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Effects of Seabird Vectors on the Fate, Partitioning, and Signatures of Contaminants in a High Arctic Ecosystem

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 Supporting Information

ABSTRACT:



Seabirds bioaccumulate contaminants from prey, transport them to their nesting sites, and deposit them in their excreta and carcasses, thereby focusing marine-derived contaminants into remote, terrestrial receptor sites. In the case of organochlorine chemicals transported by northern fulmars (*Fulmarus glacialis*) to a High Arctic seabird colony on Devon Island, Nunavut, Canada ($76^{\circ}13'N$, $89^{\circ}14'W$), this contaminant pathway dominates all others. In freshwater ponds below the nesting cliffs, concentrations of organochlorine contaminants characteristic of fulmar input were 2- to 45-fold higher in sediments and water (depending on seabird input to the particular pond) than in ponds remote from the colony. Air–water fugacity quotients for the ponds decreased with seabird input, indicating that fulmar contaminant input shifts air–water partitioning to increasingly favor volatilization to air. Although contaminant evasion from water was favored, direct evidence of it was not detected in air samples. For PCBs, congener profiles of pond sediments or water became more similar to seabird sources as seabird input increased, and less similar to air profiles. Based on measurements of contaminants in fulmars and other local environmental media, this study presents the first application of fugacities and multivariate source apportionment statistics to resolve seabird biological vectors.

INTRODUCTION

As the implementation of international agreements result in a reduction of the primary sources of contaminants such as PCBs,¹ emissions from secondary sources become increasingly important to contaminant fate and cycling.² These secondary sources, which include reservoirs like soil and vegetation, may remobilize accumulated contaminants into air and water through forest fires³ and thermal revolatilization from soils.² Similarly, contaminants in the ocean can be focused into terrestrial receptor sites by biovectors such as seabirds and anadromous fish.⁴

Seabirds accumulate contaminants from prey items obtained broadly in the ocean, but they deposit these into relatively small

areas near their nesting sites via guano, regurgitated or dropped food, shed feathers, and carcasses.⁴ Thus, seabirds are exemplary biological vectors for contaminants like persistent organic pollutants and toxic trace metals.^{5–13} The Canadian Arctic Archipelago alone supports >10 million breeding seabirds.¹⁴ Remote from primary sources of contaminants, Arctic receptor sites may become dominated by contaminants delivered by seabirds.

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For example, in the Norwegian Arctic nesting seabirds contributed 80% of the PCB inventory of Ellasjøen Lake.⁷ Large seabird colonies in the Arctic also provide nutrient subsidies in an otherwise impoverished terrestrial environment, thus seabird vectors for contaminants may be of particular concern for terrestrial food webs supported by seabird-derived nutrients.^{15,16}

Most studies have assumed that seabirds elevate contaminant concentrations in the environment near their nesting sites solely via deposition of guano.^{7,10} However, this obvious pathway could be augmented by, for example, stomach oils generated by the birds from prey items; stomach oils may be used for defense and feeding.¹⁷ Northern fulmars (*Fulmarus glacialis*) readily expel such oils when competing for nest sites and repelling predators, and oil frequently lands on nearby snow and rocks eventually washing into the catchment below the nesting cliffs. Stomach oils contain organochlorine contaminants at higher concentrations than found in guano and prey.^{18,19} Discerning the predominant sources of seabird-derived contaminants to a given coastal setting is particularly relevant as contaminant signatures differ between sources.¹⁹

Here, we present a comprehensive, multidisciplinary assessment of the effects of a large northern fulmar colony on contaminants in the nearby coastal Arctic environment. This assessment builds on the ecological, limnological, and contaminant research conducted at Cape Vera on Devon Island, Nunavut, where we have studied the fulmar life history and diet,^{20,21} the effects of the colony on water and sediment chemistry of nearby freshwater ponds,^{6,10,12,13,22} and the contamination of marine and terrestrial biota.^{15,16,18,19} Organochlorine contaminant concentrations were measured in study ponds spanning a gradient from high seabird input beneath the nesting cliffs to almost no input at 4 km distant. During three years of fieldwork, we measured concentrations of polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) in pond sediments and water, in ambient air, in fulmars, and in their guano, stomach oils, and carcasses. Fugacities were calculated to determine the effect of seabird contributions on local environmental reservoirs and to determine the direction of net flux between media. Statistical source apportionment was used to assess similarity between contaminant profiles in ponds and those in possible sources. To our knowledge, this is the first study to measure environmental concentrations of organochlorine contaminants in all relevant media such that fugacities and source apportionment statistics could be applied to the question of contaminant forcing by biovectors.

