Environmental Science Advances

ROYAL SOCIETY OF CHEMISTRY

TUTORIAL REVIEW

View Article Online
View Journal | View Issue

Cite this: *Environ. Sci.: Adv.*, 2025, **4**, 990

Do Arctic local sources of pollution influence the exposure of ringed seals (*Pusa hispida*) analyzed in contaminant monitoring programs?†

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The monitoring of contaminants in Arctic wildlife is mainly designed for long-range transported chemicals and supports international initiatives to mitigate global threats from chemical pollution. As human activities increase in the Arctic, the question of potential impacts of local pollution on wildlife exposure has emerged. In this study, we examined the current monitoring practices and the state of knowledge of pollutants in Arctic ringed seals (*Pusa hispida*) from Canada, Greenland, Norway, Russia, and Alaska. We complemented this synthesis with a case study of ringed seal exposure from different locations in Canada (2008–2022) to test the potential influence of local pollution sources. Multivariate constrained ordination was applied to examine the effects of ringed seals biological metrics (age, sex, length, and stable isotopes as dietary proxies), settlement size, and potential local contaminant sources (infrastructures, industries) on contaminant concentrations [polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), per- and polyfluoroalkyl substances (PFAS), polybrominated diphenyl ethers (PBDEs), trace elements] in seals. Results indicated that biological data, location, local source variables and sampling year explained 8.4% (PBDEs), 31.8% (OCPs), 41.0% (trace elements), 45.1% (PFAS), and 65.8% (PCBs) of the contaminant variation in seals through time. Variation partitioning showed that, out of these percentages, local source variables uniquely explained less than 4% of variation for PBDEs, OCPs, trace elements and PFAS compared to 17.5% for PCBs. Results suggest an effect of local contaminant sources that is tightly linked to study site characteristics as well as seal biology. Local sources should be identified and assessed at sampling sites close to communities to ensure efficient contaminant monitoring and effectiveness of mitigation measures.

Received 12th December 2024
Accepted 14th April 2025

DOI: 10.1039/d4va00418c

rsc.li/esadvances

Environmental significance

The monitoring of contaminants in Arctic wildlife is important to assess the long-range transport of chemicals and support international initiatives mitigating pollution. Local sources of pollution may increase with climate change and socioeconomic development of the Arctic and potentially influence levels of contaminants found in wildlife. This study examined the state of knowledge of pollutants in ringed seals (*Pusa hispida*) across the Arctic, identified examples of ringed seal exposure to local sources of pollution, and presents a preliminary analysis of the influences of local pollution in ringed seals across Canada. Results suggest an effect of local contaminant sources that is tightly linked to study sites and seal biology. Recommendations are given to better assess the potential influence of local sources in wildlife contaminant monitoring are suggested.

Introduction

Pollution is a global issue impacting ecosystems worldwide, including remote polar regions. The Arctic Monitoring and Assessment Programme (AMAP) was established in the early 1990s to monitor and assess environmental contaminants and their effects in Arctic regions.[1] For decades, AMAP has investigated levels and trends of pollutants in various environmental media and human samples and their effects on ecosystems and human health and presented these findings in scientific assessment reports and summaries for policymakers.[2,3] The long-term monitoring of contaminants in Arctic wildlife has two main purposes. The first is to assess pollutant concentrations in the Arctic environment, including their change over time, which is important information for chemicals management. This includes international initiatives to mitigate global threats from contaminants such as the United Nations Stockholm Convention

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† Electronic supplementary information (ESI) available. See DOI: https://doi.org/10.1039/d4va00418c

on Persistent Organic Pollutants (POPs). Thus, an important aspect of the Arctic monitoring programs has been to document long-range environmental transport to remote regions, as one of the screening criteria of the Stockholm Convention, as well as the effectiveness of international regulations. The second major purpose of the monitoring programs is to generate information on the potential exposure of Arctic Indigenous and local populations, who rely on local fish and wildlife in their traditional diet. It has been documented repeatedly that human populations in the Arctic are highly exposed to POPs and mercury because of the bioaccumulative characteristics of these chemicals and their biomagnification through the food web.[3]

The pollutants monitored in Arctic biota include chemicals of the original dirty dozen POPs (*e.g.*, polychlorinated biphenyls [PCBs] and organochlorine pesticides such as dichlorodiphenyltrichloroethane [DDT]), and new POPs such as some per- and polyfluoroalkyl substances (PFAS) (specifically, perfluorooctane sulfonic acid [PFOS], perfluorooctanoic acid [PFOA], perfluorohexane sulfonic acid [PFHxS]). Monitoring programs have also included various chemicals of emerging Arctic concern (CEACs), a term that summarizes compounds of concern in the Arctic that are not covered by the Stockholm Convention, such as non-regulated PFAS, emerging brominated flame retardants (EBFRs) and current-use pesticides.[4] The monitoring efforts also include mercury and trace elements in biota and thus support the United Nations' Minamata Convention on Mercury.

The Arctic is not only affected by long-range transported pollution; a wide range of pollutants may also be emitted locally, for example, from community solid waste sites, airports, sewage outflows, shipping, tourism, and resource extraction (*i.e.*, mining, oil and gas exploration).[5,6] These studies have shown that polycyclic aromatic hydrocarbons (PAHs) and other CEACs are emitted from a variety of local sources in addition to POPs from former uses. Legacy POPs, such as PCBs, have been found in high concentrations at former military installations, as well as in buildings and electrical installations in Arctic settlements.[7,8] The Arctic is experiencing fast socioeconomic development and industrialization, which will likely increase the number of local pollution sources and the variety of chemicals emitted.[5,9,10]

The ringed seal (*Pusa hispida*) is a key species for monitoring spatial and temporal trends of contaminants in the Arctic.[11–17] This long-lived species has an important role in marine food webs, is distributed across the Arctic and is of great cultural, economic, and nutritional importance for Arctic Indigenous Peoples in Canada and Greenland. Collaborations with northern and Indigenous communities, including hunters, have enabled the long-term collection of ringed seal samples at multiple locations within the Arctic.[18] The ringed seal is currently listed as a species of *least concern* by the International Union for Conservation of Nature in Greenland,[19] as *vulnerable* in Norway,[20] as *special concern* in Canada,[21] as *threatened* in the United States,[22] and is not listed in Russia.[21] Highly dependent on sea ice, ringed seals are impacted by climate-related change,[23,24] food web dynamics and disease spread.[21,22]

The objectives of this study were to (1) examine the current monitoring practices and the state of knowledge of pollutants in Arctic ringed seals from Canada, Greenland, Norway, Russia, and the United States (Alaska), (2) identify examples of local/regional source pollution in ringed seal contaminant levels, in the Arctic and beyond, (3) conduct a preliminary analysis of the influence from local Arctic contaminant sources using monitoring data in ringed seal from the Canadian Arctic, and (4) provide recommendations to better assess the potential influence of local sources in wildlife contaminant monitoring.

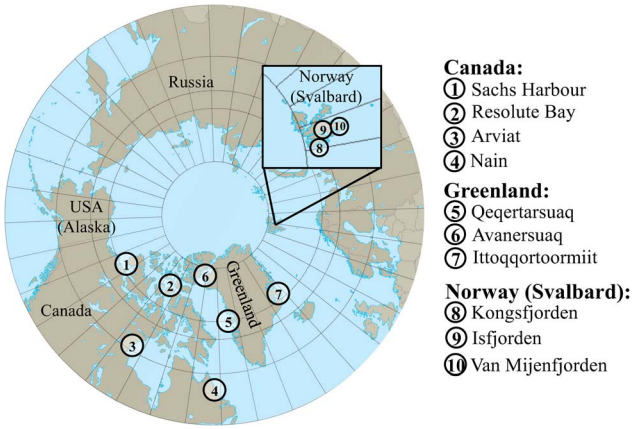

Fig. 1 Sampling locations for ringed seal long-term contaminant monitoring in Canada, Greenland (part of Kingdom of Denmark), and Norway (Svalbard).

