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Climate Variability and Temporal Trends of Persistent Organic Pollutants in the Arctic: A Study of Glaucous Gulls

JAN O. BUSTNES,**,†
GEIR W. GABRIELSEN,* AND
JONATHAN VERREAULT§

Norwegian Institute for Nature Research, The Polar Environmental Centre, NO-9296 Tromsø, Norway, Norwegian Polar Institute, The Polar Environmental Centre, NO-9296 Tromsø, Norway, and Département des sciences biologiques, Université du Québec à Montréal, C. P. 8888, Succ. Centre-ville, Montréal, Québec H3C 3P8, Canada

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The impact of climate variability on temporal trends (1997—2006) of persistent organic pollutants (POPs; polychlorinated biphenyls [PCB], hexachlorobenzene [HCB], and oxychlordane) was assessed in glaucous gulls (Larus hyperboreus) breeding in the Norwegian Arctic (n = 240). The Arctic Oscillation (AO: an index of sea-level pressure variability in the Northern Hemisphere above 20°N) with different time lags was used as a climate proxy. The estimated concentrations of POPs in glaucous gull blood/plasma declined substantially (16—60%) over the time period. Multiple regression analyses showed that the rates of decline for POPs were correlated to climate variation when controlling for potential confounding variables (sex and body condition). More specifically A0 in the current winter showed negative associations with POP concentrations, whereas the relationships with AO measurements from the year preceding POP measurements (AO preceding summer and A0 preceding winter) were positive. Hence, gulls had relatively higher POP concentrations in breeding seasons following years with high air transport toward the Arctic. Furthermore, the impact of AO appeared to be stronger for HCB, a relatively volatile compound with high transport potential, compared to heavy chlorinated PCB congeners. This study thus suggests that predicted climate change should be considered in assessments of future temporal trends of POPs in Arctic wildlife.

Introduction

After the strong reduction in the use of many chlorinated persistent organic pollutants (POPs) 30–40 years ago, the global concentrations of these chemicals declined substantially. For example in aquatic birds from temperate areas, compounds such as polychlorinated biphenyls (PCB) showed major declines in the 1970s and early 1980s (1, 2). For the Arctic regions there are few time series available for POPs, but in the Canadian Arctic legacy POPs have declined in

several seabird species after the mid-1970s (*3, 4*). In the Norwegian Arctic, PCBs decreased significantly in polar bear (*Ursus maritimus*) plasma during the 1990s (*5*), but for marine birds there have been no analyses of temporal trends from this Arctic region.

The variation in the concentrations of lipid-soluble POPs in wildlife is, however, not only influenced by the changes in the rate of chemical release into the environment. One issue that is receiving growing research attention is how a changing climate may impact the occurrence of POPs in the Arctic (6). Predicted climate change will have profound effects in the Arctic via temperature increase, more low-pressure activity, augmented runoffs from glaciers, and increased rate of sea ice melting. These climate-related variables may influence the transport and bioavailability of POPs (6-8), but the lack of long-term data series has made it difficult to assess such effects in arctic species.

In glaucous gulls (Larus hyperboreus) breeding on Bear Island (Bjørnøya) in the Barents Sea, POPs have been measured in hundreds of blood and plasma samples since the mid 1990s (9). In the present study POP data in glaucous gull blood/plasma were used to examine the temporal trends (1997–2006) of three types of ubiquitous legacy POPs (PCB, HCB [hexachlorobenzene], and oxychlordane) and to assess the impact of climate variability on POP concentrations, using the Arctic Oscillation index (AO) as a climate proxy. The AO is a measurement of low pressure activity and precipitation in the European Arctic and has previously proven useful in understanding the impacts of climate variability on contaminant pathways and trends (6). We predicted that the levels of POPs would decline as a result of long-term reductions in emissions, but the complexity of climate processes made it difficult to predict potential impacts on POPs (7, 10). Hence, the effects of climate variation on POPs concentration may be direct, through increased atmospheric transport. Under such circumstances one might expect variations between compounds with different transport potentials, e.g. between a light volatile compound such as HCB and a heavy chlorinated PCB congener with lower transport potential (6, 8, 11). However, the impact of climate may also be indirect through alterations in prey availability and composition leading to changes in POP intake and gull physiology.