■ EXPERIMENTAL SECTION

Study Site. Cape Vera ($76^{\circ}13'N$, $89^{\circ}14'W$; Figure 1), Devon Island, Nunavut in the Canadian High Arctic hosts $>20\,000$ mature northern fulmars, a species of Procellariiform seabird (commonly called “petrel” or “tubenose”), during the breeding season from early-May through mid-September.²³ Below the nesting cliffs, 13 freshwater ponds, representing a gradient of seabird input, were selected for this study. All ponds are within 4 km of each other and thus are presumably exposed to similar climatic variables and background air concentrations of contaminants. Being marine feeders, fulmars nesting at the colony do not interact directly with the ponds. Pond coordinates are given in the Supporting Information (Table S1).

Sampling. Air was passively sampled using XAD resin-based samplers described elsewhere^{24,25} (see Supporting Information).

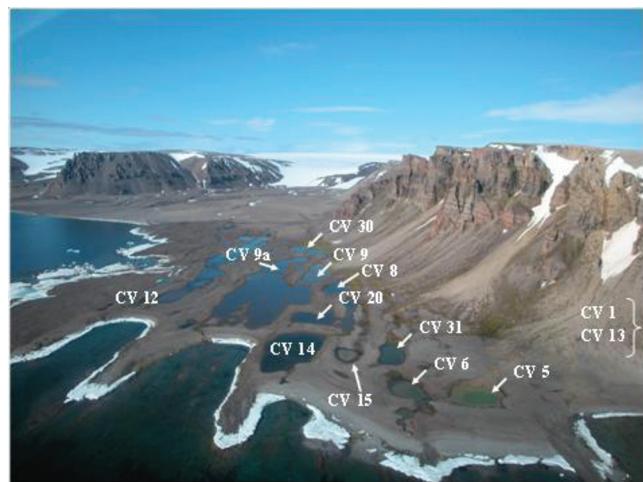


Figure 1. Aerial photo of study ponds at Cape Vera. Ponds CV 1 and CV 13 are outside the frame of the photo. Photo by J.P. Smol.

Stainless steel mesh sampling cylinders plugged with glass wool were packed with precleaned Supelpak-2 resin (Sigma Aldrich Canada), spiked with field recovery standards (CBs 30, 204, 1,3-dibromobenzene, 1,3,5-tribromobenzene, 1,2,4,5-tetrabromobenzene, δ -hexachlorocyclohexane, endrine ketone), stored in aluminum canisters with Teflon washers and kept frozen until deployment. Four samplers were deployed at the study site; two samplers were positioned at the base of the nesting cliffs and two at locations away from the colony. Air sampling was conducted for a total of two years and each set of sampling cylinders were deployed for approximately one year before replacement with fresh cylinders. Thus, data reflects the average, time integrated concentrations for 2006/2007 and 2007/2008.

Pond water was passively sampled using semipermeable membrane devices (SPMDs), which are described elsewhere^{26,27} (see Supporting Information). Briefly, 1 mL of 99% purity triolein spiked with performance reference compounds (CBs 14, 19 (in 2007 only), 32, 155, 203) was pipetted into 60 cm lengths of 3.3 cm wide polyethylene layflat tubing and heat sealed with loops at either end. SPMDs were stored in amber mason jars and kept frozen until deployment. In the field at least four SPMDs were fastened inside each sampling cylinder and suspended mid way in the water column using floats and weights. The SPMDs were deployed for 32 days in 2006 and for 16 days in 2007.

Surficial sediments were collected in 2006 and 2007. The surface of undisturbed sediments was skimmed directly into Whirl-Pack bags, excess water drained, and the bags were sealed and kept frozen until PCB/OCP analysis. Fulmar stomach oils, guano, and whole body homogenate (WBH) were collected from harvested birds in 2006 and 2007.^{18,19}

