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Wandering Albatrosses Document Latitudinal Variations in the Transfer of Persistent Organic Pollutants and Mercury to Southern Ocean Predators

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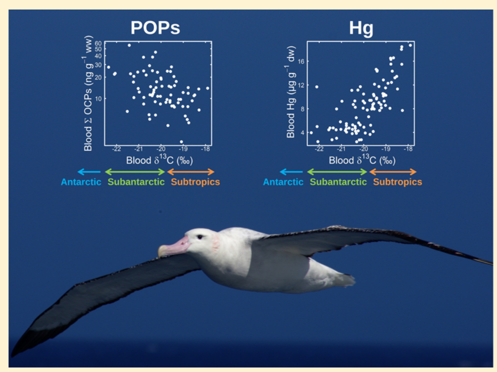
Supporting Information

ABSTRACT: Top marine predators are effective tools to monitor bioaccumulative contaminants in remote oceanic environments. Here, we used the wide-ranging wandering albatross *Diomedea exulans* to investigate potential geographical variations of contaminant transfer to predators in the Southern Ocean. Blood concentrations of 19 persistent organic pollutants and 14 trace elements were measured in a large number of individuals (N = 180) of known age, sex and breeding status from the subantarctic Crozet Islands. Wandering albatrosses were exposed to a wide range of contaminants, with notably high blood mercury concentrations. Contaminant burden was markedly influenced by latitudinal foraging habitats (inferred from blood $\delta^{13}C$ values), with individuals feeding in warmer subtropical waters having lower concentrations of pesticides, but higher concentrations of mercury, than those feeding in colder subantarctic waters. Sexual differences in contaminant burden seemed to be driven by gender specialization in feeding habitats, rather than physiological characteristics, with females foraging further north than males. Other individual traits, such as adult age and reproductive status, had little effect on blood contaminant concentrations. Our study provides further evidence of the critical role of global distillation on organic contaminant exposure to Southern Ocean avian predators. In addition, we document an unexpected high transfer of mercury to predators in subtropical waters, which merits further investigation.



INTRODUCTION

Anthropogenic activities have released intentionally and nonintentionally hundreds of thousands of different chemical compounds into the environment. Among these, persistent organic pollutants (POPs) such as organochlorine pesticides (OCPs), polychlorinated byphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) are of great concern, because they were designed to resist degradation for their agricultural and industrial applications.[1,2] Additionally, human activities have increased emissions to the environment of natural trace elements, such as metals and metalloids, raising their concentrations in a variety of ecosystems worldwide.[3] POPs and both essential and nonessential trace elements can be toxic for humans and wildlife.[4,5] Moreover, some environmental contaminants, such as POPs and mercury (Hg), bioaccumulate in biota and biomagnify up food webs.[6,7] POPs and Hg also undergo long-range atmospheric transport and deposition,[8,9] contributing to the nonpoint source contamination of remote environments. The World Ocean in particular appears to be the last sink for some POPs and trace elements,[8,10,11] but few data are available on their distribution and trophic transfer in remote oceanic areas.[12,13] This is particularly true for the southern Indian Ocean, where POPs[14−18] and trace elements[19−28] have been measured in only some biota, while information on water concentrations lacks dramatically.[12,29,30]

While direct assessment of environmental contaminants in large open water regions is logistically challenging, great insight

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into patterns of marine contamination can be obtained by using top predators as bioindicators.[31] Seabirds, in particular, have revealed important geographical trends of contaminant transfer to predators in a variety of ecosystems.[32,33] Yet, many different factors can drive variation in seabird exposure, hampering their use as reliable bioindicators of marine contamination.[34] Variability in seabird POP and trace element concentrations results not only from extrinsic factors, such as feeding habitat and trophic position,[27,35,36] but also from intrinsic factors, such as detoxification capability and nutritional condition.[2,37] Intraspecific variation in contaminant exposure has received substantial consideration (e.g., ref 38), yet marked between-individual differences often remain largely unexplained.[39,40] Overall, there is a need for more studies that concurrently assess a wide range of causal ecological factors. In particular, life-history traits have been rarely considered,[41,42] due to the paucity of long-term surveys on seabird populations giving access to individuals of known age and breeding history.[28,43,44]

