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Seabird-Transported Contaminants Are Reflected in the Arctic Tundra, But Not in Its Soil-Dwelling Springtails (Collembola)

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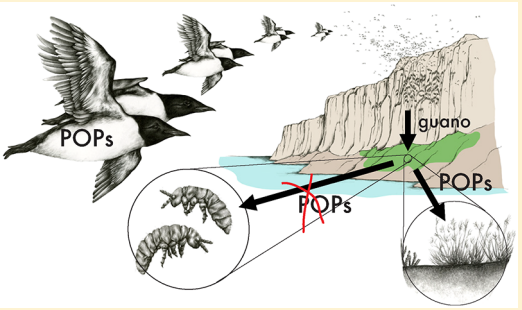
Supporting Information

ABSTRACT: Arctic-breeding seabirds contain high levels of many anthropogenic contaminants, which they deposit through guano to the tundra near their colonies. Nutrient-rich soil in vicinity to seabird colonies are favorable habitats for soil invertebrates, such as springtails (Collembola), which may result in exposure to seabird-derived contaminants. We quantified a wide range of lipid-soluble and protein-associated environmental contaminants in two springtail species (*Megaphorura arctica* and *Hypogastrura viatica*) and their respective habitats (soil/moss) collected underneath seabird cliffs. Although springtails are commonly used in laboratory toxicity tests, this is the first study to measure concentrations of persistent organic pollutants (POPs) and mercury (Hg) in springtails from the field, and to study biotransportation of contaminants by seabirds to soil fauna. We categorized the sites a priori as of low, medium, or high seabird influence, based on the seabird abundance and species composition. This ranking was reflected in increasing $\delta^{15}N$ values in soil/moss and springtails with increasing seabird influence. We found clear indications of seabirds impacting the terrestrial soil environments with organic contaminants, and that concentrations were higher in soil and moss close to the bird cliff, compared to farther away. However, we did not find a relationship between contaminant concentration in springtails and the concentrations in soil/moss, or with level of seabird influence. Our study indicates a low uptake of contaminants in the soil fauna, despite seabird-derived contamination of their habitat.



INTRODUCTION

Numerous seabird species migrate to the Arctic in early spring, where they gather in large colonies to breed.[1−3] Areas near the seabird colonies receive large amounts of litter, including guano, egg shells, regurgitated stomach oils, feathers, food remnants, and carcasses.[4,5] The biological productivity of Arctic tundra is usually limited by nitrogen availability, and fertilization from seabirds strongly stimulates the growth and diversity of vegetation.[6−8] The tundra below seabird colonies also supports a high diversity of soil invertebrates,[9] of which springtails (Collembola) commonly make the highest contribution to total biomass and biodiversity.[4,10,11] Springtails are globally distributed microarthropods, which play an important role in the decomposition of organic matter and the cycling of nutrients in soil ecosystems. They can also affect the bioavailability of contaminants to other organisms indirectly by impacting the soil characteristics.[12,13] Springtails are also important in the transfer of energy between food webs above and below ground, by being prey to small arthropods[13,14] and terrestrial bird species.[15]

In the High Arctic, springtail density and biomass are up to 20 times higher in tundra areas below bird cliffs, compared to areas not influenced by seabirds.[11] However, in addition to receiving favorable nutrients from seabird litter (e.g., guano), springtails may be exposed to anthropogenic contaminants, as seabirds function as biological vectors of industrial and agricultural pollutants biomagnified through the marine food web.[16−18] Seabirds feed relatively high in the marine food chain,[19] and may thus accumulate high levels of several persistent organic pollutants (POPs) and some inorganic contaminants.[20,21] Examples of contaminants quantified in several Arctic seabird species include polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB), various organochlorine pesticides,[22−25] polybrominated diphenyl ethers (PBDEs),[26] per- and polyfluoroalkyl substances (PFASs),[27] and mercury (Hg).[28] The Arctic is geographically remote from major pollution sources, but contamination takes place due to long-range atmospheric or ocean transport,[29−31] or actively through biotransport.[18] Biotransport of contaminants by seabirds has

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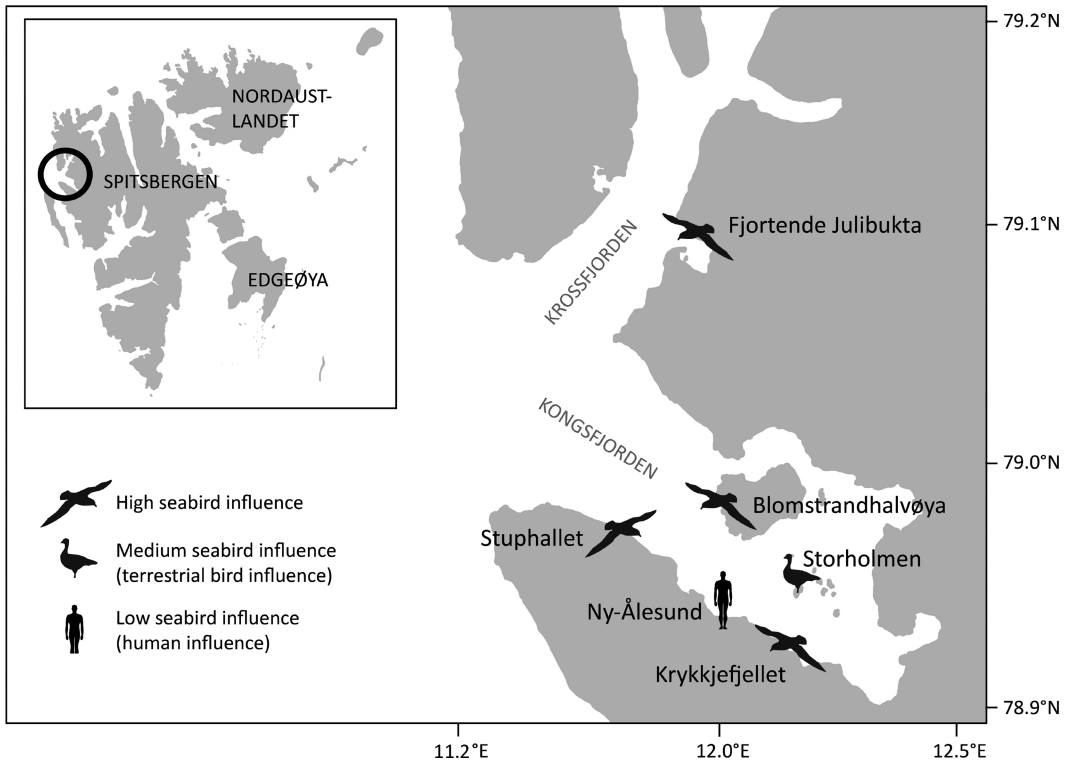