Organic Contaminant Analysis. Analytes were extracted from XAD resin and SPMDs using Soxhlet and dialysis, respectively, based on established, standardized methods.^{26,28} The extracts of two replicate SPMDs were combined for the 2007 samples because of the reduced deployment period. Analytes were extracted from sediments using an accelerated solvent extractor (ASE 200, Dionex); each sample was extracted first with hexane, then with dichloromethane. Details of the organochlorine contaminant analyses of fulmar tissues and media are given elsewhere.^{18,19} Co-extracted constituents were removed from SPMD extracts by gel permeation chromatography,¹⁸ and from

sediment extracts by liquid chromatography on a preparative LC (Agilent 1200 series) with dichloromethane. All samples were then passed through activated silica (60–100 mesh size)/sodium sulfate columns to remove water, further cleanup the extract, and to fractionate the analytes into two fractions. Fraction A, consisting of the PCBs and less polar OCPs, was eluted first from the column using hexane. Fraction B, consisting of the remaining, more polar, OCPs was eluted using 1:1 hexane: dichloromethane.

All samples were analyzed on a Hewlett-Packard 6890 series II gas chromatograph with a ^{63}Ni electron capture detector and a $60\text{ m} \times 0.25\text{ mm}$ ($0.25\text{ }\mu\text{m}$ film) DB-SMS column (J&W Scientific) and helium carrier gas at a flow rate of 2.0 mL min^{-1} . Calibration standards were run with every batch of ~ 10 samples to update concentration–response curves and to check for the possible coelution of analytes. PCBs (collectively ΣPCBs) measured were IUPAC congeners: 5/8, 18, 29, 31/28, 52, 49, 44, 66, 101, 99, 87, 110, 149, 118, 146, 153, 132, 105, 163, 138, 187, 183, 128, 156, 201, 180, 170, 195, 194, 206, 209. OCPs measured were: 1,2,3-trichlorobenzene (1,2,3-TCB), 1,2,3,4-tetrachlorobenzene (1,2,3,4-TTCB), pentachlorobenzene (PECB), α -hexachlorocyclohexane (α -HCH), hexachlorobenzene (HCB), γ -hexachlorocyclohexane (γ -HCH), heptachlor, aldrin, heptachlor epoxide (HE), γ -chlordane, α -endosulfan/ α -chlordane (α -endo./ α -chlor.), p,p' -dichloro-diphenyl-dichloroethylene (p,p' -DDE), dieldrin, endrin, β -endosulfan, p,p' -dichloro-diphenyl-dichloroethane (p,p' -DDD), o,p' -dichloro-diphenyl-trichloroethane (o,p' -DDT), p,p' -DDT, methoxychlor and mirex.

Method recoveries of spiked standards for all pond and air samples ($n = 115$) including method blanks, field blanks, and reference materials were on average $73\% \pm 31$ for CB 30 and $78\% \pm 25$ for CB 204 or 205. QA/QC details for fulmar samples are reported elsewhere.¹⁹ Standard reference materials ($n = 3$, sediments, National Institute of Standards and Technology 1944) were analyzed with each batch of sediment samples. The average concentrations of 25 PCBs and of 7 OCPs were $69\% \pm 32$ and $54\% \pm 28$ of the certified concentration, respectively. Concentrations in sediments were method blank corrected and concentrations in air and water were field blank corrected to account for possible background contamination associated with transportation to and from the field. Air concentrations were recovery corrected using the recoveries of field recovery standards. Method detection limits (MDLs), calculated as the Student's t value appropriate for a 95% confidence level times the standard deviation of the averaged blank concentrations, for ΣPCBs were 0.038 ng g^{-1} for sediments ($n = 3$), 0.9 ng SPMD^{-1} ($n = 13$), and $4.9\text{ ng sampler}^{-1}$ for air ($n = 5$). MDLs for ΣOCPs were 0.036 ng g^{-1} for sediments, 1.7 ng SPMD^{-1} , and $1.3\text{ ng sampler}^{-1}$ for air. A set of triplicates were analyzed for surface sediments; average coefficients of variation were 35% for ΣPCBs and 26% for ΣOCPs . All materials, tools, sampling equipment and glassware were solvent washed, and where possible baked. All solvents used were high-purity grade.