The present study evaluates POP and trace element concentrations in a large number of known individual wandering albatrosses *Diomedea exulans*, breeding at the subantarctic Crozet Islands, southern Indian Ocean. Blood was used as monitoring tissue, since circulating contaminants are known to reflect internal tissues concentrations.[45,46] The wandering albatross is an extremely long-lived (>50 years) cephalopod-eating seabird with one of the highest trophic levels among marine consumers of the Southern Ocean.[24,47] This top predator is thus potentially exposed to large quantities of contaminants through bioaccumulation mechanisms, as shown for Hg.[24,48,49] During the breeding period (a whole year) wandering albatrosses are central-place foragers and undertake large scale movements, foraging up to 3500 km from their nest, thus ranging from subtropical to Antarctic waters.[50,51] This provides an exceptional opportunity to investigate contaminant trophic transfer over a large latitudinal range from one single species of apex predator.

The objectives of this investigation were 3-fold. First, contaminant concentrations of the wandering albatross were described and compared to those found in closely related species worldwide, in order to set results from this study in a global context. Second, by combining information on individual traits from a long-term capture-mark−recapture survey and by using the stable isotope ratios of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) as trophic tracers, we assessed the relative contribution of intrinsic (sex, age, and breeding status) and extrinsic (feeding ecology) factors on contaminant burden. The final aim of this study was to infer potential latitudinal trends in contaminant transfer to predators in the southern Indian Ocean. Feeding ecology was expected to be more relevant than intrinsic traits in explaining between-individual variation in contaminant concentrations.[27,48] Both POP and Hg burdens were predicted to be higher in individuals feeding in cold subantarctic waters than in those feeding in the subtropics, since polar environmental characteristics (e.g., low temperature, winter darkness) favor the atmospheric deposition and persistence of these contaminants.[1,8,9]

■ MATERIALS AND METHODS

Study Site and Sampling Procedure. The study was carried out on Possession Island, Crozet Archipelago (46°S, 52°E). The island lies in the Subantarctic Zone that corresponds to the water masses situated between the northern and warmer Subtropical Zone and the southern and colder Antarctic Zone.[52] Adult wandering albatrosses return to their breeding grounds in December and females lay a single egg in late December to early January. Both parents incubate alternatively until hatching in March and most young are fledged in November. During the incubation period (21 December to 4 March 2008), a total of 180 wandering albatrosses were sampled, including breeding and nonbreeding individuals. All birds were of known age (3−49 years), sex and breeding status, since they are part of a long-term capture-mark−recapture program started in 1966.[53] Blood was taken from the tarsal vein with a 1 mL heparinized syringe and a 25-gauge needle. Plasma and red blood cells were separated by centrifugation and stored at −20 °C. POPs and trace elements were measured in plasma and red blood cells, respectively, where they preferentially partition.[49,54,55] Hence, "blood" within the whole text refers either to plasma for POPs or red blood cells for trace elements.

POP and Trace Element Analyses. POPs were measured at the laboratory EPOC-LPTC, Bordeaux, France, from plasma (N = 128, 100 μL aliquots). Targeted compounds included seven indicator PCBs (CB-28, -52, -101, -118, -138, -153, and -180), 11 OCPs (hexachlorobenzene: HCB; lindane: γ-HCH; heptachlor; cis-chlordane; trans-nonachlor; mirex; 2,4′-DDE; 4,4′-DDE; 4,4′-DDD; 2,4′-DDT; 4,4′-DDT) and one PBDE (BDE-47). POPs were quantified using gas chromatography coupled with electron capture detection (GC-ECD). The percentage of total lipids in plasma was also measured on an aliquot of 10 μL by the sulfo-phospho-vanillin (SPV) method for colorimetric determination.[56] POP results are given in both absolute concentrations in ng g^{-1} wet weight (ww) and relative to the plasma lipid weight (lw).

Fourteen trace elements were measured on lyophilized red blood cells at the laboratory LIENSs, La Rochelle, France. Total Hg was quantified with an Altec AMA 254 spectrophotometer (N = 169, aliquots mass: 5−10 mg dry weight, dw), while arsenic (As), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), selenium (Se), and zinc (Zn) were analyzed using a Varian Vista-Pro ICP-OES and silver (Ag), cadmium (Cd), cobalt (Co), lead (Pb), and vanadium(V) using a Series II Thermo Fisher Scientific ICP-MS (N = 165, aliquots mass: 20−200 mg dw). Results are presented in absolute concentrations in μg g^{-1} dw.