Figure 1. Study sites in the area of Krossfjorden and Kongsfjorden, Svalbard archipelago, Norwegian Arctic. Symbols indicate the a priori ranking of high, medium, and low seabird influence, based on seabird species present and their abundance.

mainly been studied in freshwater lakes and ponds in the Norwegian and Canadian Arctic, for example, refs 16, 18, 32, and 33. Although less studied, tundra near bird cliffs show decreasing contamination concentrations along a gradient of decreasing seabird influence.[33,34] Most studies on Arctic environmental pollution focus on the marine ecosystem, thus leaving significant knowledge gaps concerning the contamination of terrestrial environments.[35,36] To date, terrestrial studies have investigated the contamination in abiotic matrices, vegetation, and vertebrates.[6,7,37−39] However, there are to date no studies on contaminant occurrence in soil fauna, such as springtails.

The overall aim of the present study was to determine whether springtails in High Arctic tundra ecosystems are exposed to seabird-derived contaminants. We studied the two species, *Megaphorura arctica* and *Hypogastrura viatica*, which are typical for richly vegetated areas near seabird colonies but occupy different microhabitats (see below), suggesting that they might be exposed to soil contamination differently. Guano and other seabird litter are enriched in the heavier stable isotope of nitrogen ^{15}N, relative to ^{14}N, and thus have an enriched stable isotope ratio $\delta^{15}N$,[16,40] compared to terrestrial nitrogen sources of the tundra, that is, the nitrogen fixed from the atmosphere. We assume that different populations of the same springtail species feed at the same trophic level, and thus $\delta^{15}N$ was not used to determine relative trophic position in the present study, but as a proxy for seabird influence at their specific sites. We expected that seabird influence would depend on the distance from the seabird colony, with contaminant concentrations in springtails being correlated with that of their habitats, and that these correlations would be reflected in the $\delta^{15}N$ values. Biotransformation and selective elimination is assumed to be low in invertebrates, as their contaminant pattern resemble that of their ambient environment.[41] Therefore, if seabirds were the source of contamination, the contaminant pattern in habitats and springtails were expected to resemble that in seabirds.

MATERIALS AND METHODS

Study Species. *H. viatica* (1.9 mm body length) and *M. arctica* (3.5 mm body length) have very patchy distributions, forming conspicuous mass occurrences easily observed in the field, which allowed us to sample the amount of material required for analyses. Both species are often found near seabird colonies,[11,42] with *M. arctica* inhabiting the loose upper soil, whereas *H. viatica is* typically active in wet moss and on the surface of terrestrial algae.

Study Sites. Springtails and the substrate of their respective habitats were collected from six study sites in two fjord systems, Krossfjorden and Kongsfjorden in Spitsbergen, Svalbard (79°N, 11−12°E) (Figure 1). Based on the seabird species presence and their abundance at each site, the sites were a priori ranked as having low, medium, or high seabird influence (Table 1). At Stuphallet, *M. arctica* was collected at 0 m distance from the bird cliff, and *H. viatica* was sampled at 250 m distance. At Blomstrandhalvøya, *H. viatica* was sampled at 150 and 400 m distance from the bird cliff. We obtained only one sampling at the remaining study sites.

Sampling. In June 2016, samples estimated to comprise well above 10 000 individual springtails each, were collected from the different sites for chemical analyses. *H. viatica* was found on pieces of moss or on layers of cyanobacteria on the soil surface. They were sampled by gently shaking the substrate with springtails in a stainless-steel sieve (mesh size: ø 200 mm) over a plastic container, allowing the springtails to fall into the container. *M. arctica* was sampled by similarly sieving the top

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Table 1. Description of Study Sites[a]

Ranking of seabird influence	Study site	Description
LOW 	Ny-Ålesund	Ny-Ålesund has low seabird influence, and is clearly influenced by human activity, due to its vicinity of the research settlement. Samples from Ny-Ålesund were collected close to a small lagoon, with a shallow stream of water flowing from the research station.
MEDIUM 	Storholmen	Storholmen is a low lying island with a mixed influence by seabirds and terrestrial birds. Numerous common eiders (*Somateria mollissima*), which is a benthic marine feeder, and the herbivore barnacle geese (*Branta leucopsis*), breed on this island. Both species occupy relatively low trophic positions with a mean $\delta^{15}N$ in muscle=3.2 and 6.4‰, respectively [19, 79].
HIGH 	Krykkjefjellet	The seabird cliff Krykkjefjellet has a mixed seabird colony of mainly black-legged kittiwake (*Rissa tridactyla*), and also some brünnich's guillemot (*Uria lomvia*) and black guillemot (*Cepphus grylle*). The $\delta^{15}N$ of muscle in black-legged kittiwake ranges 12.7-14.9‰, brünnich's guillemot ranges 11.8-15.4‰ and black guillemot ranges 13.4-14.9‰ [19, 22, 50, 80].
	Stuphallet	The seabird cliff Stuphallet has a relative high density of Atlantic puffin (*Fratercula arctica*), but also individuals of black guillemot. The Atlantic puffin has a $\delta^{15}N$ values in muscle of 12.2‰ [81].
	Blomstrandhalvøya	The seabird cliff on the island Blomstrandhalvøya has a relative high density of black-legged kittiwake and a moderate density of northern fulmars (*Fulmarus glacialis*). The $\delta^{15}N$ of muscle in northern fulmars range between 14.0-15.3‰ [19, 80].
	Fjortende Julibukta	The seabird cliff in the bay of Fjortende Julibukta has relatively high densities of brünnich's guillemot and black-legged kittiwake. There are also some individuals of barnacle goose feeding on the grass below the colony.
		The seabird species present at Krykkjefjellet, Stuphallet, Blomstrandhalvøya and Fjortende Julibtukta are mainly pelagic-feeding and occupy therefore relatively high trophic positions in the Arctic marine food web, feeding on crustaceans and fish [82, 83].