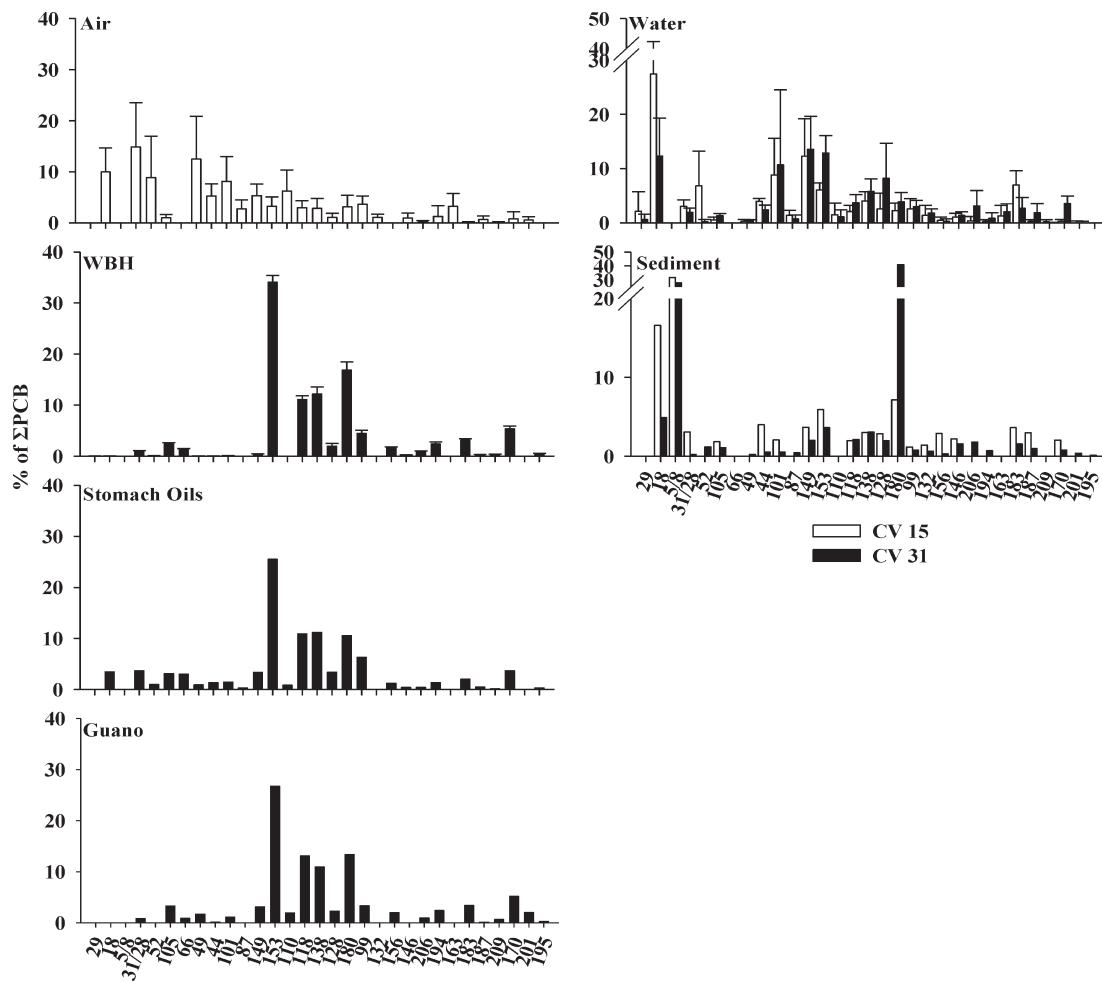
Tracing Fulmar Input to Ponds. At Cape Vera, fulmar-derived biovector elements (As, Cd, K, P, Zn) were previously identified.¹⁰ Here, we used sedimentary Zn concentrations as a tracer of fulmar input; for full details see Supporting Information. Briefly, Zn is used because of its comparatively stable geochemistry,²⁹ lack of nearby confounding sources,³⁰ and demonstrated correlation with an independent indicator of seabird input (i.e., distance from the colony).¹⁰

Sedimentary Zn concentrations were determined as described previously.¹⁰ Briefly, surficial sediment samples were freeze-

dried, ground to a fine powder, extracted using aqua regia digestion and analyzed using inductively coupled plasma mass spectrometry. The limit of detection for Zn was $0.7\text{ }\mu\text{g g}^{-1}$ dry weight. All elemental analyses were conducted by an accredited laboratory (SGS Canada Inc. Lakefield, ON).