Quality Control/Quality Assessment and other details about POP and trace element analyses are given in the Supporting Information (SI) (first paragraph and Table S1). All results are given as means ± SD. Since POP distributions were asymmetric, especially for PCBs, median rather than mean values were used for comparisons with the literature.

Stable Isotope Method. The isotopic niche of albatrosses was used as a proxy of their ecological niche.[57] The isotopic method was validated in the southern Indian Ocean, with $\delta^{13}C$ values of seabirds indicating their foraging habitats[50,58] and their $\delta^{15}N$ values increasing with trophic level.[59] The isotopic method is based on time-integrated assimilated food, with different tissues recording trophic information at different time scales. In this study, $\delta^{13}C$ and $\delta^{15}N$ values were measured in red blood cells, which provide trophic information on a few weeks before sampling,[60] thus corresponding here to the incubation period. The effect of blood $\delta^{13}C$ and $\delta^{15}N$ values on contaminant exposure was investigated using an isotopic data set that was built to study the effect of age, sex and breeding status on foraging strategies of the wandering albatross.[61]

Table 1. Model Selection for Blood Σ_7PCBs, Σ_{11}OCPs, Hg, and Cd Concentrations in Breeding Wandering Albatrosses from the Crozet Islands[a]

models	k^b	AIC_c	ΔAIC_c	w_i^c	exp. var. (%)^d
$\sum_7$ PCBs-GLM, Gamma Distribution, Inverse Link Function, N = 75 (M: 41, F: 34)					
lipid	2	452	0.00	0.29	7
lipid + age	3	452	0.39	0.24	7
lipid + δ^{13}C	3	453	0.68	0.20	7
lipid + sex	3	453	1.05	0.17	7
age	2	456	3.99	0.04	2
null	1	457	4.62	0.03	
δ^{13}C	2	458	6.09	0.01	0
sex	2	458	6.58	0.01	0
maximal: δ^{13}C + sex + age + lipid + δ^{13}C:sex + δ^{13}C:age	7	459	7.07	0.01	5
$\sum_{11}$ OCPs-GLM, Gamma Distribution, Inverse Link Function, N = 75 (M: 41, F: 34)					
δ^{13}C + sex + lipid	4	503	0.00	0.29	24
δ^{13}C + sex + lipid + δ^{13}C:sex	5	504	1.22	0.16	24
δ^{13}C + sex	3	505	1.46	0.14	21
δ^{13}C + sex + lipid + age	5	505	1.81	0.12	23
δ^{13}C + sex + age	4	506	2.61	0.08	21
δ^{13}C	2	506	3.03	0.06	20
δ^{13}C + lipid	3	506	3.09	0.06	22
Sex	2	507	3.82	0.04	19
δ^{13}C + age	3	507	3.83	0.04	21
maximal: δ^{13}C + sex + age + lipid + δ^{13}C:sex + δ^{13}C:age	7	509	5.74	0.02	22
null	1	521	17.99	0.00	
lipid	2	522	19.20	0.00	0
age	2	523	20.15	0.00	0
Hg-LM, N = 95 (M: 50, F: 45)					
δ^{13}C + sex + δ^{13}C:sex	4	1784	0.00	0.59	53
δ^{13}C + sex + age + δ^{13}C:sex	5	1786	2.27	0.19	53
maximal: δ^{13}C + sex + age + δ^{13}C:sex + δ^{13}C:age	6	1788	4.35	0.07	53
δ^{13}C	2	1789	4.70	0.06	50
Hg-LM, N = 95 (M: 50, F: 45)					
δ^{13}C + sex	3	1789	5.21	0.04	50
δ^{13}C + age	3	1790	6.42	0.02	50
δ^{13}C + sex + age	4	1791	7.03	0.02	50
δ^{13}C + age + δ^{13}C:age	4	1792	7.74	0.01	50
δ^{13}C + sex + age + δ^{13}C:age	5	1793	8.58	0.00	50
sex	2	1820	36.21	0.00	30
sex + age	3	1822	38.39	0.00	29
null	1	1853	69.25	0.00	
age	2	1855	71.22	0.00	0
Cd-GLM, Gamma Distribution, Inverse Link Function, N = 93 (M: 49, F: 44)					
δ^{13}C + sex	3	890	0.00	0.33	7
Sex	2	891	0.94	0.21	5
δ^{13}C + sex + age	4	892	1.99	0.12	6
δ^{13}C + sex + δ^{13}C:sex	4	892	2.17	0.11	6
sex + age	3	893	2.96	0.07	4
δ^{13}C + sex + age + δ^{13}C:age	5	893	3.84	0.05	4
δ^{13}C + sex + age + δ^{13}C:sex	5	894	4.11	0.04	4
null	1	895	4.99	0.03	
maximal: δ^{13}C + sex + age + δ^{13}C:sex + δ^{13}C:age	7	896	6.12	0.02	5
age	2	896	6.82	0.01	0
δ^{13}C	2	896	6.89	0.01	0
δ^{13}C + age	3	898	8.83	0.00	0
δ^{13}C + age + δ^{13}C:age	4	899	10.28	0.00	0