Monitoring contaminants in Arctic ringed seals

Contaminants in ringed seals are currently studied in monitoring programs in Canada, Greenland and Norway, although the frequency varies. Long-term temporal trends have focused on POPs, additional unregulated PFAS, and mercury (Hg), but ringed seals from different regions have also been studied for the presence of additional trace elements and CEACs. In this study, we gathered information from all studies of contaminants in ringed seals by searching 'Web of Science', 'Scopus' and 'Google Scholar' with the following keywords: ringed seals, *Pusa hispida*/*Phoca hispida*, pinnipeds, contaminants, pollution, monitoring, Arctic, north, temporal contaminant trends. Manual checks of the results lead to a few additions of studies that were known to the authors and not identified in the literature search, for example referring to one-off screening studies outside the regular time trend monitoring.

In Canada, the Northern Contaminants Program was created in the 1990s to monitor pollutants in multiple Arctic wildlife species following concerns of northerners about the high levels of contaminants found in their traditional foods. Results have been published over time in a series of annual *Synopses of Research Reports* (2005–2017) and *Canadian Arctic Contaminant Assessment Reports* (1997–2017).[25] Contaminant monitoring in ringed seal has thus been on-going for more than 30 years across the Canadian Arctic in collaboration with Indigenous partners. The current monitoring sites include Sachs Harbour (Beaufort Sea), Resolute Bay (High Arctic), Arviat (Western

Hudson Bay) and Nain (Labrador) (Fig. 1). Through this program, temporal and spatial trends of multiple contaminants *e.g.*, PCBs, polybrominated diphenyl ethers (PBDEs), OCPs, PFAS, short-chain chlorinated paraffins (SCCPs), and trace elements, including mercury, have been reported (see Table 1 for review). Multiple articles have quantified these POPs and others (*e.g.*, polychlorinated dibenzodioxins/dibenzofurans, PCDD/Fs) in ringed seals from different sites through time.[26–37] Results for legacy POPs show that PCBs and OCPs are declining in concentrations,[38] but trends were generally found to be region and tissue-specific, while the feeding ecology of the seals, inferred from stable isotopes and fatty acids, was found to be closely linked to bioaccumulation of contaminants.[39,40] Moreover, CEACs such as substituted diphenylamine antioxidants and benzotriazole UV stabilizers,[41] EBFRs,[42] and plastics[43] have also been studied in ringed seals in the Canadian Arctic.

In Greenland, the AMAP Core Programme was formally established in 1994, following earlier sample collections of ringed seals that were also included in retrospective analyses.[69] The program is ongoing and includes sampling of ringed seals approximately every two years at three sites, Qeqertarsuaq (Disko Island, central West Greenland), Ittoqqortoormiit (central East Greenland), and Avanersuaq/Qaanaaq (Northwest Greenland) (Fig. 1 and Table 1). The samples are collected by local hunters.[125] Ringed seals from Qaanaaq are only analyzed for trace elements (mercury, cadmium, selenium), while specimens from the other locations are analyzed for lipophilic POPs (*e.g.*, PCBs; OCPs including DDTs, hexachlorobenzene [HCB], hexachlorocyclohexanes [HCHs], chlordanes [CHLs] and toxaphene; PBDEs; hexabromocyclododecane [HBCD]) in blubber as well as PFAS in liver. Regular screening of CEACs (*e.g.*, EBFRs, PFAS, phthalates, octachlorostyrene) and chemicals later regulated as POPs (*e.g.*, hexachlorobutadiene, SCCPs) in ringed seals has been included in the program.[70,79–82,126,127] Suspect and non-target screening has also recently been conducted to identify CEACs in ringed seal blubber and liver samples.[86] Retrospective studies that were not continued in the regular monitoring program included PCDDs/PCDFs, coplanar PCBs and endosulfan.[68,81] POP concentrations have been found to be generally higher in the Eastern region than in West Greenland.[69,72,77] Temporal trends of chemicals have been reported in several publications (Table 1). The results show that POP concentrations have generally been decreasing in ringed seals from Greenland,[17,83] whereas PFAS trends have been more variable.[59,77]

In Norway (Svalbard), PCBs, OCPs including DDTs, CHLs, HCB, toxaphene, and PBDEs have been monitored in ringed seals with relatively low frequency since the 1990s[103,128] (Fig. 1 and Table 1). Emerging brominated flame retardants and organophosphate esters (OPEs),[102,104,107] SCCPs and medium-chain chlorinated paraffins (MCCPs), PCB/PBDE metabolites,[100,101] toxaphene,[91,100] PFAS,[105,106] and trace elements[62,95] have been reported in seals collected at different sites (see ESI Fig. S1† for specific sampling locations).[103] Time trends for PFAS were reported for the period of 1990 to 2010,[105] while trends for POPs (CHLs; dichlorodiphenyldichloroethylene [DDE]; PCB-153; HCHs; HCB) have been reported under the environmental monitoring program for Svalbard and Jan Mayen (MOSJ).[128] Ringed seals from this Arctic region have been used as a reference population for studying biotransformation and effects of contaminants in highly exposed Baltic ringed seals.[129–136]

In the United States (Alaska) and Russia, concentrations of contaminants in ringed seals have been reported, however, there are currently no known long-term contaminant monitoring programs in ringed seals in these regions, although a national Russian Arctic contaminants program has been proposed.[137] Various studies have reported levels of PCBs, DDTs, CHLs, HCHs, HCB, PBDEs, PCDD/Fs, and trace elements in ringed seal tissues collected across different regions of the Russian Arctic.[117,118,120–124] Information for ringed seals from Alaska are mainly from the 1980s and 1990s and include assessments of PCBs, DDTs, CHLs, HCHs, dieldrin and multiple trace elements (*e.g.*, Hg and methylmercury; see Table 1).[108–113,115,138] Perfluoroalkyl carboxylic/sulfonic acids (PFCAs, PFSAs) and perfluorooctane sulfonamide (PFOSA) were also reported in seal liver collected in Alaska between 2003 and 2007.[116]

The circumpolar assessment of contaminants in ringed seals has enabled comparisons between regions and over time.[10,11,13,15,29] The long-term trends established by these Arctic monitoring programs are among the longest recorded for wildlife globally. These monitoring programs have also provided a platform for assessing time trends of new POPs and CEACs in seals. For instance, temporal analyses indicated that PBDEs increased significantly from 1998 to 2008 in ringed seals from East Baffin and decreased in recent years, which fluctuated slightly over time at several other locations.[42] Increasing trends of perfluorononanoic acid (PFNA) were also reported in ringed seals from East Greenland and Western Hudson Bay, whereas PFOS trends were highly variable.[59]

The time series of contaminants in ringed seals from the Canadian Arctic and Greenland have reached a time span that provides sufficient power for statistical analyses, for example studies on the influence of climate change on contaminant levels in ringed seals.[38,39,77,83] Seawater temperature, sea ice extent, and the Arctic Oscillation Index were found to be associated with POP concentrations in ringed seal in East and West Greenland.[83] Contaminant concentrations (including PCB and Hg) were also found to be correlated with environmental factors (air temperature, precipitation, sea ice coverage, oscillation indices) in seals in the Canadian Arctic.[38,39] These influences were still minor compared to the overall decrease of most POPs due to declining primary and secondary emissions[5] but might gain importance with the changing climate.