Data Analysis. Fugacities. Fugacities (${}^31\text{ f Pa}$), which are chemical- and media-specific, were calculated as the chemical concentration ($C\text{ mol m}^{-3}$) divided by the fugacity capacity ($Z\text{ mol m}^{-3}\text{ Pa}^{-1}$) of the media (Supporting Information, Tables S2–S4). Air–water fugacity quotients, $f_{\text{Air}}/f_{\text{Water}}$, frequently used to assess the direction of the net flux of contaminants at the air–water interface of oceans,^{32,33} were computed for the study ponds. If $f_{\text{Air}}/f_{\text{Water}} = 1$, the chemical is at equilibrium between air and water and no net exchange of contaminant is expected. If $f_{\text{Air}}/f_{\text{Water}} > 1$ the deposition of the contaminant from air into water is favored, if $f_{\text{Air}}/f_{\text{Water}} < 1$ volatilization of the chemical from water into air is favored. For the present application, the shift in $f_{\text{Air}}/f_{\text{Water}}$ with increasing seabird input to the ponds is also relevant. $f_{\text{Air}}/f_{\text{Water}}$ was computed for each chemical using the f_{Water} for each water sample (i.e., SPMD) and the average f_{Air} of the eight passive air samplers.

Source Apportionment. Using all PCB congeners as variables, Mahalanobis distances between the mean vectors of each of the four PCB sources considered here (air, stomach oils, guano, and seabird carcasses) and those of each individual pond sample (either sediments or water) were computed in MYSTAT (version 12). Mahalanobis distances indicate how similar the PCB profiles in the pond water or sediment samples are to that of each source, with small distances indicating the profiles are similar and larger distance indicating the profiles are different. Rather than using the PCB air profile, we used the mean vector of three control ponds because PCB profiles in the sediments and water of the control ponds are attributable to atmospheric contributions of PCBs, and these profiles have already been subject to any congener-specific bias in the transfer of PCBs from air as a result of different physicochemical properties (e.g., the air–water partition coefficient, K_{AW} ³¹). Two sets of source apportionment analyses were performed, one for pond water and one for pond sediment. For each analysis, PCB congener concentrations in each sample were normalized to the concentration of CB 153 in the sample, and expressed in deviations from the mean of all samples in the analysis (i.e., guano, stomach oils, carcasses, and water or sediment samples) by dividing by the standard deviation. Normalizing to CB 153 instead of ΣPCBs was preferable because CB 153 was present at comparatively high concentrations in every sample, whereas the congeners comprising ΣPCBs shifted between sample types. For this preliminary statistical analysis, nondetects were replaced with the method detection limit (MDL) specific to the congener and sample type, resulting in substitutions for 30% of data set. Substitution methods for nondetects can be associated with an increased risk of type II error and results concluding that a variable is not the likely source of contamination should be viewed as suspect.³⁴ Subsequent analyses of contaminant biovectors should assess the effect of using different nondetect techniques on Mahalanobis distances. However, such treatments are unlikely to change the conclusions of the present study since trends with seabird input were noted (i.e., the null hypotheses were rejected) for both pond water and sediment analyses. Source apportionment analyses were not performed for OCPs because of the broader range of persistence between constituents which would also influence the overall OCP profile.



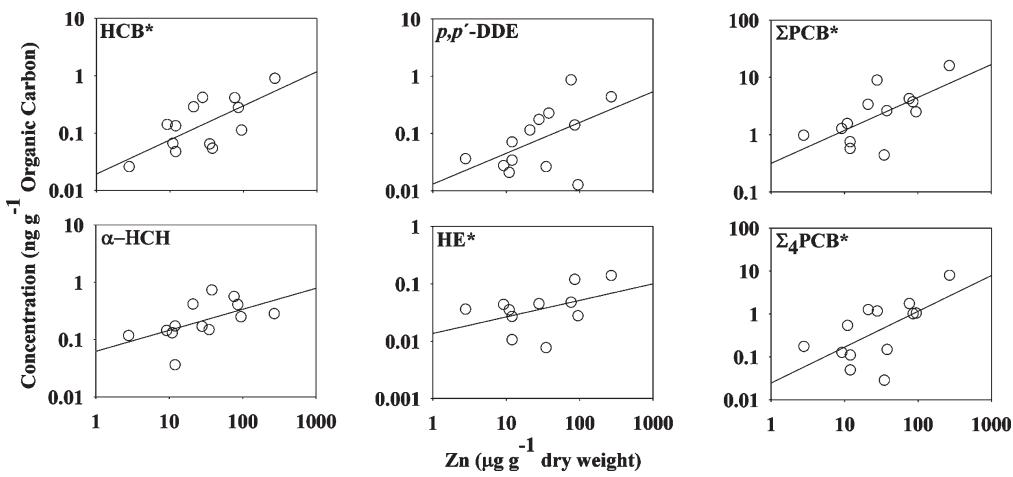


Figure 3. Surface sediment concentrations for selected chemicals and chemical groups along a gradient of seabird influence (quantified using sedimentary Zn as a tracer of seabird input) surveyed in 2006 ($n = 13$). $\Sigma_4\text{PCB}$ is the sum of CBs 118, 138, 153, and 180. Significantly positive slopes (95% confidence) are indicated with “*”.

