

SUPPORTING INFORMATION

**Influences of climate change on long-term time series
of persistent organic pollutants (POPs) in Arctic and Antarctic biota**

Katrin Vorkamp^{*},^a Pernilla Carlsson,^b Simonetta Corsolini,^c Cynthia A. de Wit,^d Rune Dietz,^e
Matthew O. Gribble,^f Magali Houde,^g Vrinda Kalia,^h Robert J. Letcher,ⁱ Adam Morris,^j
Frank F. Rigét,^e Heli Routti,^k Derek C.G. Muir^l

^a Aarhus University, Department of Environmental Science, Roskilde, Denmark;

^b Norwegian Institute for Water Research (NIVA), Fram Centre, Tromsø, Norway;

^c University of Siena, Department of Physical, Earth and Environmental Sciences, Siena, Italy;

^d Stockholm University, Department of Environmental Science, Stockholm, Sweden;

^e Aarhus University, Department of Ecoscience, Roskilde, Denmark;

^f University of Alabama at Birmingham (UAB), School of Public Health, Birmingham, AL, USA;

^g Environment and Climate Change Canada, Montréal, QC, Canada;

^h Columbia University, Department of Environmental Health Sciences, New York, NY, USA;

ⁱ Environment and Climate Change Canada, Ottawa, ON, Canada;

^j Northern Contaminants Program, Crown-Indigenous Relations and Northern Affairs, Gatineau, QC, Canada;

^k Norwegian Polar Institute, Fram Centre, Tromsø, Norway;

^l Environment and Climate Change Canada, Burlington, ON, Canada

^{*} Corresponding author. Postal address: Aarhus University, Department of Environmental Science, Frederiksborgvej 399, 4000 Roskilde, Denmark. E-mail: kvo@envs.au.dk

Table S1: List of abbreviations used in this article

Abbreviation	Full name
AMAP	Arctic Monitoring and Assessment Programme
AO	Arctic Oscillation
C	Carbon
CBz	Chlorobenzene
CEAC	Chemical of emerging Arctic concern
CHL	Chlordane
DDD	Dichlorodiphenyldichloroethane
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
ESB	Environmental Specimen Bank
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
N	Nitrogen
NAO	North Atlantic Oscillation
NIST	National Institute for Standards and Technology
OCS	Octachlorostyrene
PBDE	Polybrominated diphenyl ether
PCB	Polychlorinated biphenyl
PCDD	Polychlorinated dibenzo- <i>p</i> -dioxin
PCDF	Polychlorinated dibenzofuran
PDO	Pacific Decadal Oscillation
PFAS	Perfluoroalkyl substance
PFCA	Perfluorocarboxylic acid
PFOS	Perfluorooctane sulfonate
PFTTrDA	Perfluorotridecanoic acid
PNA	Pacific/North American Pattern
POP	Persistent organic pollutant
STAMP	Seabird Tissue Archival and Monitoring Project
TMF	Trophic magnification factor

Table S2: Summary of the studies investigating temporal trends of persistent organic pollutants in Arctic and Antarctic biota in relation to climate parameters.

