Subpopulation-Specific Influence of Diet and Other Factors on the Spatial Trends of Chlorinated, Brominated and Fluorinated POPs in Polar Bears (*Ursus maritimus*)

Robert J. Letcher[1,], Melissa A. McKinney[1], Jon Aars[2], Erik W. Born[3], Marsha Branigan[4], Emily Choy[1], Rune Dietz[5], Thomas J. Evans[6], Geir W. Gabrielsen[2], Derek C.G. Muir[7], Elizabeth Peacock[8] and Christian Sonne[5]

[1] Ecotoxicology and Wildlife Health Division, Science and Technology Branch, Environment Canada, Ottawa (robert.letcher@ec.gc.ca, melissaamckinney@gmail.com, emily.s.choy@gmail.com)
[2] Norwegian Polar Institute, Tromsø, Norway (aars@npolar.no, gabrielsen@npolar.no)
[3] Greenland Institute of Natural Resources, Nuuk, Greenland (ewb@mail.ghsdh.dk)
[4] Department of Environment and Natural Resources, Government of the Northwest Territories, Inuvik, Northwest Territories, Canada (marsha_branigan@gov.nt.ca)
[5] Aarhus University, National Environmental Research Institute, Department of Arctic Environment, Roskilde, Denmark (rdi@dmu.dk, csh@dmu.dk)
[6] United States Fish and Wildlife Service, Anchorage, Alaska, United States (Thomas_Evans@fws.gov)
[7] National Water Research Institute, Environment Canada, Burlington, Canada (derek.muir@ec.gc.ca)
[8] United States Geological Survey, Anchorage, Alaska, United States (lpeacock@usgs.gov)

A broad suite of persistent organic pollutants (POPs) in the classes of legacy organochlorine (OC) contaminants and newer brominated flame retardants (BFRs) and per- and poly-fluorinated contaminants (PFCs) (including some precursors, metabolites and isomers) , were analyzed in adipose or liver tissue from 11 polar bear (*Ursus maritimus*) subpopulations spanning Alaska east to Svalbard and collected 2005 to 2008. The influence of subpopulation (region), age, sex and especially diet, were assessed with respect to the spatial trends of these POPs in polar bears. In adipose tissue samples, although 37 polybrominated diphenyl ethers (PBDEs), total-(α)-hexabromocyclododecane (HBCD), 2 polybrominated biphenyls (PBBs), pentabromotoluene, pentabromoethylbenzene, hexabromobenzene, 1,2-bis(2,4,6-tribromophenoxy(ethane) and decabromodiphenyl ethane were screened, only 4 PBDEs, total-(α-)HBCD and BB153 were consistently found. Geometric mean ΣPBDE (4.6–78.4 ng/g lipid weight (lw)) and BB153 (2.5–81.1 ng/g lw) levels were highest in East Greenland (43.2 and 39.2 ng/g lw, respectively), Svalbard (44.4 and 20.9 ng/g lw) and Western (38.6 and 30.1 ng/g lw) and Southern Hudson Bay (78.4 and 81.1 ng/g lw). Total-(α)-HBCD levels (0.3–41.1 ng/g lw) were lower than ΣPBDE levels in all subpopulations except in Svalbard. ΣPCB levels increased in adipose tissue of subpopulations going from west to east (1797–10537 ng/g lw), and were highest of all chlorinated and brominated POP classes under study. ΣCHL levels were more spatially uniform among subpopulations (765–3477 ng/g lw). ΣDDT levels were relatively low and spatially variable (31.5–206 ng/g lw). However, elevated proportions of *p,p'*-DDT to ΣDDT in Alaska and Beaufort Sea relative to other subpopulations suggested fresh inputs from vector control use in Asia and/or Africa. For PFCs, Σ-perfluorinated sulfonate (PFSA; C_4, C_6, C_8 and C_{10}) concentrations were comprised of >99% perfluorooctane sulfonate (PFOS) (298-2686 ng/g wet weight (ww)) among subpopulations. The Σ-perfluorinated carboxylate (PFCA; C_6 to C_{15}) concentrations among subpopulations ranged from 158-880 ng/g ww, and were comprised largely of C_9, C_{10} and C_{11}, with much lesser amounts of C_8, C_{12} and C_{13}, and very low or non-detectable C_6, C_7, C_{14} and C_{15} PFCAs. Spatial trends for the 2007-2008 samples indicated that ΣPFSA (PFOS) and ΣPFCA levels were highest in bears from East Greenland and Southern Hudson Bay, and lower but comparable among the Northern and Southern Beaufort Sea, Gulf of Boothia, Lancaster/Jones Sound, Davis Strait, Baffin Bay and Chuckchi/Bering Sea (Alaska) collected samples.

Dietary variation between Alaska, Canada, East Greenland, and Svalbard subpopulations was assessed by muscle nitrogen and carbon stable isotope (SI) ratios and adipose fatty acid (FA) signatures relative to their main prey (ringed seals). Western and southern Hudson Bay signatures were characterized by depleted SIs, lower proportions of C_{20} and C_{22} monounsaturated FAs and higher proportions of C_{18} and longer chain polyunsaturated FAs. East Greenland and Svalbard signatures were reversed relative to Hudson Bay. Alaskan and Canadian Arctic signatures were intermediate. Between subpopulation dietary differences predominated over inter-annual, seasonal, sex, or age variation. Among various OCs, BFRs and PFCs, diet signatures significantly explained variation in levels of e.g., PBDE flame retardants (14-15%) and legacy PCBs (18-21%). However, dietary influence was contaminant class-specific, since only low or non-significant proportions of variation in OC pesticide (e.g., chlordane) levels were explained by diet. Hudson Bay diet signatures were associated with lower PCB and PBDE levels, whereas East Greenland and Svalbard signatures were associated with higher levels. In addition to other confounding factors, understanding diet/food web factors is important to accurately interpret contaminant trends, particularly in a changing Arctic subject to the large and complex affects of climatic warming.