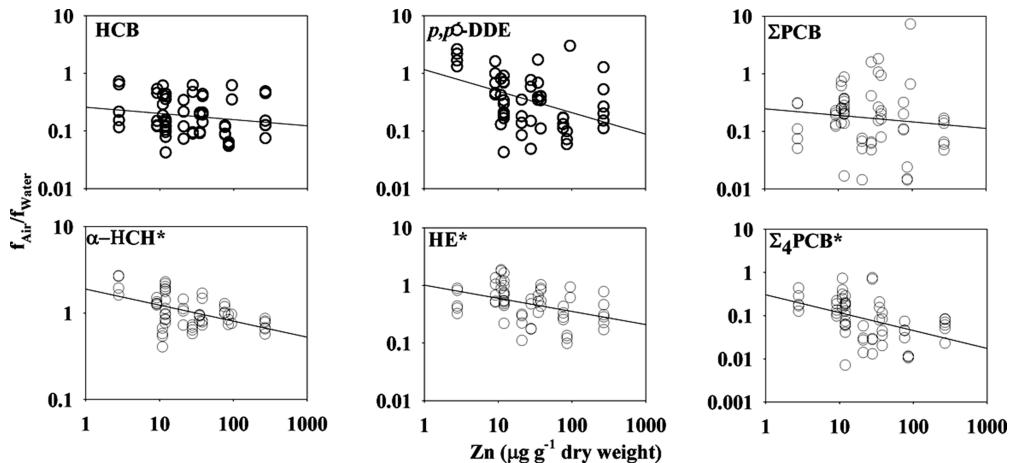


Figure 4. Fugacity quotients $f_{\text{Air}}/f_{\text{Water}}$ in freshwater ponds ($n = 13$) along a gradient of seabird input, quantified using sedimentary Zn as a tracer of seabird input. Data for all SPMDS (61 samples) are shown including replicates and samples from 2006 and 2007. $\Sigma_4\text{PCB}$ is the sum of CBs 118, 138, 153, and 180. Significantly negative slopes (95% confidence) are indicated with “*”.

Approximately 37% of ΣPCB in the water of CV 15 was contributed by PCBs characteristic of air (CBs 18, 31/28, and 52), compared to only 14% in CV 31. Similarly, in the sediments these four congeners comprised 20% and 6% of ΣPCB in CV 15 and CV 31, respectively. Higher K_{OA} congeners, including fulmar-characteristic CBs 118, 138, 153, and 180, contributed a greater percentage to ΣPCB in water in CV 31; congeners with Σ_{OA} values greater than that of CB 52 comprised 85% of ΣPCB in CV 31 as compared to 61% in CV 15. In sediments, over 40% of ΣPCB in CV 31 was CB 180, yet CB 180 comprised only 7% of ΣPCB in CV 15. The OCPs tell a similar story. From these results it is evident that where the seabird pathway has an opportunity to impinge on a given pond, the contaminant profiles in both water and sediments are more consistent with seabird sources and less consistent with air. This is the basis of the statistical assessment of source apportionment discussed below.

Organochlorine Contaminant Concentrations. As seabird input to the ponds increased, sediment concentrations of fulmartytic contaminants also increased (Figure 3). In the pond receiving the largest seabird input the concentration of ΣPCB

(15.8 ng g⁻¹ org. C) was approximately 16 times higher than in the pond receiving the least input, the concentration of $\Sigma_4\text{PCB}$ (7.9 ng g⁻¹ org. C) was 45 times higher, *p,p'*-DDE (0.4 ng g⁻¹ org. C) was 12 times higher, HCB (0.9 ng g⁻¹ org. C) was 35 times higher, α -HCH (0.3 ng g⁻¹ org. C) was 2 times higher, and HE (0.1 ng g⁻¹ org. C) was 4 times higher than in the pond receiving the least seabird input. Mean concentrations of organochlorine contaminants in water from the pond with the largest seabird input was 1.5–9.5 times higher than those of the pond with the least seabird input (Supporting Information, Figure S2).