[a]Links to refs 19, 22, 50, 79, 80, 81, 82, and 83.

layer of litter/soil. To avoid dehydration, pieces of moist moss were placed in the container until handling in the laboratory. In the laboratory, springtails were separated from debris by placing the sample on a piece of aluminum foil in a clean container and collecting the animals as they moved away from the foil with debris. Springtails were transferred to glass containers and stored at −20 °C until chemical analyses. Substrates (mixture of soil and/or moss) of the springtails' habitat were sampled using a stainless-steel knife, wrapped in aluminum foil, and stored in a zip-lock bag at −20 °C until chemical analyses. Dry matter (%) in substrates was determined gravimetrically, drying samples at 105 °C for 24 h. An overview and description of collected samples is presented in the Supporting Information (SI) (Table S1).

Sample Preparation and Chemical Analysis. Substrate samples were homogenized in a food processor (Hugin, Punica minihakker) and sifted to remove rocks. Springtails were homogenized using a mortar and pestle. Substrate and springtails were analyzed for a wide range of lipid-soluble contaminants including PCBs, organochlorine pesticides and metabolites, PBDEs, and the protein-associated PFASs at the Norwegian Institute for Air Research (NILU) in Tromsø, Norway. Total Hg was analyzed at NILU in Kjeller, Norway. Lipid-soluble analytes included the PCB congener -28, -31, -52, -99, -101, -105, -118, -138, -153, -180, -183, -187, -194, and the PBDE compounds BDE-28, -47, -99, -100, -138, -153, -154, -183, -196, -197, -206, -207, and -209. Pesticides and metabolites included HCB, α-, β-, and γ-hexachlorocyclohexane (HCH), the dichlorodiphenyltrichloroethanes *o,p′*-DDT, *p,p′*-DDT; the dichlorodiphenyldichloroethanes *o,p′*-DDD, *p,p′*-DDD; the dichlorodiphenyldichloroethylenes *o,p′*-DDE; *p,p′*-DDE; *cis*-chlordane (c-CD); *trans*-chlordane (t-CD); *cis*-nonachlor (c-NC); *trans*-nonachlor (t-NC); oxychlordane (Oxy-CD); and mirex. PFAS analytes included perfluorobutan sulfonate (PFBS), perfluorohexanesulfonate (PFHxS), perfluoroheptanesulfonate (PFHpS), linear and branched perfluorooctanesulfonate (∑PFOS), perfluorononanesulfonate (PFNS), perfluorodecanesulfonate (PFDcS), perfluorohexa-

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noic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA), and perfluorotetradecanoic acid (PFTeDA).

As this is the first time these contaminants have been analyzed in springtails collected in the field, the methodology was modified slightly from established methods of extraction and cleanup, and differed between lipid-soluble contaminants, PFASs, and Hg, as well as between matrices. Full descriptions of methodologies and quality assurance are presented in the SI (S1. Determination of lipid-soluble contaminants, S2. Determination of PFASs, S3. Determination of Hg, and S4. Quality assurance, respectively). We did not obtain sufficient biomass to conduct the analyses of all the contaminant groups for all study sites and matrices, as indicated in tables and figures.

Stable Isotope Ratios of Carbon and Nitrogen. Homogenized substrate samples were freeze-dried in a Leybold-Heraeus GT2 Freeze-Dryer, before being rehomogenized using a pestle. Substrate and springtails were analyzed for stable isotopes of nitrogen and carbon, and stable isotope ratios (δ^{13}C and δ^{15}N) were calculated by the Institute for Energy Technology (IFE), Kjeller, Norway, as described in the SI (S5. Stable isotope analysis).

Data Treatment. All statistical analyses were performed using the statistical software R (version 3.3.2, the R Foundation for Statistical Computing 2016). Contaminant concentrations in springtails is reported as wet weight data (pg/g w.w.) for organic contaminants and dry weight (pg/g d.w.) for Hg. When comparing concentrations of lipid-soluble compounds between the springtail species, lipid weight data (pg/g l.w.) were used along with w.w. to remove concentration variation caused by lipid content. Dry weight data (pg/g d.w.) is reported for substrate samples.

When the assumptions of normal distribution and heteroscedasticity were not met, as assessed with Shapiro-Wilk's test and Levene's test, respectively, data was logarithmically or square root transformed. When possible, all groups were treated the same before statistical analysis. If normal distribution and homogeneity of variance were still not obtained, nonparametric tests were applied. The significance level was set to $p = 0.05$, and p-values were two-tailed.

A test of differences in stable isotope ratios were only performed on data from Ny-Ålesund and Fjortende Julibtukta, as the remaining sites had too low a sample size ($n < 2$). Relationships among stable isotope ratios in substrate and springtails were examined with linear regression.