[a]Models are sorted by increasing ΔAIC_c (i.e., decreasing model fit). Abbreviations: AIC_c, Akaike's Information Criteria adjusted for small sample-sizes; w_i, Akaike's weights; Exp. var., explained variation. [b]Number of parameters. [c]Weight of evidence interpreted as a proportion. Weights across all models sum to 1.00. [d]Explained variation calculated from deviance or variance for GLM and LM, respectively, and adjusted depending on k and N.

Results are given in ‰ as means ± SD. Details about stable isotope analyses are given in the SI.

Statistical Analyses. All statistical analyses were performed using R 2.15.1.[62] Only POP and trace element concentrations that were above the limit of quantification (LoQ) in at least 70% of individuals were included in statistical analyses. For these POPs and trace elements, concentrations below the LoQ were substituted using 0.5·LoQ to avoid missing values distorting the statistical outcomes. Data exploration was carried out following Zuur et al.[63] with relationships between variables being tested with Pearson or Spearman correlation tests. In a first descriptive step, a principal component analysis (PCA) was carried out on log-transformed POPs and trace elements in order to highlight covariance. In a second explanatory step, univariate analyses (linear models, LM, or generalized linear models, GLM) were used to test the effect of individual traits and foraging ecology on absolute contaminant concentrations of breeding individuals. As sample sizes differed for POPs and trace elements, separate models were constructed. In order to reduce multiple testing, only the sum of PCBs ($\sum_7$PCBs) and OCPs ($\sum_{11}$OCPs) and the nonessential, potentially harmful Hg and Cd were retained as response variables. Pb and Ag were not considered as response variables, because the former had quantifiable concentrations in less than 70% of individuals, and the latter explained poorly the total variation in the PCA data set (see variable loadings on principal component axis in Table S2, SI). The $\sum_7$PCBs and sumΣ_{11}OCPs were correlated to individual PCBs and OCPs, respectively (Pearson correlation, $0.48 < r < 0.94$ for PCBs and $0.28 < r < 0.97$ for OCPs, all $p < 0.01$, N = 83), with the exception of 2,4′ DDT and heptachlor for OCPs (r = 0.12 and 0.05, p = 0.29 and 0.62, respectively). Only biologically meaningful models were constructed, with the maximal model being *Contaminant* ∼ δ^{13}C + sex + age + δ^{13}C:sex + δ^{13}C:age (with ":" indicating interactions). The percentage of lipids in plasma (hereafter lipid) was also included as a covariate in models explaining $\sum_{11}$OCPs and $\sum_7$PCBs values. Lipid content was not related to feeding habitat or age (data not shown). Blood δ^{15}N was not included in the models, since it was strongly correlated to δ^{13}C (Pearson correlation, r = 0.85, p < 0.0001, N = 104). This results from the slight, latitudinal enrichment in δ^{15}N values from cold to warm waters of the southern Indian Ocean (see also ref 50). Over the large latitudinal gradient exploited by wandering albatrosses, the trophic-level information on δ^{15}N values is thus confounded by a feeding habitat effect. Forward selection using the Akaike's Information Criterion corrected for small sample sizes (AICc)[64] was applied. Since our aim was to make inference on the variables affecting contaminant burdens, the effect of variables was inferred through Akaike's weights, and without using model averaging.[64] Finally, the effect of breeding status was separately tested on males only, since the sample of nonbreeding individuals was unbalanced (only three nonbreeding females were analyzed for contaminant concentrations). Breeding status categories thus included male immature (3−11 year-old birds with no known breeding attempts), breeding and nonbreeding individuals. GLMs were constructed in the form *Contaminant* ∼ Breeding status (+ lipid, for $\sum_{11}$OCPs and $\sum_7$PCBs) and hypothesis testing was applied (likelihood ratio test, LRT, between each model and the null model). For all analyses, model specification and validation were based on residuals analysis.[65]

