LEVELS AND TRENDS OF PERSISTENT ORGANIC POLLUTANTS IN THE ARCTIC ENVIRONMENT

Derek C.G. Muir¹, Cynthia A. de Wit², Aaron T. Fisk³

¹National Water Research Institute, Environment Canada, Burlington, ON L7R4A6, Canada ²Institute of Applied Environmental Research, Stockholm University, Stockholm, Sweden ³University of Georgia, School of Forest Resources, Athens, GA 30602-2152 USA

Introduction

Arctic biota were demonstrated to contain a range of persistent organic pollutants (POPs), particularly organochlorines (OCs) pesticides and PCBs in the 1970s, however, until the mid-1990s there was limited knowledge on the sources, pathways, levels, spatial and temporal trends and biological effects of these substances in Arctic species^{1,2}. A report by the Arctic Monitoring and Assessment Program (AMAP) reviewed the data on POPs in Arctic air, seawater, sediments, and terrestrial, freshwater and marine biota, that was available as of 1996³. That assessment has recently been updated with new data that has emerged in the last 5 years to fill important knowledge gaps⁴. There has been an expansion of the information available on levels of contaminants in air, seawater, fish, in top predators and in marine food web organisms in the past 5 years. The major findings on levels and trends of POPs, in that assessment, will be presented here.

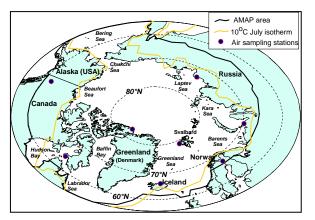
Sources and spatial trends

Information on global sources of POPs has also improved greatly with the publication of global gridded inventories for hexachlorocyclohexane (HCH)⁵, and PCBs⁶. Additional information on local sources within the Arctic has emerged. Harbours in Norway, Svalbard, Canada, and Russia, as well as areas around former military bases in Alaska, Canada and Norway (Jan Mayen; Svalbard) have been shown to have elevated levels of some PCBs in sediments and/or biota. Many of the former military sites in Canada, Alaska, Greenland and Norway have undergone cleanups in the past 5 years, which include removal of contaminated equipment and soils. The situation in the Russian Arctic regarding PCB use at military sites is unclear.

Less is known about sources of dioxin-like PCBs and polychlorinated dibenzo-p-dioxins and – dibenzofurans (PCDD/Fs) in the Arctic. A recent inventory, based on US and Canada per-capita dioxin emissions estimated for 1996, shows most of the Arctic region of North America having emissions of <0.1 ug TEQ/km²/yr and more populated areas (e.g. southern Alaska, southern Yukon, Labrador) with estimated emissions of 0.1-25 ug TEQ/km²/yr⁷. This is consistent with the very low population density given the total population of the North American Arctic of about 650,000 and land area of approximately $7.5 \times 10^6 \text{ km}^2$. By comparison the densely populated regions of North America have emissions of 100-5000 ug TEQ/km²/yr and higher values for point sources such as municipal waste incinerators. There is also local open, low temperature, burning of municipal garbage in the Canadian Arctic, Alaska and Russia, which is probably contributing to loadings of PCDD/Fs to the Arctic atmosphere. Like the PCB emissions from former military sites these emissions may be locally important sources, i.e. within a short radius of each community.

Measurements of air-borne POPs have continued at Alert (Nunavut, Canada), Pallas (Finland),

Storhovdi (Iceland), and Ny-Ålesund, Figure 1. Map of the Arctic showing the area covered by the (Svalbard, Norway) and a new site at AMAP assessment and location of air sampling stations Amderma (western Russia) began operation in 1999. Location of the stations is shown in Figure 1. Major OCs in Arctic air are α-and γ-HCH, p,p'-DDT, chlordane (CHL) compounds, endosulfan, pentachloro-anisole (PeCA), HCB and PCBs (10 to about 100 congeners)^{8,9}. Results suggest rather uniform average concentrations of most OC compounds. Amderma, in northwest Russia had average $\Sigma 10PCB$ of 4 pg/m³ in 1999-00 (the first year of operation of this site), similar to Alert and lower than Pallas or Storhofdi. New chemicals detected at several of these sites include



PBDEs, chlorinated paraffins, chlorinated naphthalenes, PeCA, tetrachloro-veratroles, Air mass back trajectories show that PCBs measured at Alert on northern Ellesmere Island originated primarily from North American sources, however, Central and Eastern Russia were also potential sources based on prevailing air mass movements¹⁰. In the Yukon and Alaska, Asian sources likely predominate based on known air mass movements^{10,11}. Less is known about long range atmospheric transport of PCDD/Fs to the North American Arctic. Commoner et al.¹² predicted total PCDD/F deposition of about 4 to 53 pg TEQ/m² yr to terrestrial surfaces near 8 communities in the eastern Canadian Arctic. PCDD/Fs were determined in soils from Alaska, the Yukon and NWT in the Canadian Arctic, western Greenland, and Norway and northwestern Russia^{13, 14}. Fluxes of total PCDD/Fs estimated from the mass of chemical in a 10x10cm (15 cm depth) core ranged from 1.2-143 ng/m^2 yr. The authors concluded that PCDD/Fs did not move appreciably from warm to cold latitudes. Most Arctic soil samples were characterized by high proportions of OCDD although TCDFs were also important contributors to total PCDD/Fs.