Species	Location	Years	Contaminants	Climate-related parameters	Associations	Reference
Freshwater biota						
Zooplankton, Arctic char	Canada	2008–2017	PCBs, CHLs, DDTs	Turbidity (proxy for permafrost slumps)	Higher concentrations of POPs in biota from lakes with higher carbon and particulate concentrations	Cabrerizo et al. (2019a; b)
Arctic char	Canada	1989–2015 ^a	ΣPCBs, ΣDDTs, HCB, ΣHCHs, toxaphene	North Atlantic Oscillation (NAO) index, temperature (air), precipitation	Positive association between HCH and precipitation; positive associations between ΣPCBs, ΣDDTs, ΣHCHs and NAO in the preceding year	Cabrerizo et al. (2018)
Arctic char	Greenland	1999–2017	ΣPCBs, PCB-52, PCB-153, ΣDDTs, α-HCH, HCB	Biological variables (δ ¹⁵ N), Arctic Oscillation (AO) index, air temperature	Positive associations between ΣPCBs, α-HCH and air temperature	Rigét et al. (2020)
Arctic char	Norway (Bjørnøya)	1998–2017	ΣPCBs, HCB	Air temperature	Positive associations between HCB and air temperature, but indications of confounding factors	AMAP (2021)
Burbot	Canada	2000–2012	PCBs, DDTs	Air temperature	Positive associations between primary productivity and concentrations of ΣDDTs and PCB congeners, but not over the entire timespan	Carrie et al. (2010)
Terrestrial mammals						
Arctic fox	Norway (Svalbard)	1997–2013	PCBs, PBDEs, CHLs, <i>p,p'</i> -DDT, <i>p,p'</i> -DDE, β-HCH, HCB, mirex	Biological variables (δ ¹³ C, δ ¹⁵ N), sea-ice coverage (proxy for seal availability), reindeer mortality	Positive association between POPs and intake of marine diet; positive association between β-HCH and sea-ice cover; negative association between HCB and reindeer mortality	Andersen et al. (2015)
Arctic fox	Norway (Svalbard)	1997–2014	PFAS	Biological variables (δ ¹³ C, δ ¹⁵ N), sea-ice coverage (proxy for seal availability), reindeer mortality	Positive association between PFAS and intake of marine and high trophic level prey. Positive association between PFOS and sea-ice cover; negative association between PFCA and reindeer mortality	Routti et al. (2017)
Marine fish						
Emerald rockcod	Antarctica	1981–2010 ^a	PCBs, PBDEs, PCDD/Fs, CHLs, DDTs, HCHs, HCB	Iceberg calving event	Increase of concentrations of ΣPCBs, <i>p,p'</i> -DDE and ΣPBDEs after iceberg calving event	Cincinelli et al. (2016)
Seabirds						
Adélie penguins	Antarctica	1964–2005	DDTs	Glacier melt timing	Stable concentrations of ΣDDT (instead of expected decrease) related to release from glacier meltwater	Geisz et al. (2008)
Common murre	United States (Alaska)	1999–2010	ΣPCBs, <i>p,p'</i> -DDE, HCB, α-HCH, <i>cis</i> -nonachlor, dieldrin, heptachlor epoxide, oxychlordane, mirex	Pacific Decadal Oscillation (PDO) index	No association found between POP concentrations and PDO (different from results for thick-billed murres, see below)	Kalia et al. (2021)
Glaucous gull	Norway (Bjørnøya)	1997–2006	ΣPCBs, HCB, oxychlordane	AO index	Positive associations of ΣPCBs, HCB and oxychlordane with AO in preceding summer and winter; but negative associations with AO in the same year	Bustnes et al. (2010)
Northern fulmar	Canada	1975–2015 ^a	PCBs, CHLs, CBz, DDTs,	AO and NAO indices,	Positive associations between chlorobenzenes, <i>cis</i> -/ <i>trans</i> -	Foster et al. (2019)