Materials and Methods

Study Area. Data were collected from breeding glaucous gulls (all >5 years of age) on Bear Island (74° 30′ N, 19° 01′ E) from late May to the end of June between 1997 and 2006 (except 1999, 2003, and 2005). Glaucous gulls were caught on their nests using a nest trap (12) and individually marked with PVC leg bands and numbered steel rings. Blood (~10 mL) was sampled from a wing vein with a syringe and frozen (-18 °C) within a few h of sampling. From 1997 to 2001 whole blood was used for analyses, while from 2002 to 2006 the blood was centrifuged before it was frozen, and plasma was used. Skull length (head + bill) and wing length (all morphometric values measured were ±0.5 mm) were recorded for all birds. Sex was determined based on the total head and bill length (females are smaller than males, with no size overlap between sexes) (12). Wing length was used as a general measure of body size. Body condition was measured as body mass corrected for body size (wing length).

Previous studies have reported that major subcolonies of glaucous gulls on Bear Island accumulate different levels of POPs as a result of dietary specialization; i.e. gulls breeding

^{*} Corresponding author phone: $+47\ 77\ 75\ 04\ 07$; fax: $+47\ 77\ 75\ 04\ 01$; e-mail: Jan.O.Bustnes@nina.no.

[†] Norwegian Institute for Nature Research.

[‡] Norwegian Polar Institute.

[§] Université du Québec à Montréal.

nearby the major seabird cliffs are feeding predominantly upon adult seabirds and their eggs and chicks. Therefore, these birds accumulate more POPs than gulls breeding at the seashore locations, which feed more on fish and crustaceans (13-15). In the present study, only blood of birds (n=240) collected from the major seabird cliffs was used, since there were no data on seashore locations after 2002.

Climate Variables. Climate variation may be measured in a multiude of ways (temperature, wind, precipitation, snowfall, sea ice distribution, glacier melting, etc.). These variables can further be divided into subvariables such as seasons. It is therefore necessary to make a priori assumptions concerning the variables to include in analyses (16). Furthermore, it has been demonstrated that large scale climate indexes often better predict variation in ecological processes than local climate (17, 18), and several studies have demonstrated the usefulness of AO (Arctic Oscilliation) and NAO (North Atlantic Oscilliation) as proxies in studying the impact of climate variation on contaminant pathways and trends in the Arctic (6-8). The AO index is an indicator of sea-level pressure variability for the Northern Hemisphere above 20°N, and its pattern has a dominant low-pressure region centered roughly over the Arctic Ocean. A positive AO index implies a strengthening of the counterclockwise polar vortex from the surface to the lower stratosphere. The asymmetries in the pressure pattern mean that cool winds sweep eastsoutheast across eastern Canada, and southwesterly North Atlantic storm tracks bring rain and mild temperatures to northern Europe. AO thus summarizes, on a large scale, a number of climate variables and is reflecting variation in the basic transport pathways of POPs (atmospheric transport), as it is a measure of air flow from temperate areas to the Arctic (6). Data on the AO for all relevant years were obtained from the Web site of the National Weather Service, Climate Prediction Centre (http://www.cpc.noaa.gov/ products/ precip/CWlink/daily_ao_index/ao_index.html). To assess the seasonal impacts and the tempral effects of climate variables on the accumulation of POPs in glacous gulls, three different measurements of AO with different time lags from contaminant sampling were used in the analyses: 1) AO_{current winter} (the monthly means of the December-March for a given breeding season); 2) $AO_{preceding\; summer}$ (the monthly means of the June-September AO during the year prior to POP measurements); and 3) $AO_{preceding\ winter}$ (the monthly means of the December-March during the year prior to POP measurements).

