggs from South Greenland (1986–2003). *Environmental Science and Technology* 39:8199–8206.
- VORKAMP, K., M. THOMSEN, S. MØLLER, K. FALK, AND P. B. SØRENSEN. 2009. Persistent organochlorine compounds in Peregrine Falcon (*Falco peregrinus*) eggs from South Greenland: Levels and temporal changes between 1986 and 2003. *Environment International* 35:336–341.
- WANIA, F. 2003. Assessing the potential of persistent organic chemicals for long-range transport and accumulation in polar regions. *Environmental Science and Technology* 37:1344–351.
- WANIA, F. 2006. Potential of degradable organic chemicals for absolute and relative enrichment in the Arctic. *Environmental Science and Technology* 40:569–577.
- WANIA, F. 2007. A global mass balance analysis of the source of perfluorocarboxylic acids in the Arctic Ocean. *Environmental Science and Technology* 41:4529–4535.
- WIENER, J. G., D. P. KRABBENHOFT, G. H. HEINZ, AND A. M. SCHEUHAMMER. 2003. Ecotoxicology of mercury. Pages 409–463 in D. J. Hoffman, B. A. Rattner, G. A. Burton Jr., and J. Cairns, Jr. (Eds.). *Handbook of Ecotoxicology*, 2<sup>nd</sup> Ed. Lewis Publishers, Boca Raton, Florida, USA.
- WOLKERS, H., B. VAN BAVEL, A. E. DEROCHE, Ø. WIIG, K. M. KOVACS, C. LYDERSEN, AND G. LINDSTRÖM. 2004. Congener-specific accumulation and food chain transfer of polybrominated diphenyl ethers in two arctic food chains. *Environmental Science and Technology* 38:1667–1674.