			OCS, mirex, photomirex	precipitation, sea-ice extent, air and sea surface temperature	nonachlor, dieldrin, mirex, photomirex and NAO	
Thick-billed murre	Canada	1975–2015 ^a	PCBs, CHLs, CBz, DDTs, OCS, mirex, photomirex	AO and NAO indices, precipitation, sea-ice extent, air and sea surface temperature	Positive associations between some PCB congeners, <i>p,p'</i> -DDE, chlorobenzenes, OCS, dieldrin and precipitation; negative associations between PCB-170, PCB-180, oxychlordane, <i>cis</i> -nonachlor, heptachlor epoxide and precipitation	Foster et al. (2019)
Thick-billed murre	Canada	1975–2013 ^a	ΣPCB, <i>p,p'</i> -DDE, HCB, oxychlordane, heptachlor epoxide, dieldrin	Biological variables ($\delta^{15}\text{N}$)	Positive association between trophic position and rate of POP decline	Braune et al. (2015)
Thick-billed murre	Canada	1993–2015	PCB-153, PBDE-47, PFOS, <i>p,p'</i> -DDE, α -HCH	AO and NAO indices, precipitation, sea-ice coverage, sea level pressure ^a , air and land surface temperature, wind speed	Positive associations between POP concentrations and AO or NAO indices (after a time-lag); positive associations of POPs with greater ice-coverage and earlier freeze-up of sea-ice; negative association between POP concentrations and temperature	AMAP (2021)
Thick-billed murre	United States (Alaska)	1999–2010	ΣPCBs, <i>p,p'</i> -DDE, HCB, α -HCH, <i>cis</i> -nonachlor, dieldrin, heptachlor epoxide, oxychlordane, mirex	PDO index	POP concentrations were highest when PDO was near 0 and lowest during PDO extremes.	Kalia et al. (2021)
Marine mammals						
Beluga	Canada	1989–2015	PCBs, CHLs, DDTs, HCHs, HCB, dieldrin, mirex	Biological variables ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$)	Associations between ΣPCBs and $\delta^{13}\text{C}$; associations between dieldrin, mirex and both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$	Noël et al. (2018)
Ringed seal	Canada (multiple locations)	1972–2016 ^a	ΣPCBs, ΣCBz/HCB, ΣCHLs, ΣDDTs, ΣHCHs, oxychlordane, trans-nonachlor ^b	AO and NAO indices; Pacific/North American (PNA) pattern; sea-ice coverage, air temperature	Positive associations between POPs and AO/NAO of preceding year, negative associations between POPs and AO/NAO of the year of sampling for most locations, but variations between locations; positive associations of ΣPCB and ΣCBz and sea-ice coverage	Houde et al. (2019)
Ringed seal	Canada	1993–2008	PCBs, CHLs, DDTs, HCB, HCHs, dieldrin, mirex, toxaphene	Sea-ice (time of breakup)	Higher concentrations of some PCBs and <i>p,p'</i> -DDT in years with earlier sea ice break-up	Gaden et al. (2012)
Ringed seal	Greenland (East)	1986–2017	ΣPCB, PCB-52, PCB-153, ΣDDT, α -HCH, HCB	Biological variables ($\delta^{15}\text{N}$), AO index, sea-ice coverage, seawater temperature	Positive association between all compounds and $\delta^{15}\text{N}$; positive association between HCB, α -HCH and AO; positive association between HCB and seawater temperature	Rigét et al. (2020)
Ringed seal	Greenland (West)	1994–2017	ΣPCB, PCB-52, PCB-153, ΣDDT, α -HCH, HCB	Biological variables ($\delta^{15}\text{N}$), AO index, sea-ice coverage, seawater temperature	Positive association between PCB-52 and $\delta^{15}\text{N}$; negative association between AO and α -HCH; positive association between ΣPCB, PCB-52, PCB-153, ΣDDT and temperature; positive association between ΣPCB, ΣDDT and salinity; positive association between sea ice coverage and all compounds except α -HCH	Rigét et al. (2020)

Polar bear	Canada	1991–2007	ΣPCBs, ΣPBDEs, ΣCHLs, ΣDDTs, α -HCH, β -HCH	Biological variables ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, fatty acid signatures), sea-ice (time of breakup)	Associations between POP concentrations and diet composition; associations between time of sea-ice break-up and diet composition	McKinney et al. (2009)
Polar bear	Canada	WHB ^c : 1991–2015 SHB ^d : 2007 or 2008–2015	PCB-153, PBDE-47, PFOS, p,p' -DDE, α -HCH	AO and NAO indices, precipitation, sea-ice coverage, sea level pressure ^a , air and land surface temperature, wind speed	Positive associations between POP concentrations and AO or NAO indices (after a time-lag); positive associations of POPs with later freeze-up of sea-ice; negative association between POP concentrations and temperature as well as precipitation	AMAP (2021)
Polar bear	Greenland	1984–2011	ΣPCBs, ΣPBDEs, PFAS, ΣCHLs, ΣDDTs, HCB, ΣHCHs, OCS, dieldrin, mirex, toxaphene	Biological variables ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, fatty acid signatures)	Associations between POP concentrations and diet composition	Dietz et al. (2021)
Polar bear	Norway (Svalbard)	2000–2014	PFAS	Biological variables ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$), sea-ice (proxy for seal availability)	Positive associations of most PFAS with trophic level / intake of marine diet	Routti et al. (2017)
Polar bear	Norway (Svalbard)	1997–2017	ΣPCBs, ΣOH-PCBs, PBDE-47, PBDE-153, p,p' -DDE, HCB, β -HCH, oxychlordan	Biological variables ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$)	Positive associations of POPs with trophic level / intake of marine diet	Lippold et al. (2019)