Knowledge of levels of persistent OCs in Arctic wildlife has expanded in the past 5 years especially in the case of Alaska, Greenland, Svalbard/Barents Sea, and western Arctic Russia⁴. Species studied for the first time in detail include marine zooplankton (*Calanoid* sp.), mussels (Mytilus sp) (Alaska, Faroe Islands, and Canada), soils (Norway, Canada/Russia), lichen and mosses (Russia/Alaska), bowhead whales and Arctic fox (in Alaska and Canada), bearded seals (Alaska and Russia), bald eagles (Alaska/Aleutians), black guillemots (Faroes) and Greenland shark (Canada/Greenland). Measurements continued on many key AMAP species such as Atlantic cod, beluga whales, ringed seals, polar bears, seabird eggs and liver (kittiwake, thick-billed murre, northern fulmar, black guillemots). This work confirmed previous observations that levels of total PCBs (Σ PCB) and total DDT (Σ DDT) compounds in polar bear, ringed seals and seabirds, are highest in the European Arctic and lowest in the western North American Arctic while HCH isomers (Σ HCH) are higher in the North American Arctic. Highest concentrations of PCBs in polar bear were found in samples from the Kara Sea and Franz Josef Land. A surprising finding has been relatively high toxaphene in harp and ringed seals from Svalbard and the White Sea. The

circumpolar trend of OCs in zooplankton differs from that observed in higher trophic level organisms in that there were not large differences in levels of major OCs between the North American Arctic and the European Arctic.

Transport of POPs in migratory animals may also be a significant pathway, in the Arctic, where many important species such as seabirds, cetaceans (beluga, bowhead, minke) and pinnipeds (harp seals), salmon and Atlantic cod are migratory⁴. Ewald et al.¹⁵ described the entry of PCBs and DDT into the grayling population of the Copper River in Alaska by migrating sockeye salmon. The grayling in a salmon spawning lake had contaminant concentrations more than double those found in grayling from a nearby, but salmon-free, lake. Detailed studies of the food web of Lake Ellasjøen at Bjørnøya (Bear Island) have shown that a biological pathway (seabird guano) is probably responsible for the elevated levels of OCs in Arctic char from this lake¹⁶.

Temporal trends

Hung et al.^{8,9} found significant declines of α -HCH, γ -HCH, PeCA, chlordane-related compounds and PCB 28, 101, 153 and 180 over a 5-year period (1993-1998) as well as increases in endosulfan and p,p'-DDE. Detailed analysis (including temperature normalization of the data) of temporal trends at the other air sampling sites has not been carried out.

The information available on temporal trends of POPs in Arctic biota has been extended so that a 25 to 30 year perspective is available for polar bears, seabirds and ringed seals in the Canadian Arctic, as well as for fish in the Swedish Arctic. Studies covering a 10-15 year period are available for polar bear at Svalbard, peregrine falcons in Alaska, Atlantic cod in Iceland, glaucous gulls in Svalbard, burbot, lake trout and beluga whales in Canada and walrus in northwestern Greenland. Levels of Σ PCB, Σ DDT, Σ HCH and HCB in pike and Arctic char from the Swedish Arctic have declined significantly since the late 1960s into the 1990s, decreasing at a rate of approximately 4% to 10% per year⁴. However, no significant decline has been observed for the past 10 years for HCH isomers and HCB, while Σ PCB and Σ DDT continue to decline at about the same rate in the 1990s as in the 1980s. Σ DDT and Σ HCH both declined significantly in liver of burbot from 8 Russian north flowing rivers between 1988 and 1994 in parallel to declines of these pesticides in river water¹⁷.

Levels of Σ PCB and Σ DDT decreased significantly in eggs of kittiwake, thick-billed murre, northern fulmar, in the Canadian Arctic archipelago (1975-1998), while Σ CHL, dieldrin and mirex levels decreased in kittiwake eggs only¹⁸. Σ DDT also declined significantly in female ringed seals from three locations in the Canadian Arctic archipelago, between the early/mid-1970's to late-1990's/2000. Σ DDT had the largest decline of any "legacy" organochlorines – 2.5 x at Ausuittuq, 3.3 x at Ikajutit and 2.0x at Sachs Harbour over 13-30 years¹⁹. Temporal trends of PCB 153 concentrations in polar bear blood were studied using annual samples collected between 1990 and 1998²⁰. Decreases of ca. 40% occurred in the early 1990s, and stabilized thereafter. This is a more rapid trend than was found for PCB 153 in Hudson Bay polar bears during the same time period.