Chemical Analyses. Analyses of POPs in glaucous gull blood/plasma samples were carried out at the Environmental Toxicology Laboratory at the Norwegian School of Veterinary Science in Oslo (NVH), Norway (1997–2001), and the National Wildlife Research Centre in Ottawa (NWRC), Canada (2002–2006). The analytical details (extraction, cleanup, partitioning, and instrumental analysis) employed for the determination of POPs in plasma and whole blood have been comprehensively described elsewhere (12, 15, 19). In 1999 an interlaboratory test was performed between the NWRC and the NVH. In this method validation test, 20 plasma samples of polar bears from Svalbard were analyzed for PCBs and selected chlorinated pesticides. The results showed excellent agreement between the laboratories for the PCB congeners used in the present study as well as HCB and oxychlordane. Since these OCs are the main ones found in glaucous gulls at levels that are generally comparable to the polar bear, the results of the PCB, HCB, and oxychlordane analyses between the two laboratories can be considered directly comparable. In this study, PCB was the sum of CB-99, -118, -153, -138, -180, and -170, and we used lipidnormalized POP concentrations in order to control for extractable lipid content differences between plasma and whole blood samples (20).

Data Analyses. Multiple samples from the same individuals were removed so that there was only one blood/plasma sample for each individual. When there was more than one sample per individual, the observation from the year with the lowest total sample size was included in the final data set.

The data were analyzed using Linear Models in R (21). The goodness of fit of linear models was assessed using partial residual plots and influence values (22). Simple regression analyses were used to estimate changes in levels over the years, while the information criterion approach was used for selecting the most parsimonious models among a set of potential predictors. We chose the most parsimonious statistical models based on the Akaike's Information Criteria corrected for small sample size (AICc). More specifically, we calculated the relative likelihood of each model using AICc weights derived from difference in AICc values between the best model (lowest AICc) and other models (16). To assess the robustness of the model selection procedure we used variable importance (sum of AIC weights of all models including this variable). Variables with high importance (i.e., >0.8) come out systematically in all best models, whereas variables with lower importance cannot be reliably seen as having an effect (16). To estimate the concentrations in the early and late sampling period we back-transformed from log (ln) values in the regression models (year vs POP concentrations).

Results and Discussion

Temporal Trends. Despite substantial interannual variation in POP concentrations (Table 1, Figure 1), simple regressions showed significant (p < 0.05) or near significant (p < 0.1) declines (range: 16-60%) in POP concentrations, except for oxychlordane in females (Table 2). The relationships were particularly strong for PCB in both sexes as well as for HCB in males (>50% decline; Table 2). By comparison, in aquatic birds from temperate regions, the major reductions in POPs (e.g., PCB) typically occurred in the 1970s, after which the concentrations leveled out. For example, in herring gulls (Larus argentatus) from the North American Great Lakes (1) and in guillemots (Uria aalge) from the Baltic Sea (2) the concentrations of PCB in eggs had decreased by nearly 70% by the early 1980s. In contrast, the concentrations of PCB in liver samples of glaucous gulls from Bear Island seemed to have decreased to a lesser extent (~30%; 16 vs 24 ppm, respectively) in the 17-year period between 1972 (23) and 1989 (24). Although different PCB congeners may have been selected for quantification in these glaucous gull studies, the data still suggest a noteworthy decline in POP concentrations in glaucous gull after the mid 1990s. Hence, major reductions may have started later in this Arctic-breeding species relative to aquatic birds from temperate and industrialized regions. A slower reduction of POPs in the Arctic compared to temperate areas has also been suggested based on surveys of POPs in seabird eggs from both the Canadian Arctic (3) and the European Subarctic (25). This difference might be due to time-lags between reduced inputs from the POP sources and detectable reductions in Arctic biota. One potential bias in the present study is that our data are limited to one colony site. Glaucous gull may feed upon a large spectrum of prey items, including terrestrial food (26), which may imply that data from one location may not necessarily reflect the general trends in marine birds from a larger region. However, in the Norwegian Arctic, glaucous gulls almost exclusively feed upon marine preys (13, 26). Hence, gulls from Bear Island are not expected to differ substantially from other avian marine top predators in this region, although actual concentrations and strength of trends may vary as a result of inter- and intraspecific trophic level variation (13, 14).