These long-term monitoring programs have mainly been designed to detect chemicals that are transported from industrialized mid-latitude locations to the Arctic *via* air, oceans, and rivers representing long-range environmental transport, together with the purpose of monitoring human exposure. As more information emerges on local pollution sources in the Arctic, such as local infrastructures and industrial activities,[6] and projected increases in human activity are foreseen in a warming Arctic,[10,139] local influences on wildlife exposure need

Table 1 Contaminants analyzed in tissues of ringed seals in the Arctic. Studies on time trends in ringed seals up to 2013 were previously compiled by Vorkamp and Muir[15]

Region/site	Chemicals[a]	Year of collection	Tissue	Reference
Canada				
Holman (now Ulukhaktok)	PCB, DDT	1972, 1981	Blubber	26
Resolute Bay, Grise Fiord, Admiralty Inlet	PCB, DDT, CHL, HCH, HCB	1972, 1975, 1976, 1983, 1984	Blubber, liver	44
Belcher Islands	PCB, DDT, CHL, HCH, dieldrin	1989	Blubber, kidney, liver, muscle	27
Sachs Harbour, Ulukhaktok, Paulatuk, Shingle Point, Salluit, Wakeham Bay, George River, Inukjuak, Resolute Bay, Sanikiluaq, Eureka	Total Hg, Cd, Pb, Se, Cu, Zn	1989–1994	Liver, kidney, muscle	45
Belcher Islands (Sanikiluaq)	PCB, DDT, CHL, HCH, dieldrin, mirex	1991	Blubber	28
Qikiqtarjuaq, Cambridge Bay, Coral Harbour, Kangiqsujuaq Kangiqsualujjuaq, Inukjuak, Pangnirtung, Rankin Inlet, Resolute Bay, Sachs Harbour, Talurjuaq, Tuktoyaktuk	PCB, DDT, HCH, CHL, HCB, toxaphene, dieldrin	1983–1989	Blubber	46
Ulukhaktok, Grise Fiord	PFCA, PFSA, FOSA	1998, 2001	Liver	47
Ulukhaktok	PCB, PCDD/F	1981, 1991, 1996, 2000	Blubber	29
Quaqtaq, Salluit, Kangirsuk	Toxaphene	1998	Blubber	48
Qikiqtarjuaq	PCB, DDT, CHL, HCH, HCB, dieldrin, mirex	2001	Blubber, liver, muscle	30
Resolute Bay, Arviat	PFCA, PFSA, 8:2/10:2 FTCA/FTUCA	1972, 1992, 1993, 1998, 2000, 2004, 2005	Liver	49
Inukjuak, Pangnirtung, Grise Fiord, Gjoa Haven, Pond Inlet, Arctic Bay, Sachs Harbor, Qikiqtarjuaq, Nain, Arviat, Resolute Bay	PFCA, PFSA, PFOSA, 8:2/10:2 FTUCA, 8:2 FTA	2002–2005	Liver	50
Sachs Harbour	PFCA, PFSA, 8:2/10:2 FTA, 8:2/10:2 FTUA, 7:3 FTA/FTUA	2004	Blubber, liver, blood	31
Ulukhaktok, Co-op Site, Woodward Point, Kuuk River	Hg	14th century–2003	Canine teeth	51
Ulukhaktok	HCH	1978–2006	Blubber	32

Table 1 (*Contd.*)

Region/site	Chemicals[a]	Year of collection	Tissue	Reference
Ulukhaktok	Total Hg	1973–2007	Muscle	33
Ulukhaktok	PCB, DDT, CHL, HCH, HCB, dieldrin, mirex, toxaphene	1993–2008	Blubber	52
Beaufort Sea, Lancaster Sound, East Baffin, Hudson Bay	Hg	1999–2009	Liver, muscle	53
Beaufort Sea, Lancaster Sound, East Baffin, Hudson Bay	PCB, DDT, HCB, CHL, HCB, HCH, toxaphene, PBDE, HBCD, PFCA, PFSA, PFOSA	1972–2010	Blubber, liver	54
Ulukhaktok	PCB, DDT, CHL, HCB, heptachlor epoxide, dieldrin, mirex, endrin	1972–2010	Blubber	34
Saglek Fjord	PCB, DDT, HCH, CHL, HCB, dieldrin	2008–2011	Blubber	35
Nachvak Fjord, Saglek Fjord, Okak Bay, Anaktalak Bay	PCB	2008	Blubber	36
Sachs Harbour, Ulukhaktok, Arviat, Inukjuaq, Grise Fiord, Arctic Bay, Resolute Bay, Pangnirtung, Gjoa Haven, Pond Inlet, Nachvak Fjord, Saglek Fjord, Okak Bay, Anaktalak Bay	Total Hg, Cd	2007–2011	Liver, muscle	37
Arviat, Inukjuaq, Resolute Bay, Grise Fiord, Arctic Bay, Sachs Harbour, Pangnirtung, Ungava, Qikiqtarjuaq, Nain, Okak, Saglek, Gjoa Haven	PBDE, EBFR, dechlorane plus	1998–2013	Blubber	42
Sachs Harbour, Ulukhaktok, Arviat, Kangiqsualujjuaq, Quaqtaq, Inukjuaq, Grise Fiord, Arctic Bay, Resolute Bay, Pangnirtung, Qikiqtarjuaq, Gjoa Haven, Makkovik, Nain	PCB, DDT, HCB, HCH, CHL, toxaphene	1972–2016	Blubber	38
Resolute Bay, Arviat, Sachs Harbour, Lake Melville	Substituted diphenylamine antioxidants, benzotriazole UV stabilizers	2016–2017	Liver	41
Ulukhaktok	PBDE	1981–2015	Blubber	55
	Total Hg, Se	1972–2017	Liver, muscle	39

Table 1 (*Contd.*)

Region/site	Chemicals[a]	Year of collection	Tissue	Reference
22 sites from Beaufort Sea, Central Arctic, Baffin Island, Hudson Bay, Ungava, Nunatsiavut				
Arviat	Total Hg	2003–2015	Muscle	56
Arviat	SCCP	2017	Blubber	57
Arviat, Nain, Resolute Bay, Sachs Harbour	SCCP	2016	Blubber	58
Arviat	PFOS, PFNA	1992–2020	Liver	59
Greenland				
Greenland	Total Hg	1984–1987	Liver, kidney, muscle	60
Greenland	Pb, Cd, Hg, Se	1978–1991	Liver, kidney, muscle	61
Avanersuaq, Danmarkshavn, Ittoqqortoormiit, Nanortalik, Upernavik, Uummannaq, Kong Oscars Fjord	Hg, Cd, Se	1978–1986	Liver, kidney, muscle	62
Ittoqqortoormiit, Avanersuaq, Qeqertarsuaq, Nanortalik	PCB, DDT, HCH, HCB, *trans*-nonachlor, toxaphene	1994	Blubber	63
Ittoqqortoormiit, Qeqertarsuaq, Avanersuq	PCB, DDT, CHL, HCH, HCB, Cd, Hg, Se	1999–2000	Blubber, liver, kidney, muscle	64
Ittoqqortoormiit	PCB, DDT, PBDE, CHL, HCH, HCB, toxaphene	2001	Blubber	65
Ittoqqortoormiit, Qeqertarsuaq	PFCA, PFOS, PFHxS, PFOSA	1982–2003	Liver	66
Ittoqqortoormiit, Qeqertarsuaq, Avanersuq	PFOA, PFOS, PFHxS, PFOSA	1998–2002	Liver	67
Ittoqqortoormiit	PCDD/PCDF, coplanar PCB	1986–2003	Blubber	68
Ittoqqortoormiit	PCB, PBDE	1986–2004	Blubber	69
Ittoqqortoormiit, Qeqertarsuaq	HBCD, TBBPA, methyl-TBBPA	2002	Blubber, liver	70
Ittoqqortoormiit, Qeqertarsuaq	HCH	1986–2006	Blubber	71
Qeqertarsuaq	PCB, DDT, PBDE, CHL, HCH, HCB, toxaphene	1982–2006	Blubber	72
Ittoqqortoormiit	PCB, PBDE, HBCD	1986–2008	Blubber	17

Table 1 (*Contd.*)