Two-sample Student's t test was applied when data points were normal distributed and had equal variance, and the Welch two-sample t test[43] in the case of heteroscedasticity.

For both substrate and springtails, compounds quantified above the limit of detection in a minimum of 65% of the samples were included in multivariate statistical analyses. The following compounds were therefore excluded from both matrices: PCB-105, -183, -194; PBDE-28, -99, -100, -138, -153, -154, -183, -196, -197; o,p'-DDT; p,p'-DDT; o,p'-DDD; p,p'-DDD; o,p'-DDE; mirex; PFBS; PFHxS; PFHpS; PFNS; PFDcS; PFHxA; PFHpA; PFDA; PFUnDA; PFTrDA; PFTeDA. PCB-28/31, -101, -187; PBDE-47, -207; β-HCH; PFDoDA were eliminated only from substrate samples, and PBDE-206, $\sum$PFOS, PFOA, PFNA were eliminated only from springtail samples. Values below the respective LOD of contaminants included in the statistical analyses were replaced with randomly selected numbers between 0.5 × LOD and LOD in a uniform distribution. A total of 9.7% of the contaminant data were replaced, of which 35 of 273 data points were for substrate, and 22 of 312 data points were for springtails. All concentrations of contaminants detected are presented in the raw data (SI Table S2). Multivariate statistics were conducted using the R-package *vegan*.[44] Only *H. viatica* was included in the multivariate statistics, as only a total of two samples were obtained for *M. arctica,* in which only Hg and PFASs were quantified due to matrix limitations. Principal component analyses (PCAs) were conducted to analyze the variations in concentrations among and within study sites and matrices, and the relationships with δ^{13}C, δ^{15}N, and lipid content of springtails. Redundancy analysis (RDA) was performed to test significance of the explanatory variables to account for the variation in concentrations. Only lipid-soluble POPs were included in the PCAs, as uncertainties were associated with the lack of standardized pretreatment of substrate samples for Hg analysis and due to low recovery of PFASs in substrate. The analytes were centered (mean = 0), but not scaled to unit variance. For both matrices, the explanatory variables included in the PCAs were δ^{13}C and δ^{15}N. For *H. viatica*, lipid content (%) and contaminant exposure from the ambient habitat were added as additional explanatory variables. The latter was represented by log transformed d.w. concentrations of p,p'-DDE, *trans*-nonachlor, and PCB-153 quantified in substrate. In the PCA for substrate (SI Figure S1), we chose one contaminant vector from each of the PCAs main three clusters of vectors with similar structure in their variance among the samples. These representative compounds were included in the PCA for *H. viatica* to examine the relationship between the contaminants in springtails and their habitat. The same explanatory variables were included as active variables in the RDA, with the exception of lipid content, which was entered as a covariate for *H. viatica*. Significant explanatory variables were forward selected using permutation tests.

RESULTS AND DISCUSSION

Seabird Influence on the Tundra. We used δ^{15}N values of the springtails and the substrate as a proxy for seabird influence and found a decreasing gradient from sites of high to medium to low seabird influence (Fjortende Julibukta > Blomstrandhalvøya (150 m) > Stuphallet (0 m) > Blomstrandhalvøya (400 m) > Krykkjefjellet > Stuphallet (250 m) ≈ Storholmen > Ny-Ålesund) (Table 2). Lower δ^{15}N values in substrate and springtails from Ny-Ålesund with low seabird influence compared to Fjortende Julibukta with high (Welch two-sample t test, $p = 0.0001$; two-sample t test, $p =$ 0.8e-05, respectively), confirms that δ^{15}N is a suitable proxy for seabird influence. In addition, the decreasing trend in δ^{15}N with increasing distance from the colony, are in accordance with our a priori ranking of the sampling sites based on bird species and abundance, and fit with previously reported results.[6−8] The low δ^{15}N values in springtails from Ny-Ålesund with low seabird influence are closer to the δ^{15}N values measured in springtails from other geographical regions, for example, refs 45 and 46. The δ^{15}N in springtails increased with increasing δ^{15}N in substrate across all sites (linear regression, $R^2 = 0.96$, $p < 0.0001$), thus, marine-derived nutrients from seabirds are reflected in both the springtails and their habitat.

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Table 2. Contaminant Concentrations in Substrate (Soil/Moss) (pg/g d.w.) and Springtails (Hg in pg/g d.w., Rest of Compounds in pg/g w.w.), Stable Isotopes Ratios ($\delta^{13}C$ and $\delta^{15}N$), Dry Matter in Substrate, and Lipid Content in Springtails[a]