TABLE 1. Sample Sizes and Concentrations (μ g/g, Lipid Weight) of Three POPs in Blood of Female and Male Glaucous Gulls Breeding on Bear Island between 1997 and 2006

				fe	males					n	nales		
year		n	mean	median	SD	min	max	n	mean	median	SD	min	max
1997													
	PCB	21	63.39	55.72	58.84	5.10	290.91	17	136.58	75.16	144.72	25.98	623.6
	HCB	21	2.47	2.45	2.45	0.15	4.84	17	6.76	5.52	5.67	0.98	21.5
1998	oxychlordane	21	3.45	2.67	3.17	0.33	15.7	17	6.85	4.38	6.00	1.65	25.7
1550	PCB	8	40.17	34.69	22.17	16.54	83.97	8	88.51	87.70	39.38	32.25	144.45
	HCB	8	2.71	2.99	1.61	0.70	5.91	8	4.19	3.41	2.22	1.59	7.57
	oxychlordane	8	1.63	1.36	0.86	0.80	3.22	8	3.33	2.47	1.84	1.25	6.67
2000	PCB	15	48.16	38.97	22.32	19.76	86.65	10	64.58	67.39	36.95	13.87	132.80
	HCB	15	1.89	1.85	0.64	1.05	3.42	10	2.29	1.83	1.34	0.65	4.31
	oxychlordane	15	1.60	1.40	0.72	0.78	3.39	10	1.75	1.74	0.87	0.34	3.24
2001	•												
	PCB	19	85.34	68.46	69.93	18.53	273.11	16	106.88	70.22	104.63	30.73	420.49
	HCB	19	5.27	4.36	3.04	1.33	13.49	16	5.90	5.69	2.18	2.55	9.61
2002	oxychlordane	19	2.88	1.94	1.94	0.94	7.64	16	3.31	2.56	2.12	1.37	8.67
2002	PCB	34	21.29	17.88	11.30	6.45	53.19	34	30.27	27.56	17.96	7.53	90.07
	HCB	34	0.36	0.32	0.18	0.11	0.78	34	0.53	0.42	0.36	0.09	1.37
	oxychlordane	34	1.44	1.24	0.87	0.42	4.58	34	1.88	1.89	0.95	0.49	4.18
2004	PCB	14	49.87	42.66	29.52	22.23	132.35	10	52.61	45.65	25.23	25.51	93.95
	HCB	14	3.84	2.83	2.03	2.20	7.93	10	3.98	3.35	1.76	2.06	7.18
	oxychlordane	14	4.64	3.66	3.01	1.47	11.39	10	3.83	2.93	2.38	1.39	8.06
2006	oxyomor auno			0.00	0.01	,	11.00		0.00	2.00	2.00	1.00	0.00
	PCB	19	24.50	20.67	13.98	7.47	57.07	15	66.06	53.46	50.26	7.71	167.84
	HCB	19	1.59	1.32	0.90	0.56	3.69	15	3.29	3.17	2.18	0.69	8.85
	oxychlordane	19	1.72	1.44	0.87	0.57	3.49	15	4.20	3.95	3.21	0.74	11.82