Region/site	Chemicals[a]	Year of collection	Tissue	Reference
Ittoqqortoormiit, Qeqertarsuaq, Avanersuq	Hg	1984–2010	Liver	73
Ittoqqortoormiit	PCN	1986, 2000, 2006	Blubber	74
Qaanaaq	PFCA, PFSA	1984, 1998, 2006	Liver	75
Ittoqqortoormiit	HBCD	1986–2010	Blubber	76
Ittoqqortoormiit, Qeqertarsuaq	PFCA, PFOS, PFHxS, PFOSA	1982–2010	Liver	77
Qeqertarsuaq	PCB-52, PCB-153, *p*,*p*′-DDE, HCB, HCH	1994–2010	Blubber	23
Ittoqqortoormiit, Qeqertarsuaq	Toxaphene	1986–2012	Blubber	78
Ittoqqortoormiit, Qeqertarsuaq	PCB, BDE-47, HBCD, EBFR, dechlorane plus	2012	Blubber	79
Ittoqqortoormiit	PFCA, PFSA, F-53B, FOSA	2011, 2012	Liver	80
Ittoqqortoormiit, Qeqertarsuaq	Endosulfan, SCCP, octachlorostyrene	1986–2014	Blubber	81
Ittoqqortoormiit, Qeqertarsuaq	HCBD	2014	Blubber	82
Ittoqqortoormiit, Qeqertarsuaq	PCB, DDT, HCB, α-HCH	1986–2016	Blubber	83
Qeqertarsuaq, Ittoqqortoormiit, Qaanaaq	SCCP	2016	Blubber	58
Avanersuaq/Qaanaaq	Hg, Se, Cd	2018	Liver	84
Qaanaaq, Qeqertarsuaq	Hg, Se, Cd	2008	Liver	85
Ittoqqortoormiit	Non-target screening	2018	Blubber, liver	86
Ittoqqortoormiit	PFOS, PFNA	1986–2021	Liver	59
Norway				
Spitsbergen, Svalbard	PCB, DDT, HCH, CHL, PCDD/F, HCB, dieldrin	1986	Blubber	87
Tempelfjorden and Kongsfjorden, Svalbard	PCB, DDE	1990	Blubber, kidney, liver	88
Northern coast of Norway	PCB, DDT, CHL, HCB, HCH, Hg, Cd	1987–1994	Blubber	89
Svalbard	Cd, Hg, Zn, Se	1978–1986	Liver, kidney, muscle	62
Kongsfjorden, Svalbard	PCB, toxaphene	1996	Blubber	90

Table 1 (*Contd.*)

Region/site	Chemicals[a]	Year of collection	Tissue	Reference
Kongsfjorden, Svalbard	Toxaphene	1992	Blubber	91
East of Svalbard (northern ice area)	PCB, DDT, HCH, CHL, HCB	1995	Blubber	92
Kongsfjorden, Svalbard	PCB, DDE	1992	Blubber	93
Kongsfjorden, Svalbard	PCB, DDT, CHL, HCH, HCB, Cd	1994, 1996	Blood	94
Kongsfjorden, Svalbard	Hg, Se, Cd, Pb	1996	Liver, kidney, muscle	95
Barents Sea	PCB, *p*,*p*′-DDE, HCH, CHL, HCB	1995	Blubber	96
Kongsfjorden, Svalbard	PCB, DDT	1996, 1998	Liver	97
Kongsfjorden, Svalbard	PCB, PBDE, HCB, CHL, DDE, toxaphene	1996, 2004	Blubber	98
Tempelfjorden and Van Mijenfjorden, Svalbard	PCB, OH-PCB, $MeSO_2$-PCB	2007	Liver, plasma	99
Tempelfjorden and Van Mijenfjorden, Svalbard	*p*,*p*′-DDE, $MeSO_2$-DDE, CHL, HCB, toxaphene	2007	Liver, plasma	100
Tempelfjorden and Van Mijenfjorden, Svalbard	PBDE, OH-PBDE	2007	Liver, plasma	101
Tempelfjorden, Svalbard	EBFR, PBDE	2007	Liver	102
Kongsfjorden, Svalbard	PFCA, PFSA, FTOH, 6:2 FTS, FTCA, FTUCA, EBFR, SCCP, MCCP	2010	Plasma	103
Kongsfjorden, Svalbard	OPE	2010	Blubber	104
Kongsfjorden, Tempelfjorden, Billefjorden, Van Mijenfjorden, Svalbard	PFCA, PFSA, FASA	1990–2010	Plasma	105
Tempelfjorden, Svalbard	PFOS, PFOA, F53, F53B	2007	Liver	106
Tempelfjorden, Yoldiabukta, Svalbard	EBFR	2014	Blubber	107
United States (Alaska)				
Barrow (now Utquigvik) and Nome	PCB, DDT, HCB, γ-HCH, CHL, heptachlor epoxide, dieldrin	1988–1989	Liver, kidney, blubber	108
Bering Sea	PCB, DDT, CHL	1989–1995	Blubber	109
Barrow, Nome	PCB, DDT, HCB, γ-HCH, CHL, dieldrin, heptachlor epoxide, 35 elements	1988, 1991	Liver, kidney, blubber	110

Table 1 (*Contd.*)

Region/site	Chemicals[a]	Year of collection	Tissue	Reference
Barrow, Nome	36 elements, methylmercury	1988–1989	Liver, kidney	111
Barrow, Nome	DDT, PCB, HCB, HCH, CHL, mirex, toxaphene, dieldrin, As, Cd, Cu, Hg, Se, Ag, V, Zn	1988–1993	Liver	112
Barrow, Nome	PCB, DDT, HCB, HCH, CHL, toxaphene, heptachlor epoxide, dieldrin, 37 elements	1988–1993	Liver, kidney, blubber	113
Barrow	PCB, DDT, HCH, CHL, HCB	1996	Blubber	114
Barrow, Nuiqsut, Point Lay	HCB, CHL, DDT, HCH, PCB	1997–1999	Blubber	115
Point Hope, Shishmaref, Little Diomede, Hooper Bay	PFCA, PFSA, PFOSA	2003–2007	Liver	116
Russia				
Dikson	DDT, PCB, CHL, HCH, HCB	1995	Blubber	117
Sorocskaya Guba Bay, Lake Lagoda, Lake Saimaa	HCB, PCB, DDT, HCH	1988–1995	Blubber	118
Dvina Bay, Onega Bay	HCH, CHL, DDT, HCB, PCB	1998, 2001	Blubber	119
Lavrentiya	PCB, PCDD/F	2002	Blubber	120
Lavrentiya Bay	HCH, DDT, PBDE, CHL, HCB, PCB, toxaphene, Hg, Pb, Cd	2000, 2002	Blubber, kidney, liver, muscle	121
Kalgalaksha Bay, Vaygach Island, Dikson Island, Vankarem	DDT, PCB, PBDE, PCDD/F, HCH, CHL, HCB, mirex, methoxychlor, endrin, toxaphene	2001–2005	Blubber	122
Enmelen, Sireniki	DDT, HCH, HCB, CHL, PCB, mirex	2016	Blubber, muscle	123
Enmelen, Sireniki	Pb, As, Cd, Hg, Cu, Zn, Ni, Cr, Al, Mn, Ba, Sr, Co, V, Be, Mo, Sn, Sb	2016	Blubber, muscle	124

[a] PCB: polychlorinated biphenyls; DDT: dichlorodiphenyltrichloroethane; DDE: dichlorodiphenyldichloroethylene; HCH: hexachlorocyclohexane; HCB: hexachlorobenzene; HCBD: hexachlorobutadiene; CHL: chlordanes; PCDD/F: polychlorinated dibenzodioxins/dibenzofurans; HBCD: hexabromocyclododecane; TBBPA: tetrabromobisphenol A; PCN: polychlorinated naphthalenes; EBFR: emerging brominated flame retardants; PBDE: polybrominated diphenyl ethers; OH-PCB/PBDE: hydroxylated metabolites of PCBs and PBDEs, respectively; $MeSO_2$-PBDE: methylsufonyl metabolite of PBDEs; SCCP: short-chain chlorinated paraffins; MCCP: medium-chain chlorinated paraffins; UV: ultra-violet; PFCA: perfluoroalkyl carboxylic acids; PFSA: perfluoroalkyl sulfonic acids; PFOS: perfluorooctane sulfonic acid; PFOA: perfluorooctanoic acid; PFNA: perfluorononanoic acid; FASA: perfluoroalkane sulfonamide; PFHxS: perfluorohexane sulfonic acid; F53: polyfluorinated ether sulfonate; F53B: chlorinated polyfluorinated ether sulfonate; FTOH: fluorotelomer alcohols; FTA: fluorotelomer saturated acid; FTUA: fluorotelomer unsaturated acid; FTCA: fluorotelomer carboxylic acids; FOSA: heptadecafluorooctane sulfonamide; PFOSA: perfluorooctane sulfonamide; Al: aluminium; Ag: silver; As: arsenic, Ba: barium; Be: beryllium; Cd: cadmium; Co: cobalt; Cr: chromium; Cu: copper; Hg: mercury; Ni: nickel; Mn: manganese; Mo: molybdenum; Pb: lead; Sr: strontium; Sn: tin; Sb: antimony; Se: selenium; V: vanadium; Zn: zinc.

to be considered. Furthermore, screening efforts for CEACs are increasing and include current-used chemicals for which emission sources are more difficult to identify. Local sources of CEACs in the Arctic might be associated with ongoing use (*e.g.*, in consumer products, building materials, vehicles), rendering them difficult to control. This raises the question of the potential influence of chemicals emitted from Arctic local sources on ringed seal exposure and consequences for interpretation of findings from ongoing spatial and temporal trend monitoring studies.