Ikonomou et al.²¹ reported exponential increases in total (Br_2-Br_7)-PBDEs (Σ PBDEs) in male ringed seals aged 0-15 years from Holman in the western Canadian Arctic over the period 1981 to 2000. Σ PBDEs increased 9-fold over this period. Similar increase of PBDEs were found in beluga

whales²². The doubling time for PBDEs in beluga during the early 1990s ($t_{2=3.0}$ years) was faster than in Holman ringed seals (t2=4.5 years).

References

¹ Muir, D.C.G., Wagemann, R., Hargrave, B.T., Thomas, D., Peakall, D.B. and Norstrom, R.J. (1992). Sci. Total Environ. <u>122</u>, 75.

²Barrie, L.A., Gregor, D., Hargrave, B., Lake, R., Muir, D.C.G., Shearer, R., Tracey, B., and Bidleman, T. (1992). Sci. Total Environ. 122, 1.

³de March, B.G.E., C.A. de Wit and D.C.G. Muir (Eds). (1998). In: Arctic Assessment Report, Chapter 6. Arctic Monitoring and Assessment Programme. Oslo. Norway. Pp. 183-372.

⁴de Wit, C., Fisk, A.T. Hobbs, K.E. Muir, D.C.G. Gabrielsen, G., Kallenborn, R., Krahn, M., Norstrom, R., and Skaare, J. (2002). In: Arctic Assessment Report, Arctic Monitoring and Assessment Programme. Oslo. Norway, in press.

⁵Li, Y.F. (1999). J. Geophys. Res. 104, D19, 23,785.

⁶Breivik, K., Sweetman, A., Pacyna J.M., and Jones, K.C. (2002). Sci. Total Environ. 290, 199. ⁷Cohen, M.D., Draxler, R.R., Artz, R., Commoner, B., Bartlett, P., Cooney, P., Couchot, K.,

Dickar, A., Eisl, H., Hill, C., Quigley, J., Rosenthal, J.E., Niemi, D., Ratte, D., Deslauriers, M.,

Laurin, R., Mathewson-Brake, L., McDonald, J. (2002). Environ. Sci. Technol. 36, 4831.

⁸Hung, H., Halsall, C.J., Blanchard, P., Li, H.H., Fellin, P., Stern, G., and Rosenberg, B. (2001). Environ. Sci. Technol. 35:1303.

⁹Hung, H., Halsall, C.J., Blanchard, P., Li, H.H., Fellin, P., Stern, G., and Rosenberg, B. (2002). Environ. Sci. Technol. 36, 862.

¹⁰Stern, G.A., Halsall, C.J., Barrie, L.A., Muir, D.C.G., Fellin, P., Rosenberg, B., Rovinsky, F.Ya, Kononov E.Ya. and Pastuhov, B. (1997). Environ. Sci. Technol. 31, 3619.

¹¹Bailey, R., Barrie, L.A., Halsall, C.J., Fellin, P., and Muir, D.C.G. (2000). J. Geophys. Res. Atmos. 105:11805.

¹²Commoner, B., Barlett, P.W., Eisl, H., and Couchot, K. (2000). North American Commission for Environmental Cooperation, Montreal QC (www.cec.org). 101 pp.

¹³Wagrowski, D.M., and Hites, R.A. (2000) Environ. Sci. Technol. <u>34</u>, 2952.

¹⁴Brzuzy, L.P., and Hites, R.A. (1996). Environ. Sci. Technol. 30, 1797.

¹⁵ Ewald, G., Larsson, P., Linge, H., Okla, L., and Szarzi, N. (1998). Arctic <u>51</u>, 40.

¹⁶Evenset, A., Christensen, G., Kallenborn, R., and Skotvold, T. (2002). Akvaplan-niva report. Tromso, Norway. 33pp.

¹⁷Zhulidov, A.V., Robarts, R.D., Headley, J.V., Liber, K., Zhulidov, D.A., Zhulidova, A.V., and Pavlov, D.F. (2002). Sci. Total Environ. 293, 231.

¹⁸Braune, B.M., Donaldson, G.M., and Hobson, K.A. (2001). Environ. Pollut. 114, 39.

¹⁹Muir, D., Fisk, A., and Kwan, M. (2001). In: S. Kalhok (Ed.), Synopsis of Research Conducted Under the 2000-2001 Northern Contaminants Program, pp. 208-214. Indian and Northern Affairs Canada, Ottawa, ON, Canada.

²⁰Henriksen, E.O., Wiig, Ø., Skaare, J.U., Gabrielsen, G.W., and Derocher, A.E. (2001). J. Environ. Monit., 3,493.

²¹Ikonomou, M.G., Rayne, S., and Addison, R.F. (2002). Environ. Sci. Technol. 36:1886.

²²Stern, G.A. and Ikonomou, M. (2000). In: Kalhok, S. (Ed), Synopsis of Research Conducted Under the 1999-2000 Northern Contaminants Program, pp. 227-232. Indian and Northern Affairs Canada. Ottawa, ON.