The Impact of Climate on POP Accumulation. To assess the relationships between climate variability and temporal changes in POP concentrations, models with the three AO predictor variables (AO $_{current\ winter}$, AO $_{preceding\ summer}$, and AO $_{preceding\ winter}$) were examined for each compound, controlling for year, sex and body condition (Table 3). First, males had higher blood concentrations of POPs than females (Tables 1 and 2), which is typically being observed in studies of glaucous gulls shortly after egg-laying (9, 27, 28). This sexspecific difference in POP concentrations is the result of maternal transfer of POPs to the eggs (29) and potentially to specialization of males and females on certain food items (feeding ecology) as suggested elsewhere for herring gulls (30). However, the most parsimonious models did not include the interactions between sex and year (variable importance [VI] between 0.22 and 0.32), suggesting that POP trends were more or less similar for males and females. Therefore, interactions between sexes were not considered further. The relationships between body condition and POP concentrations were weak, with the exception of oxychlordane (Table 3). This was unexpected since studies of glaucous gulls have shown increasing blood concentrations of POPs as a function of decreasing body condition (27, 28).

There was no evidence that males and females responded differently to climate variation, as the interactions between sex and AOs on POP concentrations had low variable importance (0.23 < VIs < 0.34). The best models included all three measurements of AO (VIs > 0.76; Table 3). More specifically, AO current winter was negatively associated with POP concentrations, whereas the relationships with AO preceding summer and AO preceding winter were positive (Figure 1, Table 3). These differences may be caused by the time elapsed since the POPs were transported to the Arctic (9–18 months for AO preceding summer and AO preceding winter and 3–6 month for AO current winter) and the likelihood that these compounds reached species occupying the top of the food chain (6, 31). Hence, the negative effect of AO current winter on POP levels may

be an indirect relationship, caused by a modulation in intake/ assimilation of POPs induced by dietary change and/or by physiological changes (e.g., nutritional status and body condition) in the gulls. For example, the winter climate might impact the preys that gulls are feeding upon during the following summer. Hence, after cold winters high trophic preys with high loadings of POPs may be less abundant than after mild winters. Alternatively, the negative relationship may be a result of different wintering locations of glaucous gulls in mild years vs cold years; e.g. in colder winters (AO⁻) glaucous gulls may be forced to migrate further south in the North Atlantic or Barents Sea where background levels and thus the dietary intake of POPs may be higher than in the Arctic regions (32). Such relationships have been demonstrated in herring gulls where birds moved to more contaminated southerly Great Lakes locations in severe winters, after which their eggs contained higher concentrations of POPs the following breeding season, compared to mild winters (33).

The positive relationship between the AO indices from the year prior to POP measurements was consistent with predictions by MacDonald et al. (6). Hence, more northwards air transport seems to have increased background levels of POPs in the Arctic, which again resulted in higher POP loadings accumulating in glaucous gulls at the top of the food chain in the following breeding season. Moreover, air mass transported northwards during the summer may contain higher concentrations of POPs than in the winter since high temperatures in temperate regions favor volatilization from the Earth's surface (7, 31). However, the winter season accounts for about 80% of the annual south-to-north air mass transport (34). It may thus be difficult to predict what season that will be the most important contributor to increasing POP concentrations in glaucous gulls. To test this linkage, models were run for PCB and HCB using $AO_{preceding\ winter}$ and $AO_{preceding\ summer}$ (in addition to year and sex). The results showed that AO_{preceding winter} explained two