Examples of known local or regional sources of contamination in seals

Contaminants in ringed seals have also been analyzed outside the Arctic where the environment is impacted from both local sources and long-range transported pollutants. One example is the Baltic Sea where ringed seal tissues have been analyzed to study time trends of contaminants and their effects.[133] This semi-enclosed brackish water environment receives pollution from inland activities located in the vast catchment area of nine heavily populated countries, including municipal, agricultural, and industrial sources.[140–142] In addition, ship traffic, fisheries and tourism may lead to direct emissions to the sea that also receives contaminants from the deposition of airborne pollutants. Given these aggregated pollution sources, the Baltic Sea is less suitable than the Arctic to reflect the long-range environmental transport of chemicals that is assessed for the Stockholm Convention. Consequently, the research and monitoring of contaminants in ringed seals of the Baltic Sea have not distinguished between different types of sources. Starting from the late 1960s, high levels of legacy contaminants have been reported in ringed seals from the Baltic Sea[143,144] and the contaminant exposure has been associated with effects on reproduction, immune, and endocrine functions as well as population declines.[129,133,134,136,145–148] The Baltic Marine Environment Protection Commission, formerly Helsinki Commission (HELCOM), was established in 1974 to protect the marine environment of the Baltic Sea from all sources of pollution. Time trends indicated that some chemical groups such as PBDEs, PCBs, DDTs, and PCDD/Fs have declined since the 1970s in ringed seal tissues, whereas other chemicals (brominated dioxins and furans) remained stable.[97,149]

Ringed seals potentially exposed to locally emitted contaminants were studied at Saglek Bay (Nunatsiavut, northern Labrador, Canada), where a former military radar station with PCB contamination was located. Although PCB contaminated soils were removed in the 1990s and early 2000, the adjacent bay had become contaminated with PCBs, leading to elevated PCB levels in sediments, invertebrates, fish, seabirds, and ringed seals.[6,7] Unique PCB profiles (high proportions of highly chlorinated congeners) and high PCB concentrations were found in ringed seal tissues compared to other Canadian locations.[35] Moreover, stable isotopes of nitrogen and carbon, fatty acid, and satellite telemetry data indicated that the home range/habitat use of seals and time spent in the inlets were important determinants of their PCB concentrations, indicating local contamination.[36,150]

Local sources of pollution, along with long-range pollution, were suggested to contribute to plasma PFAS concentrations and blubber PBDE concentrations in another ice-associated seal species, the Weddell seal (*Leptonychotes weddellii*) of the Antarctic.[151,152] These seals were sampled close to the permanent research stations of the United States, New Zealand, and Italy. The local source hypothesis was supported by geographical differences in PFAS levels in seawater indicating potential point sources of contamination[153] as well as PCB, PBDE, and trace element concentrations reported at high levels in wastewater sludge, sediment, invertebrates, and/or fish around the American station.[154–156] These studies also reported a rapid decrease in concentrations with increasing distance from the point source. Together these examples suggest that local sources might influence the levels and patterns of contaminants in ringed seals and related species, but the information is limited to a handful of studies with only one on Arctic seal populations.

Case study: the influence of local anthropogenic sources on contaminant concentrations in ringed seals analyzed under the Canadian monitoring program

To increase our understanding of the potential influence of local anthropogenic sources on the ringed seal contaminant exposure in the Arctic, we conducted an exploratory pilot study based on data from the Canadian Northern Contaminants Program. Canadian ringed seal samples are typically collected near communities where local pollution sources might exist. Specific anthropogenic local pollution sources have not been formally studied in the four communities with ongoing ringed seal monitoring across the Canadian Arctic (*i.e.*, Sachs Harbour, Northwest Territories; Resolute Bay and Arviat, Nunavut; Nain, Labrador). For this case study, we incorporated information on human population sizes, community infrastructures, mines, airports, military sites, and industries that have been identified as potential local sources of pollution in other studies[6,157,158] (Tables 2 and S1†). However, limited data existed on contaminant emissions for locations included in those studies. A distance-based redundancy analysis (db-RDA) with forward selection of variables and variation partitioning was performed to analyze this environmental dataset. Constrained ordination is a multivariate regression that helps visualize the relationship between response variables (herein the suite of contaminants in seal tissues) and a set of explanatory variables (herein the potential local source pollution, seal biological data, and study site location). In turn, variation partitioning teases apart the unique *versus* combined effect of explanatory variable bundles represented *via* Venn diagrams. Several studies have applied redundancy analyses to link contaminant profiles to environmental variables (*e.g.*, water chemistry, habitat characteristics, biological factors) in fish, invertebrates, and marine mammals.[159–161] Here, we build on this

Table 2 Summary of the observation- and community-level variables used in the distance-based redundancy analysis and variation partitioning, including their units of measurement and relevant references. Additional references can be found in Table S1

Level	Variable	Units/scale	Reference
Observation-level	Age	Years	Ref. 39 and Houde *et al.* unpublished
	Sex	Categorical (male/female)	
	Size (maximum girth)	cm	
	Carbon isotope (δ^{13}C)	‰	
	Nitrogen isotope (δ^{15}N)	‰	
	Sampling year	Year	
Community-level	Sampling location	Categorical (Sachs Harbour, Resolute Bay, Arviat, Nain)	Ref. 39 and Houde *et al.* unpublished
	Human population	Human population size in each settlement	NASA Socioeconomic Data and Applications Center (SEDAC) population estimator[162]
	Mines	Distance to closest active mine, km	Minerals and mining database (Natural Resource Canada; **https://natural-resources.canada.ca/our-natural-resources/minerals-mining/10858**)
	Power source	MW h per year	Remote communities energy database (Natural Resource Canada; **https://open.canada.ca/data/en/dataset/0e76433c-7aeb-46dc-a019-11db10ee28dd**)
	Airports	Number of flights per year	Flightradar24 **https://www.flightradar24.com**
	Military site	Distance to nearest Distant Early Warning (DEW) line site, km	GoogleEarth for distance measurements
	Solid waste	Distance from landfill to shoreline, km	GoogleEarth for distance measurements
	Wastewater	m^3 per day	Nunavut, Labrador, and Northwest Territories water board issue permits

previous experience by testing the additional influence of local point source contamination in Arctic wildlife.

A total of five db-RDAs were realized, with each focusing on a different contaminant class as the response matrix: trace elements (liver), PCB homolog groups (blubber), individual PBDEs (blubber), OCPs (blubber), and PFAS (liver) measured in ringed seals from the four Canadian locations between 2008 and 2022 (Fig. 1). Specific chemicals included in each contaminant class are detailed in Table S2.† For this study, airports, wastewater facilities, solid waste disposal sites, mines, and military sites identified in and around communities where seal sampling occurred were considered as local source variables. Source of energy and human population size through time were also considered as local source variables. The information was collected from available public sources (Tables 2 and S1†). For most of the local source variables, obtaining specific information (*e.g.*, volume of solid waste produced each year) and quantifying certain variables (*e.g.*, emission from airports) proved challenging due to the lack of available data. In cases where data were only available for a single point in time (*e.g.*, power use, flow of wastewater), this static value was used for all years for that community. Due to this limitation, we acknowledge that these variables will not fully capture temporal variability of contaminants emitted from point sources. By their nature, the local source variables may correlate strongly with the location effect. Biological characteristics of the ringed seals such as the age of the animal, sex, and maximum length were included in the analyses as they are known to influence contaminant variation in ringed seals.[38,42] Carbon (δ^{13}C) and nitrogen (δ^{15}N) bulk stable isotopes were also included as these variables are important indicators of trophic level.[24] Detailed methods for obtaining each biological variable are provided in Houde *et al.*[38] Lastly, location (community) and sampling years were also included as explanatory variables.