Study site	Ny-Ålesund			Storholmen	Krykkje-fjellet	Stuphallet		Blomstrandhalvøya		Fjortende Julibukta			
Substrate sample-ID	SUB1	SUB2/3	SUB4	SUB5	SUB6	SUB7	SUB8	SUB9	SUB10	SUB11	SUB12	SUB13/14	SUB15
Compounds pg/g d.w.													
Hg	55020	22078	31796	115219	87244	36024	26146	93965	14895	12013	52633	34061	56152
∑PFASs	2050	2017	3891	372	2671	1173	98.7	832	1090	363	442	219	497
∑PCBs	353	211	323	219	10284	20166	<LOD	386	230	219	660	220	1416
∑PBDEs	877	434	3194	19,2	507	456	476	252	811	117	27.7	69.2	334
∑DDTs	9440	1043	3548	349	351	6256	314	8.82	429	11.2	21.9	23.3	30.6
HCB	307	147	498	276	1385	386	274	325	117	162	558	191	865
∑HCHs	227	211	396	111,9	415	355	58.5	207	15.5	14.5	52.3	14	141
∑Chlordanes	173	138	280	364	1510	3865	330	248	188	142	227	91.6	447
Mirex	<LOD	<LOD	<LOD	<LOD	143	244	<LOD	<LOD	24.3	<LOD	23.3	<LOD	28.0
$\delta^{13}C$ (‰)	-25.4	-25.4	-26.3	-26.3	-27.0	-23.0	-27.1	-24.8	-17.8	-23.5	-25.9	-24.8	-23.3
$\delta^{15}N$ (‰)	3.83	5.47	3.97	7.96	10.8	19.9	10.2	24.4	18.0	26.0	28.6	29.9	30.9
Dry matter (%)	39.3	25.9	17.0	16.7	35.3	67.4	13.3	21.3	35.5	39.9	24.5	73.4	20.0
Springtail sample-ID	MA1	HV2	HV3	HV4	HV5	MA6	MA7	HV8	HV9	HV10	HV11	HV12	HV13	HV14	HV15
Compounds pg/g d.w.															
Hg	49752	12988	18228	11021	15577	88644	n.a.	19830	13747	13081	13012	10655	10616	9732	9309
pg/g w.w.															
∑PFASs	474	1297	2193	2851	n.a.	271	n.a.	n.a.	<LOD	395	260	402	197	117	196
∑PCBs	1936	2739	1630	1502	1193	n.a.	n.a.	1333	843	724	1269	860	735	844	976
∑PBDEs	2392	481	435	4138	54.2	n.a.	n.a.	1602	<LOD	353	932	1152	324	16	589
∑DDTs	493	549	439	1003	264	n.a.	n.a.	263	60.6	41.9	649	174	165	360	180
HCB	603	3052	3355	2953	5000	n.a.	n.a.	4478	1896	2353	3252	3139	2171	2960	2956
∑HCHs	172	487	276	240	139	n.a.	n.a.	177	209	210	171	171	138	116	159
∑Chlordanes	386	602	527	530	831	n.a.	n.a.	1020	618	733	407	367	335	120	373
$\delta^{13}C$ (‰)	-19.6	-21.8	-23.6	-22.0	-25.6	-28.0	-27.7	-28.0	-23.1	-25.0	-23.0	-23.2	-23.4	-23.3	-23.0
$\delta^{15}N$ (‰)	7.11	3.61	3.60	3.84	7.91	12.9	17.7	6.40	22.7	14.5	27.0	27.2	27.2	27.1	26.9
Lipid (%)	3.39	10.1	8.87	9.23	4.47	n.a.	n.a.	7.03	9.85	7.83	10.9	12.1	10.6	8.21	12.7

[a]Springtail sample-ID reflect species; MA = *M. arctica* and HV = *H. viatica*. Abbreviations: n.a. = not analyzed, n.d. = not detected, <LOD = below the limit of detection, d.w. = dry weight, w.w. = wet weight, Hg = mercury, PFASs = per- and polyfluoroalkyl substances, PCBs = polychlorinated biphenyls, PBDEs = polybrominated diphenyl ethers, DDTs = dichlorodiphenyltrichloroethane with metabolites, HCB = hexachlorobenzene, HCHs = hexachlorocyclohexanes.

The highest $\delta^{15}N$ were 27‰ in springtails and 31‰ in substrate, which both markedly exceed the 15‰ previously found in seabird tissues from the same area.[47−50] Similarly, high $\delta^{15}N$ values have also been found in earthworms living in penguin rookery soil (25.4‰, SD = 1.2).[51] Even higher $\delta^{15}N$ values than in our substrate have been found in soil in other ecosystems associated with seabird colonies, both in the Antarctic,[52] and in the Arctic.[5,34] The higher $\delta^{15}N$ in the terrestrial environment than in the seabirds themselves, is caused by fractionation during microbial transformation of uric acid to ammonia (NH_3) followed by ammonia volatilization.[51,53,54]

We did not find any difference in $\delta^{13}C$ between sites of low and high seabird influence (Ny-Ålesund and Fjortende Julibukta) for springtails or substrate (*t* test, $p = 0.08$ and $p = 0.1$, respectively). Thus, $\delta^{13}C$ is not a suitable indicator of seabird influence on terrestrial ecosystems, consistent with previous findings,[6,7,38,55] as the main carbon source to springtails is likely from CO_2 uptake from air by vegetation. The $\delta^{13}C$ values in most of our samples are consistent with the average terrestrial $\delta^{13}C$ value found in C3 photosynthesizing plants of −27.8‰.[56]

Contaminant Concentrations and Patterns in Substrate. Our statistical analyses were limited owing to lack of true replicates. One substrate sample (soil and/or moss) was collected along with each springtail sample at the different sites. The most apparent finding in substrate was higher concentrations of chlordanes, HCB, high-chlorinated PCBs, and DDTs (mainly *p,p*-DDE) in substrate collected directly beneath the bird cliff at Stuphallet (SUB7) compared to a 250 m distance from the seabird colony (SUB8) (Table 2). All PCB congeners were below the limit of detection in SUB8 (SI Table S2), whereas ∑PCB reached 20.2 ng/g d.w. at SUB7. ∑DDTs was almost 20 times higher, and chlordanes more than 10 times higher close to the bird cliff, compared to further away (Table 2). The findings at Stuphallet indicate that seabirds influence the tundra in close vicinity to their colony. At Blomstrandhalvøya, such a strong difference between soil sampled close to the birth cliff and farther away was not found. However, the sample closest to the cliff was from a distance of 150 m, and not directly beneath the cliff as in Stuphallet.