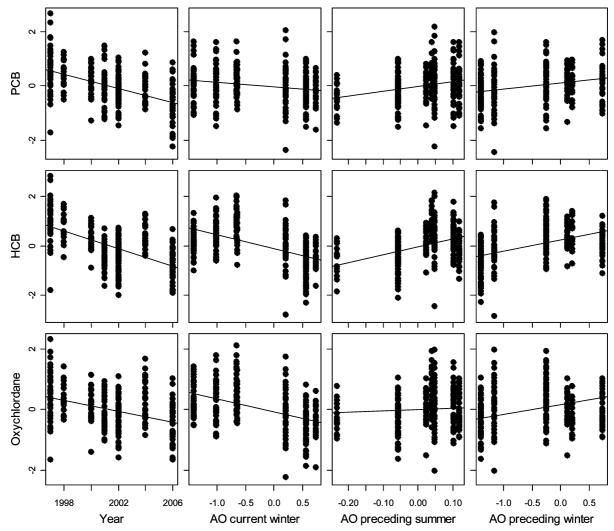


FIGURE 1. Relationships between the concentrations of three legacy POPs (PCB, HCB, and oxychlordane) in blood/plasma of glaucous gulls breeding on Bear Island, and the predictors: year, Arctic Oscillation (AO) in the current winter, AO in the preceding summer, and AO in the preceding winter. The plots are partial residuals based the most parsimonious models in Table 3; i.e. the effects of all four predictors have been controlled for each other, in addition to sex and body condition, if included in the top models.

TABLE 2. Estimated Concentrations (μ g/g, Lipid Weight) and Percentage Total Change of Three Legacy POPs between 1997 and 2006 in Male and Female Glaucous Gulls Breeding on Bear Island^a

		1997	2006	change %	intercept	slope	p	R ²
PCB								
	males	76.53	30.97	-59.5	211.3	-0.10	0.0004	0.10
	females	47.69	22.84	-52.1	174.1	-0.082	0.0004	0.09
HCB								
	males	2.96	1.15	-61.1	215.3	-0.104	0.015	0.05
	females	1.81	1.07	-40.9	123.3	-0.058	0.09	0.02
oxychlordane								
	males	3.08	1.99	-35.4	102.0	-0.05	0.07	0.02
	females	2.00	1.69	-15.5	45.5	-0.02	0.38	0.006

^a The estimates are based on simple regressions between POP concentrations and year.

to three times as much of the variation in PCB (9% vs 4%, respectively) and HCB (31% vs 10%) concentrations relative to $AO_{preceding\ summer}$ (Figure 2). Hence, the present data suggest a stronger effect of $AO_{preceding\ summer}$ compared to $AO_{preceding\ summer}$. Moreover, if climate variation is of prime importance for POP accumulation in glaucous gulls, then a greater impact of AO would be expected for lighter vs heavier chlorinated compounds. That is the volatile HCB is more prone to long-range transport compared to heavy chlorinated PCB congeners (7, 11, 31, 35). This was apparent for

AO_{preceding summer} and AO_{preceding winter} that explained a higher percentage of variation in HCB concentrations (41% based on the sum of partial R^2 for the variables) compared to higher chlorinated and less-volatile PCBs (13% [Figure 2]).

There are, however, other confounding variables that have not been accounted for in the present study. For example, changes in a species' diet may affect the accumulation dynamics of POPs over time, which may overshadow or confound temporal variations of POPs (36, 37). Unfortunately, temporal variation in glaucous gull diet on Bear Island has

TABLE 3. Model Selection for the Impact of Different Variables on the Concentrations of Three POPs in Glaucous Gulls from Bear Island between 1997 and 2006^d