All statistical analyses were conducted in R version 4.3.1. Prior to performing the db-RDAs, contaminant data were transformed using the *decostand* function from the *vegan* package in R[163] to select the best transformation among chord, Hellinger, and log-chord. The transformation that provided the highest R^2 value was retained. Multicollinearity among explanatory variables was assessed using Variance Inflation Factor (VIF). The human population size variable exhibited high collinearity and was removed to ensure better model interpretation for all db-RDAs. Using the *ordistep* function, a forward selection approach using bundles (local source bundle: mine, power source, airports, military sites (Distant Early Warning [DEW] line sites), wastewater, solid waste; biological variable bundle: seal age, sex, size, δ^{13}C, δ^{15}N) was then conducted to ensure the inclusion of local source variables in the model while also reducing redundancy. The forward selected variables,

along with sampling year and location, were incorporated into the db-RDA models using the *capscale* function in the *vegan* package with Euclidean distance.[163] To identify the relative contribution of local source contamination on the observed contaminant variation, variance partitioning using all the explanatory variables was run with the *varpart* function. This approach allowed for a direct comparison between Venn diagrams and ensured that all predefined variable bundles (*e.g.*, local source, biological, location and sampling year) had representation in the diagrams. All local source contamination variables were considered community-level variables as they were spatially structured factors tied to community-based characteristics. This contrasts with observation-level variables that were specific to individual seals.

Case study results and discussion

The forward selected db-RDA results showed that seal biological data, community location, local source variables and sampling year explained a considerable portion of the ringed seal contaminant variation. These models explained 41.0% of the variation in trace element concentrations in ringed seals (R^2 adjusted for number of variables and sample size), 45.1% for PFAS levels, 8.4% for PBDEs, 31.8% for OCPs, and 65.8% for PCBs. Results of forward selected variables included in each db-

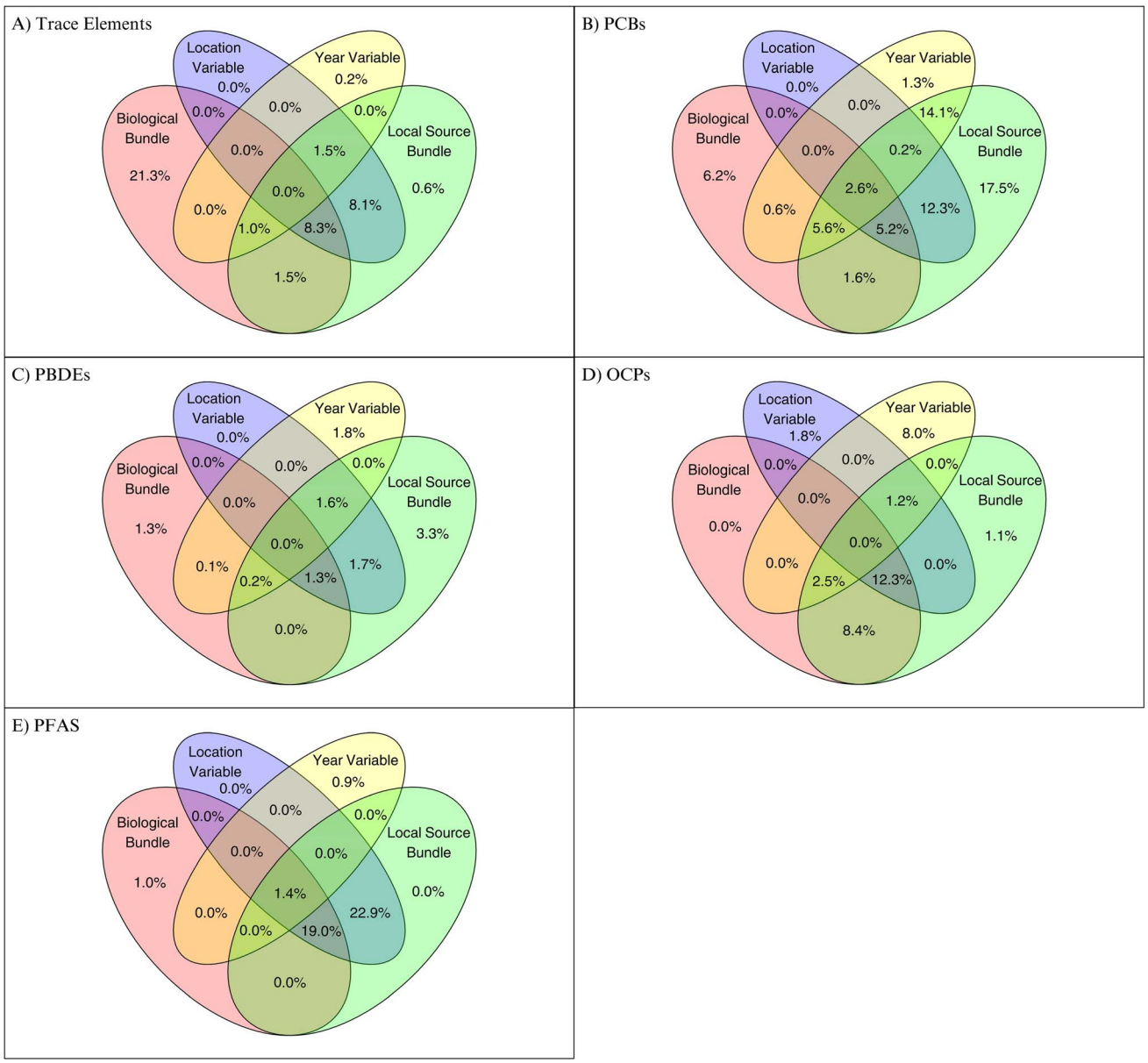

Fig. 2 Venn diagrams of the variation partitioning of the contaminant data explained by biological variable bundle (age, sex, $\delta^{13}C$, $\delta^{15}N$ and size) (red), sampling location (blue), sampling year (yellow) and local source variable bundle (human population size, power source, mine, wastewater facility, solid waste disposal, DEW line sites, airports) (green). Values (%) represent the contribution of explained contamination variation by each variable/bundle. Size of circles not representative of contribution.

RDA can be found in Table S3.† The variation partitioning (Fig. 2) showed that the local source variables uniquely explained none of the variation in PFAS concentrations, 0.6% in trace elements, 3.3% in PBDEs, 1.1% in OCPs, and 17.5% in PCBs. The partition of variation uniquely explained by location variables was consistently 0% for all contaminant groups other than OCPs (1.8%) (Fig. 2). It is challenging to separate the effect of community-level source variables from the geographical position of the community and, as expected, the contribution of the location variable overlapped with local contaminant source variables (0–22.9% co-explained). Generally, the biological variable bundle and the sampling year represented large unique contributions to the explained variation for all contaminant groups. These results are expected as biological variables are known to be closely related to contaminant concentrations in seals *via* diet differences, age, sex, and body condition of the animal,[13] and sampling year captures temporal changes in ringed seal contaminant levels likely related to changes in emissions (*e.g.*, as a consequence of chemical regulation) and environmental changes (*e.g.*, ice cover, oscillation patterns).[38]