∑PCBs in 11 substrates sampled at larger distances from bird cliffs ranged between 0.2 and 1.4 ng/g d.w. (Table 2), and were within the same range as concentrations in soil samples from the coastal area of Kongsfjorden.[57] The two remaining

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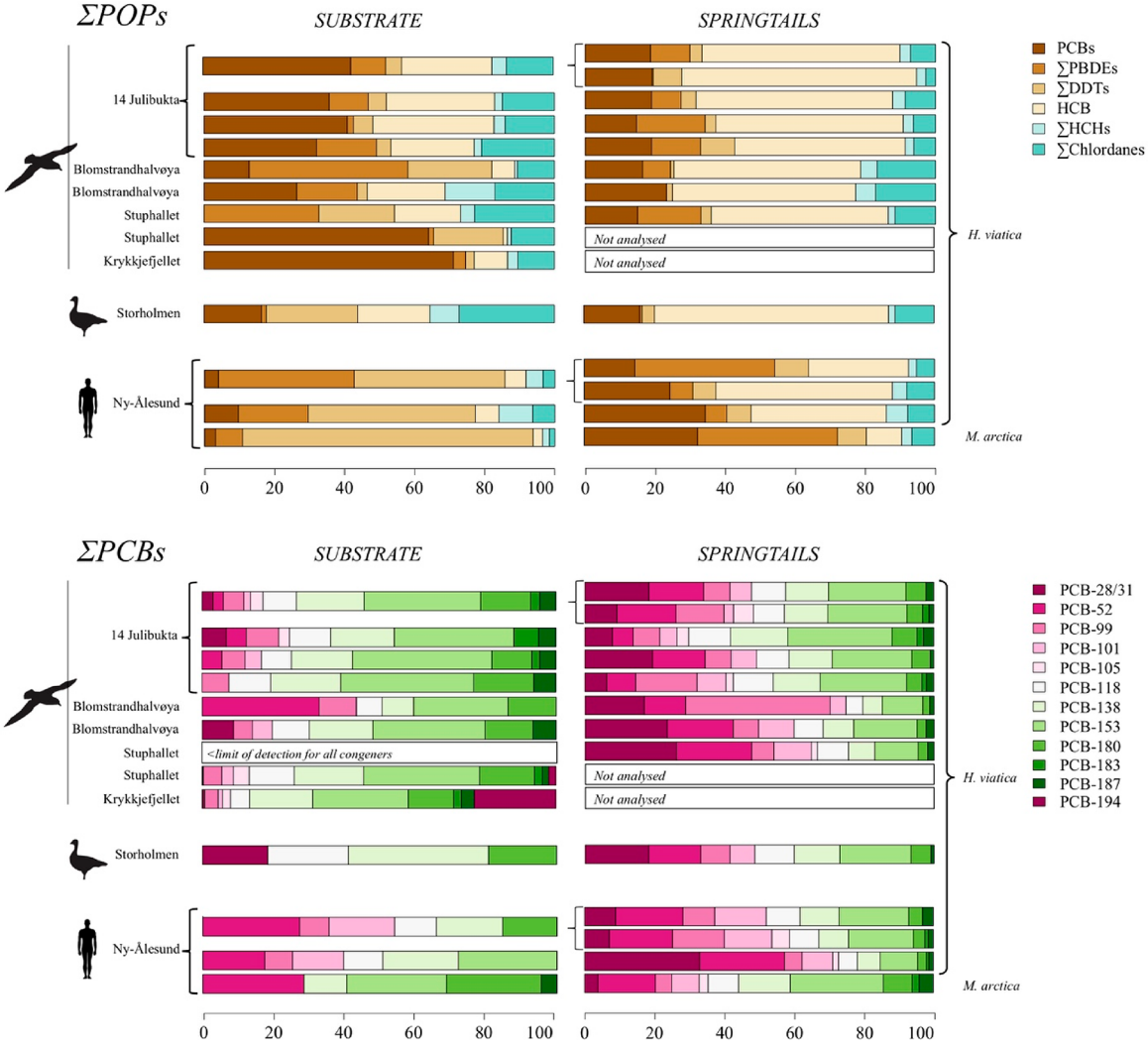

Figure 2. Relative proportions of organic contaminant groups to the sum (∑) of all groups (top) and relative proportions of polychlorinated biphenyls (PCB) congeners to ∑PCBs (bottom) in substrates (left panel) and springtails (right panel). Samples of substrates are presented horizontal to their respective springtail sample. In Fjortende Julibukta and Ny-Ålesund, two springtail samples were collected along with one substrate sample, indicated on the figure. Abbreviations: PBDEs = polybrominated diphenyl ethers, DDT = dichlorodiphenyltrichloroethane and its metabolites, HCB = hexachlorobenzene, HCH = hexachlorocyclohexanes, chlordanes = chlordane and its metabolites.

substrate samples, collected directly beneath bird cliffs, indicated increased levels due to impact by seabirds: in addition to SUB7 mentioned above, SUB6 had a ∑PCB level of 10.3 ng/g d.w. at Krykkjefjellet. Similar to PCBs, the highest ∑chlordane levels in substrate were also found at these two sampling sites (Table 2). The remaining samples had a ∑chlordane level below 1 ng/g d.w., similar to concentrations in lichens and vegetation elsewhere in the Arctic.[58] When detected, PFOS was the dominating PFAS with concentrations up to 3.8 ng/g d.w. in substrate, outranking most lipophilic pollutants (SI Table S2 of raw data). PFOS show highest levels in the location characterized by human impact (Ny-Ålesund), and in the samples collected directly beneath the bird cliffs at Krykkjefjellet and Stuphallet (sample ID SUB6, SUB7). It appears as even though seabirds may be efficient vectors for PFOS to the tundra, human activities are important sources of PFAS.[39]