				slope estii	slope estimates (SE) P						
	intercept	year	sex (males vs females)	wing (body size)	body mass	A0 current winter	A0 preceding summer	A0 preceding winter	k B ²	AAICc	AICc weights
PCB		-	880	27 0	0.45	92.0	800	0.87			
model rank		_	60.0	t	6			0.0			
_	268.7	$-0.129 (0.017)^c$ ($0.423~(0.086)^c$			-0.169 (0.092)	$1.72 (0.54)^b$	$0.217 (0.08)^{a}$	7 0.34		0.167
2	282.0	-0.137	χ_{Θ}	0.0075	-0.00070	-0.202		0.191	9 0.35	0.278	0.145
ო	266.2	-0.129	χ°	0.0058		-0.189		0.193	8 0.34		0.126
4	281.2	-0.135	%		-0.00053	-0.174		0.222			0.118
2	246.9	-0.118	*					0.330	6 0.33		0.085
9	258.0	-0.123	**		-0.00049			0.336	7 0.33		0.055
HCB											
variable importance		_	0.90	0.28	0.32	_	_	_			
model rank											
_	361.4	$-0.177 (0.019)^{c}$	$0.343~(0.095)^c$			$-0.565 (0.101)^{c}$		$0.451 (0.093)^{c}$	7 0.61	0	0.478
2	366.9	-0.180 x	*		-0.00023		3.19	0.452	8 0.61		0.183
ო	362.2	-0.177	%	-0.0018				0.458	8 0.61		0.175
4	366.8	-0.179	°×	-0.0013	-0.00020	-0.563		0.458		4.009	0.064
2	345.3	-0.169			69000'0			0.442	7 0.60		0.054
oxychlordane											
variable importance		_	0.92	0.52	0.76	_	0.89	0.92			
model rank											
-	189.2	$-0.091 (0.018)^{c}$ ($0.535 (0.196)^c$	0.0073 (0.0048)	$-0.0011 (0.0005)^{a}$	$-0.434 (0.093)^{c}$	$1.310 (0.55)^{a}$	$-0.240 (0.087)^{b}$	9 0.20		0.308
2	188.4	060'0-	%		-0.0009	-0.407	1.373	-0.211	8 0.20		0.278
ო	167.3	-0.080	%			-0.399	1.587	-0.218			0.105
4	165.3	-0.080	*	0.0046		-0.415	1.576	-0.237	8 0.19	3.313	0.059
2	162.9	-0.080		0.0103		-0.438	1.542	-0.264	7 0.18		0.051

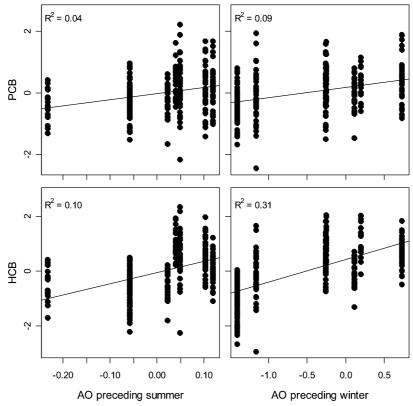


FIGURE 2. Relationships between the concentrations of PCB and HCB in blood/plasma of glaucous gulls breeding on Bear Island and the predictors: Arctic Oscillation (AO) in the preceding summer and AO in the preceding winter. The plots are partial residuals, and both predictors have been controlled for each others, in addition to year and sex. Partial R^2 values are shown in the plots.

not been systematically recorded between 1997 and 2006 and thus remains a confounding factor in the present study. The issue of climatic effects is thus complex, and it is important to keep in mind the correlational nature of the herein reported data.

In conclusion, there were negative temporal trends in POP concentrations also when a suite of influential biological and environmental variables was considered in multiple regression models (VIs_{year} \approx 1; Figure 1, Table 3), suggesting reduced influx of these chemicals to the Arctic since the mid 1990s. As such, legacy POPs may be a diminishing health risk to Svalbard glaucous gulls. However, the effects of POPs are not solely determined by their concentrations as other anthropogenic or natural stressors (e.g., food scarcity, predation, climate change, and diseases) may influence or add to the effect potential. Hence, if future changes in the Arctic regions lead to increased stress, the POPs may still be a significant health hazard to the Arctic-breeding glaucous gulls (9, 38). Furthermore, this study lends some but not conclusive support to the notion that climate variability impacts the transport and fate of POPs, suggesting that future climate change could modulate POP trends in the Arctic (6), although such an impact may be indirect. This is worrying, but it is uncertain whether this will be sufficient to modulate the downward trends of legacy POPs. However, new persistent pollutants are being detected in the glaucous gull and include a wide range of emerging chlorinated, brominated, and fluorinated chemicals in which some are currently under production and usage (18, 39, 40). Therefore further studies into the linkages between climate change and concentrations of legacy and emerging POPs in the Arctic are needed.

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