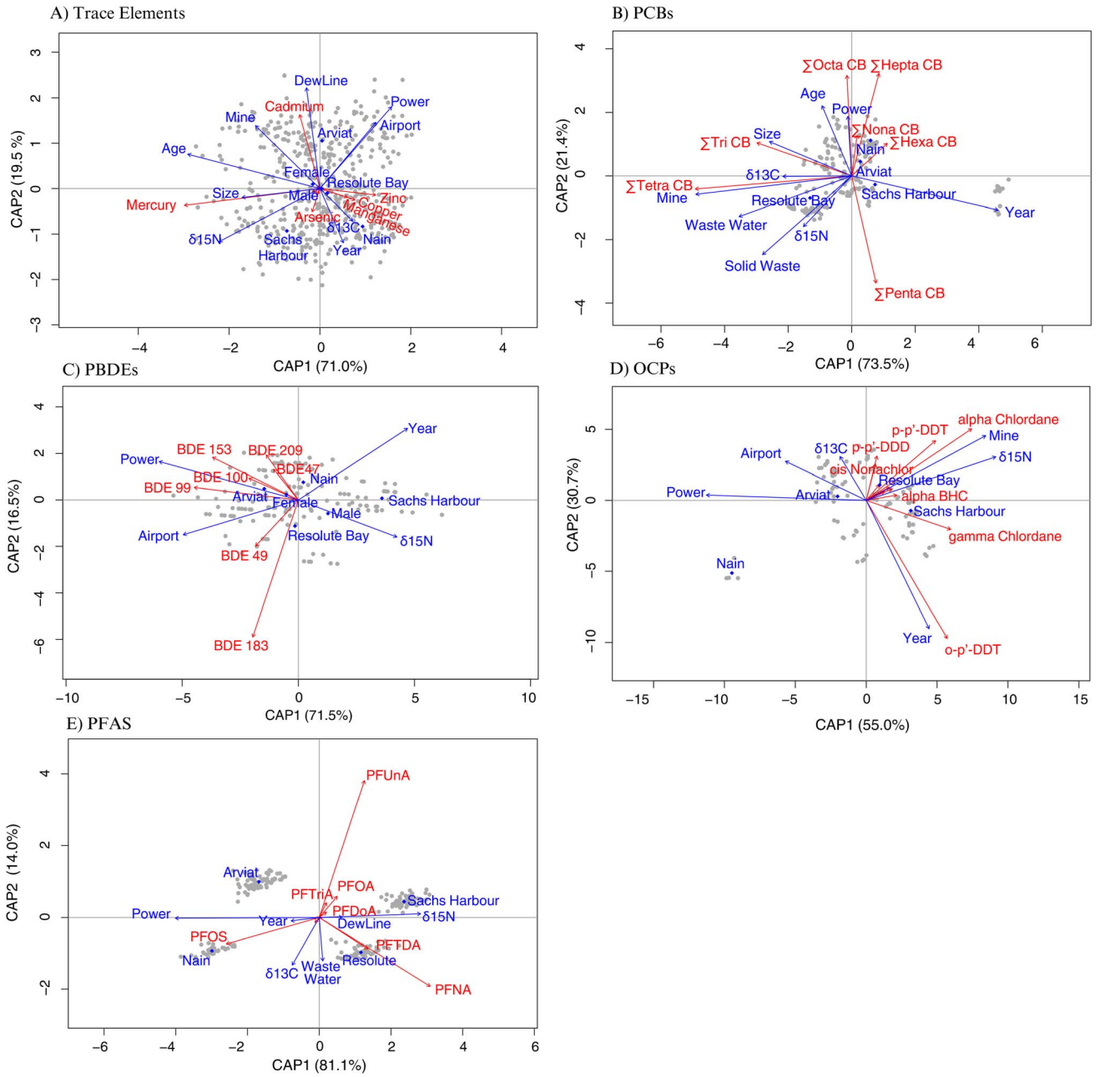

Fig. 3 Distance-based redundancy triplots illustrating the relationships between the response, in red, of (A) trace elements, (B) PCB concentrations (summed by homologue groups), (C) PBDEs, (D) OCPs, and (E) PFAS in ringed seals. In blue: a subset of explanatory variables based on forward selection (power source, wastewater facility, solid waste disposal, proximity to mine and DEW line sites, number of flights at airports, location, age, sex, year of capture, $\delta^{13}C$, $\delta^{15}N$ and size of seals). Direction and length of arrows represent variation contribution to contaminant concentrations. Canonical axes (CAP) represent constrained ordination axes. Ringed seal individuals represented in grey points.

Nonetheless, it is interesting to note that a large part of the location, local source, and biological effects could not be distinguished as 1.3–19.0% of the variation in seal contamination was co-explained by all three bundles. While the local source bundles did not show strong unique contributions on their own, their combined influence with the other variables is evident.

For the trace element db-RDA, distance from the mine and distance to military sites (DEW line site) showed negative relationships with copper and manganese, suggesting that the closer these infrastructures/sites are, the higher the copper and manganese concentrations in liver of ringed seals (Fig. 3A). Distance to military sites (DEW line sites) showed a positive relationship with cadmium suggesting that the further the military site, the higher the cadmium concentrations. While these relationships are unusual, a combination of factors not assessed in this study, including regional land cover, geochemical composition and permafrost thaw could potentially explain these findings.[164,165] As seen in Fig. 2A and 3A, biological variables (*e.g.* seal size) also play a substantial role in explaining trace metal variation. For PCBs, the db-RDA indicated numerous relationships between the PCB homolog groups and local source variables (Fig. 3B). Community power consumption showed positive relationships with the octa, hepta, and nona PCB homolog groups. The tetra PCB homolog group showed a positive relationship with distance to mine. Out of all db-RDA models, PCBs showed the highest explained variance and a large shared and unique contribution of the bundle of the local sources in the variance partitioning. PCBs were widely used in infrastructure such as electrical transformers, capacitors, and building materials prior to their ban and these results might be a reflection of PCB from older infrastructures although actual measurements are limited. PCBs were detected in soils near the Resolute Bay airport and studies of DEW line sites have shown evidence of the spread of PCBs from the radar stations to the surrounding region.[166–168]

Fire training sites, airports, landfills, wastewater discharges, and consumer goods have been identified as typical local sources of PFAS in the Arctic (reviewed by Lohmann *et al.*[59]). Our results for PFAS indicated that individual seals analyzed through the monitoring program were grouped strongly by location indicating geographic influence on contamination patterns (Fig. 3E). However, variance partitioning revealed that location itself did not explain unique variation rather it was shared with the biological and local source bundles (Fig. 2E). These results suggest that while geographical differences influence PFAS variation, likely related to source regions and long-range transported patterns (*e.g.*, sources in Asia affecting the western and High Arctic *vs.* North America and Europe affecting the eastern part of Canada). Besides geographical differences related to long-range transport, previous studies have documented PFAS contamination near Arctic airports (Resolute Bay, Canada, and Svalbard, Norway) further supporting the role of local source influences.[168,169]

Although the variation explained for PBDEs was small, some weak relationships could be observed. Power source showed positive relationships with PBDE-47, -99, -100, -153, and -209, suggesting that an increase in annual power usage could have influenced concentrations of these congeners in ringed seal blubber (Fig. 3C). Even though the production and use of PBDEs have been phased out since the mid-2000s in North America and Europe, older products and infrastructure may still contain PBDEs from before the bans.[170] For OCPs, the db-RDA showed a positive relationship between α-chlordane and distance to mine and a negative relationship between number of flights and γ-chlordane (Fig. 3D). In the variance partitioning, the local source bundle co-explains the majority of the OCP variance with the biological bundle and location variable, suggesting that a combination of geographical factors and seal characteristics are influencing the relationships seen in the db-RDA. As agricultural activities are limited or non-existent in the Canadian Arctic, the unique local influence for OCPs is likely small.

The persistence of the regulated contaminants studied here, along with the potential influence of historical contamination and secondary emissions, makes it difficult to pinpoint with certainty if relationships observed through the db-RDA analyses are solely due to local source influences. Our results indicate that it is likely a combination of factors that influence POP concentrations in ringed seals. Conducting the same analyses with non-regulated chemicals of current concern (*e.g.* plastic related compounds, EBFRs, emerging PFAS) would be interesting to do, however, long-term trends for these compounds are generally not available and their levels in seals are usually lower than legacy contaminants. Overall, results from this pilot study suggest that human-associated Arctic point sources may contribute to some degree to the contaminant exposure of ringed seals, but that this influence is likely small and difficult to distinguish from biological and temporal variables and geographical differences due to long-range transport.