AO: Arctic Oscillation; CBz: chlorobenzene; CHL: chlordan; DDE: dichlorodiphenyldichloroethylene; DDT: dichlorodiphenyltrichloroethanes; HCB: hexachlorobenzene; HCH: hexachlorocyclohexane; NAO: North Atlantic Oscillation; OCS: octachlorostyrene OH-PCB: hydroxylated polychlorinated biphenyl; PBDE: polybrominated diphenyl ether; PCB: polychlorinated biphenyl; PCDD/F: polychlorinated dibenzo-*p*-dioxins and furan; PDO: Pacific Decadal Oscillation; PFAS: per- and polyfluoroalkyl substance; PFOS: perfluorooctane sulfonic acid; PNA: Pacific/North American Pattern; ^aVarying between locations and/or compound groups; ^bmultiple other compounds analysed, but not included in analysis with climate parameters; ^cWestern Hudson Bay; ^dSouthern Hudson Bay

References

- AMAP, 2021. AMAP Assessment 2020: POPs and Chemicals of Emerging Arctic Concern: Influences of Climate Change. Arctic Monitoring and Assessment Programme (AMAP), Tromsø, Norway. viii+142 pp
- Andersen, M.S., E. Fuglei, M. König, I. Lipasti, Å.Ø. Pedersen, A. Polder, N.G. Yoccoz and H. Routti, 2015. Levels and temporal trends of persistent organic pollutants (POPs) in arctic foxes (*Vulpes lagopus*) from Svalbard in relation to dietary habits and food availability. *Science of the Total Environment*, 511: 112-122
- Braune, B.M., A.J. Gaston, K.A. Hobson, H.G. Gilchrist and M.L. Mallory, 2015. Changes in trophic position affect rates of contaminant decline at two seabird colonies in the Canadian Arctic. *Ecotoxicology and Environmental Safety*, 115: 7-13
- Bustnes, J.O., G.W. Gabrielsen and J. Verreault, 2010. Climate variability and temporal trends of persistent organic pollutants in the Arctic: a study of glaucous gulls. *Environmental Science and Technology*, 44: 3155-3161
- Cabrerizo, A., D.C. Muir, G. Köck, D. Iqaluk, and X. Wang, 2018. Climatic influence on temporal trends of polychlorinated biphenyls and organochlorine pesticides in landlocked char from lakes in the Canadian High Arctic. *Environmental Science and Technology*, 52: 10380-10390