In contrast, the fulmar colony appears to have little impact on local air concentrations. Cluster analyses (agglomerative hierarchical clustering; S-Plus 8.0), performed separately on both PCB and OCP contaminant concentrations and on contaminant profiles (a total of four analyses) of air samples from both sampling years showed no clustering of samples collected near or remote from the seabird colony. Contaminant profiles were computed as the concentration of each analyte divided by either ΣPCB or ΣOCP . Variables used for PCB and OCP cluster analyses included all constituents measured. Consequently, all

Table 1. Atmospheric Concentrations (pg m^{-3}) of Organochlorine Contaminants at Cape Vera Devon Island and Nearby Arctic Locations

location	sampling period	HCB	<i>p,p'</i> DDE	α -HCH	HE	ΣPCB^a	reference
Cape Vera	2006–2007	81.92 ± 17.04	0.34 ± 0.07	26.82 ± 4.56	2.78 ± 0.43	4.27 ± 1.52	the present study
	2007–2008	128.16 ± 13.13	0.98 ± 0.86	39.00 ± 5.06	2.24 ± 0.42	17.89 ± 11.52	
Alert	2005	52 ± 21	0.30 ± 0.26	13 ± 5.7	0.84 ± 0.29	4.9 ± 4.9	37
	2004	29 ± 21	0.44 ± 0.33	11 ± 6.1	0.65 ± 0.30	2.3 ± 1.3	
Alert	2000–2003		0.41		0.57		38
Kinngait	2000–2003		0.43		0.56		
Devon Island	2000–2001	79.03	5.80		(0.36) ^b		25 ^c
Eureka	2000–2001	94.84	(0.23)		0.44		
Alert	2000–2001	68.49	nd ^d		(0.18)		

^a ΣPCB = sum of 10 AMAP CBs (28, 31, 52, 101, 105, 118, 138, 153, 156, 180). ^b Concentrations in brackets are below the MDL. ^c Converted to units of pg m^{-3} using sampling rate of $0.52 \text{ m}^3 \text{ d}^{-1}$ sampler⁻¹, and deployment period of 365 days. ^d nd = not detectable

air samples from both sampling years were considered together as replicates.

The average air concentrations measured at Cape Vera in each of the two sampling years are comparable with literature values reported in the Canadian Arctic (Table 1). Thus, the thermodynamic implication that the fulmar colony at Cape Vera is a net source of contaminants to the air, either via the volatilization of contaminants from the nesting cliffs where seabird materials are deposited, or from the pond reservoirs which receive seabird input, was not directly detected. Given that the wind speeds at Cape Vera often exceed 85 km h^{-1} ,³⁶ the air compartment at the site must be well mixed, and it is possible that the seabird signal is diluted too quickly to be detected. This is consistent with the results of a salmon biovector model, which suggested that anadromous salmon biovectors would have a negligible effect on contaminant concentrations in air, but a significant impact on contaminant concentrations in water, sediment, invertebrates, and fish.⁴

Fugacities and Contaminant Fate. As seabird input increased, $f_{\text{Air}}/f_{\text{Water}}$ in the ponds decreased (Figure 4). For HCB and HE the average value of $f_{\text{Air}}/f_{\text{Water}}$ in the pond with the least seabird input was approximately 1.5-fold higher than that of the pond with the most. For α -HCH and *p,p'*-DDE the average value of $f_{\text{Air}}/f_{\text{Water}}$ was 3- and 6-fold higher, respectively, in the pond with the least seabird input. Values of $f_{\text{Air}}/f_{\text{Water}}$ in the control ponds were consistently within an order of magnitude of one and decreased with seabird input, whereas, values of $f_{\text{Air}}/f_{\text{Water}}$ in the pond that received the most seabird input, CV 31, were consistently less than one. Taken together these results indicate that seabird input shifted air–water partitioning in the ponds at Cape Vera to increasingly favor organochlorine chemical evasion at the air–water interface, especially at ponds receiving the highest seabird input.

In general, fugacities in the pond water were higher than or comparable to those in the sediments (see Supporting Information, Figure S3), which is consistent with aqueous phase contaminants from seabird materials being rinsed from the nesting cliffs into the ponds via the water inflow, contributing to elevated water fugacities. Higher fugacities in water imply favored partitioning from water into sediments for chemicals such as HCB, HE and ΣPCB , whereas *p,p'*-DDE, α -HCH, and ΣPCB appear to be closer to equilibrium between the water and sediments.