There was no clear relationship between seabird influence (δ^{15}N) and substrate concentrations of Hg (Table 2). Although one study found Hg concentrations to be higher in seabird affected sites compared to nonaffected sites,[59] several studies report inconsistent trends between Hg and seabird transport.[38,60,61] Effects of seabird influence might be masked by the relative proportion of moss in the sample material, as the Hg concentration increased with the proportion of moss in the sample material (linear regression, n = 13, R^2 = 0.76, p-value = 0.0001) (SI Figure S3 and Table S2). Mosses are often used as bioindicators of heavy metal pollution,[62] as bryophyte tissues accumulate metals due to high cation exchange capacities, no cuticle and a high surface-area-to-volume ratio.[63,64]

When excluding PFASs due to low analytical recovery, the dominating contaminant groups in substrate from sites of medium and high seabird influence were PCBs, HCB, and chlordanes, whereas samples with low seabird influence were dominated by DDTs (mainly p,p-DDE) (Figure 2). A figure of relative concentrations including PFASs are presented in SI Figure S4, along with an explanation of its exclusion in Figure 2 (SI Figure S6. Exclusion of PFAS). In substrate sampled directly beneath the colonies in Krykkjefjellet and Stuphallet, PCBs made up a higher fraction of the total contaminant load, and were dominated by congeners 138 and 153, which is

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similar to results in seabirds.[22,48,50] The high relative contribution of highly chlorinated PCBs was also evident in substrate samples from other sites with high seabird influence (Blomstrandhalvøya, Fjortende Julibukta), despite the concentrations being lower than in Krykkjefjellet and Stuphallet. This is likely due to the samples being collected further from the bird cliffs, where guano deposition is lower. Sites of low seabird influence had a pattern that was indicated to be influenced by atmospheric long-range transport and deposition, with higher proportion of low-chlorinated PCBs.[30] This is consistent with previous findings[18] of higher proportions of higher chlorinated PCBs in samples taken from a lake highly influenced by seabirds, compared to a lake with low seabird influence.

Contaminant Concentrations and Patterns in Springtails. As each springtail sample consisted of a huge number of randomly sampled animals, we assume each sample represent a good estimate of contaminant concentrations (Table 2). We found no apparent trend between increasing seabird influence and level of contamination in springtails, as ∑PFASs, ∑HCHs, DDTs, and PCBs were higher in springtails from Ny-Ålesund, the site of low seabird influence, compared to those with higher seabird influence. HCB was highest in Storholmen with medium seabird influence, and several samples from Ny-Ålesund (low seabird influence) exceeded those from sites of high seabird influence. There was no clear relationship between distance to bird cliffs and levels of contamination in springtails (sample HV9 and HV10 from Blomstrandhalvøya, taken at a 150 and 400 m distance, respectively). However, we were not able to compare samples from Stuphallet, as the number of *M. arctica* found at 0 m from the colony was not sufficient in biomass for chemical analyses.

Neither was any trend between Hg concentrations in *H. viatica* and seabird influence apparent, as the concentrations at Ny-Ålesund and Storholmen with low and medium seabird influence exceeded several of the springtail samples from sites of high seabird influence (Table 2). The relatively high Hg concentrations found in springtails in Ny-Ålesund is likely caused by historical coal mining operations and fossil fuel combustion.[65,66]

HCB contributed the highest relative proportion to the sum of organic contaminants in springtails, followed by PCBs and PBDEs or chlordanes (Figure 2). A high relative abundance of HCB is consistent with what has been observed in other terrestrial herbivores (although vertebrates) from the Canadian Arctic,[67] and is likely to originate from the air−plant−animal pathway, which is suggested to be the main pathway of POPs to Arctic terrestrial food chains.[67,68] β-HCH was less prevalent than α- and γ-HCH in springtails, including those from seabird influenced sites, which is similar to the HCH pattern in air[30,68] (SI Table S3). The low contribution by β-HCH was inconsistent with expectations as β-HCH dominates the HCH load in most seabirds,[23,41] and the pattern is likely to reflect uptake of HCHs from air. The higher biomagnification factor for β-HCH compared to α- and γ-HCH[69] could explain why β-HCH was detected in springtails.

Unlike substrate, lower chlorinated congeners dominated the PCB pattern in springtails (Figure 2), which is surprising as they have a relatively high lipid-content (3.4% in *M. arctica* and a mean of 9.4% in *H. viatica*). The higher contribution of high-chlorinated PCBs in substrates compared to springtails can be due to physicochemical properties, as high-chlorinated PCBs have higher affinity to particles and soil, thereby reducing the bioavailability for respiratory uptake to organisms,[70] as well as being resistant to degradation.[71] The PCB congener pattern in springtails resembles the one found in earthworms,[72] who have a low lipid content, where less lipophilic PCBs are easier taken up as they are more water-soluble. However, the resemblance of PCB patterns in springtails and Arctic air[73,74] may also indicate atmospheric delivery.

Comparing Contaminant Concentrations between Springtail Species. As expected, due to different habitats and physiology, contaminant levels differed between *M. arctica* and *H. viatica*. Ny-Ålesund (with low seabird influence) was the only site where both springtail species were sampled in sufficient amounts to conduct chemical analyses (*H. viatica*; n = 3 and *M. arctica*; n = 1). There, PFAS concentrations were more than twice as high in *H. viatica* as in *M. arctica*, whereas the opposite species difference was seen in the Hg concentrations. On a wet weight basis, *M. arctica* had higher concentrations than *H. viatica* for several important compounds such as PCB-138, PCB-153, and *p,p*-DDE (SI Table S3 of raw data). *M. arctica* had a lower lipid content of 3.4% compared to *H. viatica* with a mean lipid content of 9.4 ± 0.6% (n = 3) (Table 2), which results in higher lipid adjusted concentrations of contaminants in *M. arctica* compared to *H. viatica* with higher lipid content (Figure 3). When adjusted for lipid content, all chlorinated and brominated contaminants were higher in *M. arctica* compared to *H. viatica*, with the exception of HCB.