- Cabrerizo, A., D.C.G. Muir, C. Teixeira, S.F. Lamoureux and M. J. Lafrenière, 2019a. Snow deposition and melting as drivers of polychlorinated biphenyls and organochlorine pesticides in Arctic rivers, lakes, and ocean. *Environmental Science and Technology*, 53: 14377-14386
- Cabrerizo, A., D.C. Muir, A.O. De Silva, S. Lamoureux and M. Lafrenière, 2019b. Influence of permafrost disturbances on temporal trends of legacy and emerging persistent organic pollutants in landlocked Arctic char from lakes in the Canadian High Arctic. Society of Environmental Toxicology and Chemistry (SETAC) Europe 29th Annual Meeting, 26-30 May 2019, Helsinki, Finland
- Carrie, J., F. Wang, H. Sanei, R.W. Macdonald, P.M. Outridge and G.A. Stern, 2010. Increasing contaminant burdens in an Arctic fish, burbot (*Lota lota*), in a warming climate. *Environmental Science and Technology*, 44(1): 316-322
- Cincinelli, A., T. Martellini, T. K. Pozo, P. Kukučka, O. Audy and S. Corsolini, 2016. *Trematomus bernacchii* as an indicator of POP temporal trend in the Antarctic seawaters. *Environmental Pollution*, 217: 19-25
- Dietz, R., F.F. Rigét, I. Eulaers, J.-P. Desforges, K. Vorkamp, R. Bossi, J. Søndergaard, P. Ambus, M. McKinney, R.J. Letcher and C. Sonne, 2021. Unexpected increases of persistent organic pollutants and mercury levels in East Greenland polar bears (UNEXPECTED). Aarhus University, DCE – Danish Centre for Environment and Energy, 44 pp. Technical Report no. 214. <http://dce2.au.dk/pub/TR214.pdf>
- Foster, K.L., B.M. Braune, A.J. Gaston and M.L. Mallory, 2019. Climate influence on legacy organochlorine pollutants in Arctic seabirds. *Environmental Science and Technology*, 53: 2518-2528
- Gaden, A., S.H. Ferguson, L. Harwood, H. Melling, J. Alikamik and G.A. Stern, 2012. Western Canadian Arctic ringed seal organic contaminant trends in relation to sea ice break-up. *Environmental Science and Technology*, 46: 4427-4433
- Geisz, H.N., R.M. Dickhut, M.A. Cochran, W.R. Fraser and H.W. Ducklow, 2008. Melting glaciers: A probable source of DDT to the Antarctic marine ecosystem. *Environmental Science and Technology*, 42: 3958-3962
- Houde, M., X. Wang, T.L.L. Colson, P. Gagnon, S.H. Ferguson, M.G. Ikonou, C. Dubetz, R.F. Addison and D.C.G. Muir, 2019. Trends of persistent organic pollutants in ringed seals (*Phoca hispida*) from the Canadian Arctic. *Science of the Total Environment*, 665: 1135-1146
- Kalia, V., S.S. Schuur, K.A. Hobson, H.H. Chang, L.A. Waller, S.R. Hare and M.O. Gribble, 2021. Relationship between the Pacific Decadal Oscillation (PDO) and persistent organic pollutants in sympatric Alaskan seabird (*Uria aalge* and *U. lomvia*) eggs between 1990 and 2010. *Chemosphere*, 262: 127520
- Lippold, A., S. Bourgeon, J. Aars, M. Andersen, A. Polder, J.L. Lyche, J. Bytingsvik, B.M. Jenssen, A.E. Derocher, J.M. Welker and H. Routti, 2019. Temporal trends of persistent organic pollutants in Barents Sea polar bears (*Ursus maritimus*) in relation to changes in feeding habits and body condition. *Environmental Science and Technology* 53: 984-995
- 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
- Noël, M., L.L. Loseto and G. Stern, 2018. Legacy contaminants in the eastern Beaufort Sea beluga whales (*Delphinapterus leucas*): Are temporal trends reflecting regulations? *Arctic Science*, 4: 373-387

Rigét, F., K. Vorkamp, I. Eulaers and R. Dietz, 2020. Influence of climate and biological variables on temporal trends of persistent organic pollutants in Arctic char and ringed seals from Greenland. *Environmental Science: Processes & Impacts*, 22: 993-1005

Routti, H., J. Aars, E. Fuglei, L. Hanssen, K. Lone, A. Polder, Å.Ø. Pedersen, S. Tartu, J.M. Welker and N.G. Yoccoz, 2017. Emission changes dwarf the influence of feeding habits on temporal trends of per- and polyfluoroalkyl substances in two Arctic top predators. *Environmental Science and Technology*, 51: 11996-12006