Challenges in estimating the contribution of local sources to contaminants in ringed seals

Specific source contributions may be difficult to assess for contaminants in high trophic level marine species due to their typically large-scale spatial movements and their foraging ecology across different food webs.[171,172] Some ringed seals from different parts of the Arctic have been reported to travel over large distances (>1000 km) at least seasonally or during certain life stages.[173–175]

Ringed seal samples intended for contaminant monitoring are often collected at significant distances from hamlets to avoid the influence of local sources. For example, none of the ringed seals from Svalbard were collected close to human settlements. Although local pollutant sources have been described for Svalbard,[8,169,176] their contribution to pollutant exposure in the environment remains at a small spatial scale (<20 km)[177] and thus their influence on pollutant exposure for species that move over large spatial scales, like ringed seals, is difficult to assess. Spatial differences may also be related to local oceanographic conditions as suggested for differences in PFAS concentrations observed in ringed seals sampled in different fjords from the western coast of Svalbard.[105] In

Greenland, the monitoring sites are located far (>200 km) from active and major legacy mine sites[178] and other industrial point sources. Local settlements adjacent to the monitoring sites are comprised of small communities of <1000 residents. Therefore, the influence of local point sources is considered low but cannot be ruled out and should be assessed on a compound-specific basis. Previous studies from Greenland showed indications of influences of community size on levels of PCBs and PBDEs in shorthorn sculpin (*Myoxocephalus scorpius*), whereas no difference was seen for OCPs in the same samples.[179] Potential local contaminant sources at the Greenland monitoring sites include sewage and open waste burning from waste dumps in the settlements, release of effluents and spills from harbors as well as shipping (including cargo ships, cruise ships, and fishing vessels), but no detailed information is available for a quantitative assessment. In Canada, local inputs of PCBs from DEW line sites were reported for soil,[166,167] and human activities in and around Iqaluit, Nunavut were found to be a source of PCBs, Hg, PAHs, and PFAS in sediment from Frobisher Bay.[180] PCB and DDT levels in seawater collected around Cambridge Bay, Nunavut also suggested local inputs from the town and military site.[181,182] Chemical concentrations found in water and sediment may distinctly represent local source signals as compared to pinnipeds, that accumulate contaminants over a long-life span and integrate exposure from different sources including different prey species.

Recommendations to better assess the potential influence of local sources in wildlife contaminant monitoring

Long-term monitoring of contaminants in marine mammals such as ringed seals has important applications in the protection of Arctic ecosystems and human health as it integrates temporal changes in water quality and ecosystem state, provides information on contaminant exposure from traditional foods to local population, identifies emerging issues, and supports the evaluation of the effectiveness of mitigation of recognized pollutants. The question whether the measurements are influenced by emissions from local sources is important regarding the assessment of long-range environmental transport under the Stockholm Convention. Furthermore, a general understanding of contaminant transport and accumulation in Arctic ecosystems is needed for efficient regulatory action.

One key aspect in understanding the potential influence of local sources in remote regions is the knowledge of the history of the site/community and its surroundings, as compiled in part for this study. Past and present potential local/regional sources of contamination should be identified. These can include industrial facilities (*e.g.*, mines), military sites, transportation (*e.g.*, airport, port), power/heating systems, wastewater treatment, waste disposal/landfills, livestock, and urban runoff. Knowledge is needed of which substances are actually emitted from these sources. Secondary sources of contamination in the area should also be noted. For example, permafrost thawing and glacier melting sites can potentially act as secondary sources for certain contaminants.[183–186] Maps indicating the location of pollution sources should be created with this information and, ultimately, modeling and machine learning can be used to track sources of pollution.[187,188] Integration of hydrological and air modeling could also help predict the dispersion patterns of contaminants around local sources and help identify sites for sample collection.

Ringed seals or other marine mammals may not be the perfect indicator species for monitoring local contamination sources. However, if the primary purpose of the monitoring campaign is human contaminant exposure, local sources should be considered as a representation of the real-life situation, including all possible dietary exposure pathways. It will also be relevant to be able to identify pollution source mitigation measures as different environmental management agencies will likely be responsible for actions at local, regional and international levels.

Although POPs, such as PCBs and OCPs, have been banned for up to several decades, their monitoring is still crucial, for several reasons. The effectiveness of regulations should be ensured, as reflected in decreasing concentrations in Arctic ringed seals. Furthermore, despite decreasing concentrations, some POPs, such as PCBs, are still at levels in wildlife and humans that cause health concerns and require attention.[189] Other compounds, such as long-chain PFCAs are increasing in some Arctic wildlife populations and cause co-exposure situations that are far from understood.[59] Climate change affects the environmental distribution of POPs and/or ecosystem structures, which is captured in an integrated way in the contaminant time series.[190] In addition to POP and heavy metal monitoring, ringed seals also have been important species for screening studies and should be considered for further surveillance of CEACs.[4]

Higher resolution analyses of local sources might identify specific compounds that are relevant to monitor locally, in connection with specific sources, such as wastewater emissions or landfill leachates. In addition to collecting samples from wildlife for contaminant monitoring, abiotic (air, water, sediment) and other biotic (invertebrates, fish) samples around the northern communities and from the species habitat and food web should be collected. However, these collections are not always simple due to the remote locations and harsh environmental conditions. This highlights the importance of community engagement and collaboration with Indigenous Arctic Peoples.[18,191] Local communities have information about potential sources of contamination as well as valuable insights and knowledge about local environmental issues, and know when and where to collect samples. Local knowledge can also help inform the movement of animals and support interpretation of contaminant data. When resources and expertise are available, satellite tracking devices and photography (*e.g.* camera trap sampling stations) can also be used to investigate the movement ecology of the studied wildlife and links to potential contaminant exposure.[192] Other techniques such as compound specific isotopic composition and chemical fingerprinting can also be of interest to help identify specific sources of contamination.

Finally, the monitoring of contaminants in Arctic wildlife requires balancing the ongoing measurements at the same sites

over time with adjustment based on the emergence of new contaminants and evolving local conditions. Contaminant monitoring should also consider any changes in the species that are being harvested by local people, such that contaminant data are most relevant for communities.

Conclusion

Arctic monitoring programs including ringed seals (and/or other marine mammals) have been designed to monitor concentrations of long-range transported chemicals over time and generate information on exposure of local human populations *via* their traditional diet. As Arctic data are often used to assess long-range environmental transport, it is important to be able to relate measurements in the Arctic to local or distant sources. Knowledge of sources is also important to enable mitigation actions *via* the competent authorities at the local, regional or international level. This study of contaminants in ringed seals from the Arctic highlights the valuable knowledge obtained from long-term monitoring of contaminants. Our preliminary analysis using monitoring data from the Canadian Arctic detected limited effects of local sources of contamination accumulation in seals. However, it should be noted that our analysis was biased towards chemicals which are banned globally and no longer included in products of current use. As climate change continues and as Arctic communities continue to change in size, infrastructure, and tourist and industrial activity, among other factors, the relative importance of local *versus* distant contaminant sources affecting wildlife and country foods may change in the future. The assessment of local emissions of contaminants within the Arctic becomes therefore more critical and study designs should consider these developments.

Data availability

The list of potential local sources of contamination can be found in the ESI.† Contaminants data from Norway are available on the Environmental monitoring of Svalbard and Jan Mayen website (**https://mosj.no/en/indikator/influence/pollution/pollutants-in-ringed-seals/**). Metadata from Canada can be found on the Polar Data Catalogue (**https://www.polardata.ca/**) and the Environment and Climate Change open government portal (**https://open.canada.ca/en**). Greenland biota contaminant data can be accessed at the International Council for the Exploration of the Sea (ICES) Data Centre *via* the DOME database (**https://dome.ices.dk**). Some temporal trend information can be obtained from the Arctic Monitoring Assessment Programme (AMAP) assessment data portal (**https://www.amap.no/ahat**). Additional data can be available on request.

Author contributions

M. Houde – conceptualization, investigation, writing – original draft, writing – review & editing, project administration. N. Facciola – data curation, methodology, formal analysis, writing – original draft, writing – review & editing. H. Routti – data curation, writing – original draft, writing – review & editing. K. Vorkamp – investigation, data curation, writing – original draft, writing – review & editing. J. Søndergaard – data curation, writing – review & editing, validation, project administration. D. C. G. Muir – writing – review & editing, validation.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

The monitoring of contaminants in Arctic ringed seal could not be done through time without the collaboration of northern and Indigenous communities and hunters. We are very grateful for their knowledge and expertise that are the basis of this long-term work. In Canada, the Nunatsiavut Government and the Hunters and Trappers organizations of Sachs Harbour, Arviat and Resolute are acknowledged for their co-leadership in ringed seal contaminants monitoring. Zofia Taranu from Environment and Climate Change Canada is thanked for the help with statistics and her review of the manuscript as well as Cynthia de Wit (Stockholm University) and Melissa McKinney (McGill University) for their input on the text.

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