When considering springtail samples from all sites, Hg in *M. arctica*, found in areas of both low and high seabird influence, were up to 4 times higher than the highest Hg concentration in *H. viatica* (Table 2). Overall, *M. arctica* appeared to have a lower concentration of HCB compared to *H. viatica*, whereas no apparent trend was found for the remaining organic compounds when considering all study sites. These contamination differences between the two springtail species emphasize that species identity is important when evaluating contaminant exposure even within a defined taxon. Contamination concentrations and patterns may vary between vegetation and zones of the soil horizon, depending on the compounds and soil characteristics.[75,76] *M. arctica* appears to be tolerant of high concentrations of seabird-derived nutrients and thus the soil chemistry there, as it is commonly found closest to seabird colonies compared to other springtail species[11]; personal observation. In Ny-Ålesund, where both species were found, the δ^{15}N value in *M. arctica* (7.1‰) exceeded the δ^{15}N in *H. viatica* (3.6−3.8‰, n = 3), suggesting different microhabitat occupation by the species. Different habitats lead to exposure differences between the species, and could thus explain different contaminant levels.

Relationship between Contamination in Substrate and Springtails. There were no apparent trends between the total organic contaminant load in substrate and springtails (Figure 2). The few replicates per study site limited the opportunity for comparison between sites, and we therefore performed principal component analyses (PCAs) to detect trends within our contaminant data. A total of 21 and 24 compounds were detected in at least 65% of substrate and springtails samples, respectively, and applied in the multivariate statistics. Of these compounds, 17 were detected in both matrices. The PCAs on contaminant concentrations in substrate and springtails, with associated explanation and discussion is found in SI Figures S1, S2, and S7. Principal Component Analyses. The PCA on substrate contamination (SI Figure S1) is strongly influenced by the samples taken

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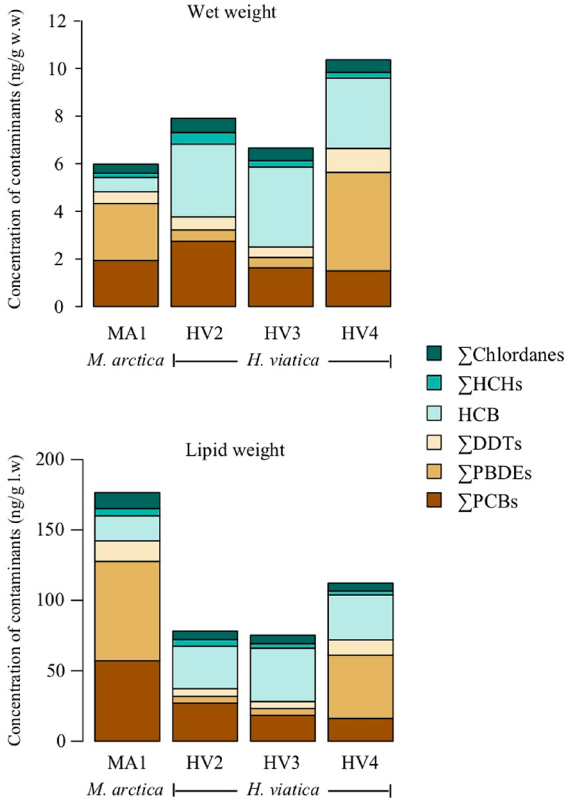

Figure 3. Lipid adjusted concentrations of sums ($\sum$) of chlorinated and brominated contaminants (ng/g l.w.) in *M. arctica* (n = 1) and *H. viatica* (n = 3) from Ny-Ålesund. Each replicate consisted of ≫10.000 pooled individuals. Abbreviations: PCBs = polychlorinated biphenyls, PBDEs = polybrominated diphenyl ethers, DDTs = dichlorodiphenyldichoroethylene and its metabolites, HCB = hexachlorobenzene, HCHs = hexachlorocyclohexanes, chlordanes = the chlordane compounds *trans*- and *cis*-chlordane, *trans*- and *cis*-nonachlor, and oxychlordane.

directly below the seabird colonies (Stuphallet and Krykkjefjellet), reflecting the high contaminant concentrations found on the tundra in these sites. The PCA on contaminant concentrations in springtails (SI Figure S2), however, did not detect any apparent trends between seabird influence ($\delta^{15}N$) and contamination in springtails, nor a clear relationship with contaminant concentrations in substrate.

A positive relationship between seabird influence and contaminant load was evident when comparing substrate samples at different distances from the seabird colony, although not across all study sites. Previous studies reflect clear trends between seabird influence and contamination in freshwater systems,[16,18] whereas inconsistent associations between seabird influence and recipient organisms are found in terrestrial areas,[34,38] including our study. This, along with the lack of correlations between contaminant concentrations in substrate and springtails, reflects the complexities of soil communities. Our study indicates that there is low uptake of contaminants in the soil fauna, despite the seabird-dervied contamination of their habitat. In general, there is limited knowledge on the levels and distribution of most contaminants in terrestrial soil, vegetation, and animals in the Arctic, which thus provides limited basis for comparison.[58,77,78] This calls for further research on the Arctic terrestrial environments to obtain a more comprehensive understanding of pollution dynamics, and we therefore consider this study to contribute valuable knowledge to the field of arctic ecotoxicology.

■ ASSOCIATED CONTENT

Supporting Information

This information is available free of charge via the Internet at The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b05316.

Method description of the determination of lipid-soluble contaminants, PFASs and Hg; method description of the quality assurance of the chemical analyses; method description of stable isotope analysis; arguments for excluding PFASs from certain comparisons; principal component analyses (PCAs); figures of PCAs; figure comparing Hg concentration in substrate to the relative amount of moss in sample material; relative proportions of contaminant groups including PFASs; table containing a description of samples; tables containing raw data of contaminant concentrations in springtails and substrate (PDF)

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Notes

The authors declare no competing financial interest.

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