Source Apportionment. Mahalanobis distances between pond water PCB profile vectors and those of the seabird sources

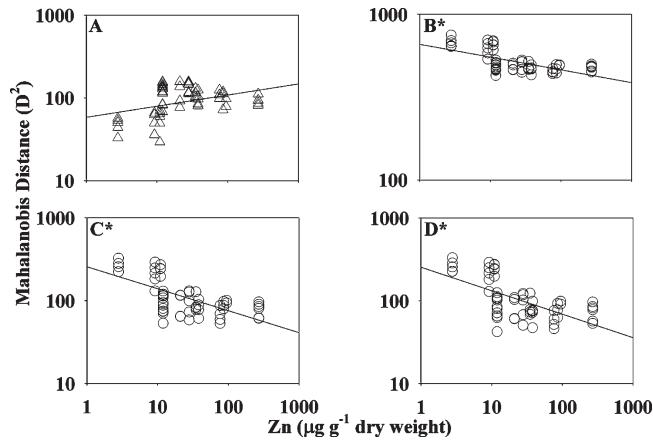


Figure 5. Mahalanobis distances between PCB profiles in pond water samples and those of possible sources: (A) atmospheric (distance to profiles in control ponds), (B) northern fulmar stomach oil, (C) carcasses, and (D) guano along a gradient of seabird input quantified using sedimentary Zn. Significantly negative slopes (95% confidence) are indicated with “*”.

decreased with increasing seabird input to the ponds (Figure 5). Simultaneously, the distances between the pond water PCB profile vectors and those of the control ponds, which reflect atmospheric input, increased with increasing seabird input. Thus, as seabird input increased, the PCB profile of water in the ponds became more similar to those of the seabird sources and less similar to those of the control ponds. Similar trends were also found for the distances between pond sediments and the sources (Supporting Information, Figure S4).

The relationship between seabird input and Mahalanobis distances was stronger for water samples than for sediment samples. This could be an artifact of the large number of replicates for water samples. However, the transfer of seabird materials and associated PCBs into the pond water at Cape Vera is probably a more direct, physical process with less opportunity for the operation of fractionating processes involving physicochemical properties of the PCB congeners. In contrast, sediment contaminant concentrations reflect, in whole or in part, water–sediment partitioning and the production of particulate organic matter in the ponds, both of which provide opportunities for partitioning that selects for physicochemical properties of chemicals (e.g., K_{OW}).³¹) Thus, the transfer of seabird-derived PCBs

from water to sediments will not be equally efficient for all congeners and the PCB signature in sediments will be less consistent with those of the sources than the water profile. Further research is required to assess if Mahalanobis distances computed between sediments and sources meet the assumption that chemical transfer is independent of the PCB congener. However, the use of source apportionment statistical analyses for seabird biovectors, and indeed for biovectors in general, shows promise that warrants additional study.

Using four approaches, we have assessed the relative effects of atmospheric deposition and seabird biovectors on contaminants in the ponds at Cape Vera. Seabirds dominate the supply of contaminants to ponds directly receiving runoff from the nesting cliffs and provide a secondary source in this otherwise remote coastal area. As a result of food web transfers, the northern fulmar colony at Cape Vera favors the transferring of certain organochlorine contaminants (i.e., CBs 118, 138, 153, and 180, HCB, *p,p'*-DDE, α -HCH, and HE) for which the concentrations and fugacities in affected ponds become elevated. In the case of these contaminants, volatilization from pond water to air becomes favored and the contaminant profiles in the water and sediments increasingly reflect seabird sources. These results imply that biovector pathways need to be considered explicitly in chemical transport modeling aimed at projecting ecosystem exposures, particularly along Arctic coastlines supporting numerous large seabird colonies. The focusing of contaminants into Arctic receptor sites is of particular relevance to resident and migratory wildlife whose survival depends on obtaining food at "oases" sustained by seabird-derived nutrients, which presently provide a simultaneous dose of contaminants.

■ ASSOCIATED CONTENT

S Supporting Information. Details: passive sampling and seabird input tracers. Tables: pond GPS coordinates, fugacity equations, PCB/OCP physicochemical properties. Figures: OCP profiles in ponds and sources, PCB/OCP concentrations (water), fugacities in ponds, Mahalanobis distances (sediments). This material is available free of charge via the Internet at <http://pubs.acs.org>.

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