■ RESULTS

POP and Trace Element Concentrations and Associations. Among the 33 targeted POPs and trace elements, 30 were detected in blood of wandering albatrosses from the Crozet Islands (see Table S3 and S4, SI). The POP pattern was dominated by OCPs (58% of $\sum_{19}$POPs), with the highest median concentrations being reported for 4,4′-DDE and HCB (5.4 and 1.8 ng g^{-1} ww, respectively). Other compounds with quantifiable concentrations in most individuals (>70%) were 4,4′-DDD, mirex and trans-nonachlor. Noticeably, the isomers 2,4′-DDT and 4,4′-DDT had quantifiable concentrations in more than 60% of individuals. The $\sum_{11}$OCPs ranged from 1.3 to 56 ng g^{-1} ww. Indicator PCBs accounted for 40% of the $\sum_{19}$POPs, with congeners CB-138, CB-153, and CB-180 having quantifiable concentrations in most individuals (>70%). The highest median concentration was however reported for CB-118 (2.3 ng g^{-1} ww). The Σ_7PCBs ranged from 0.1 to 676 ng g^{-1} ww. Only 18% of individuals had quantifiable concentrations of BDE-47, with values ranging from < LoQ to 1.9 ng g^{-1} ww (BDE-47 accounted for 2% of $\sum_{19}$POPs). Blood POP concentrations presented large between-individual variation, with coefficients of variation (CVs) being particularly high for PCBs (range 119−296%, Table S3, SI).

Among the 14 trace elements, only three were not detected in any individual (the essential Co, Mn, and V), while seven were quantifiable in more than 70% of individuals, including both essential (Cu, Fe, Se, Zn) and nonessential (Ag, Cd, Hg) elements. Fe and Se reported the highest concentrations among essential elements (2326 ± 345 and 77 ± 33 μg g^{-1} dw, respectively). Notably, Hg had quantifiable concentrations in all individuals and showed the highest concentrations among nonessential elements (7.7 ± 3.6 μg g^{-1} dw). Between-individual variation was less pronounced for trace elements than POPs, with the nonessential Ag and Pb having the highest CVs (189% and 95%, respectively, Table S4, SI).

PCA analyses included the eight POPs and seven trace elements that had quantifiable concentrations in more than 70% of individuals (see Table S2, S3, and S4, SI). POPs and Hg contributed markedly to the total variation in the data set. Strong associations were identified within PCBs and within OCPs, but the two POP classes were not associated with each other as shown by the PCA circle of correlations (Figure S1, SI). Hg was potentially negatively associated with OCPs, but no association with other metals was clearly identified.

Explanatory Factors of Between-Individual Variation in Exposure. Univariate analyses were applied to disentangle the influence of sex, age, and feeding habitat (δ^{13}C) on $\sum_7$PCBs, $\sum_{11}$OCPs, Hg and Cd burdens in breeding wandering albatrosses (Table 1). For the $\sum_7$PCBs, multiple models had a similar support (ΔAICs < 2), but explained only 7% of the total variation (Table 1). Plasma lipid content was clearly the most influential variable, as shown by the sum of Akaike's weights across all models (Table 2). $\sum_7$PCBs

Table 2. Sum of Akaike's Weights Across All Models of Each Tested Explanatory Variable for Blood Σ_7PCBs, Σ_{11}OCPs, Hg, and Cd Concentrations in Breeding Wandering Albatrosses From the Crozet Islands

	sum of Akaike's weights across all models			
explanatory variables	$\sum_7$ PCBs	$\sum_{11}$ OCPs	Hg	Cd
age	0.29	0.26	0.31	0.31
lipid	0.91	0.65		
sex	0.19	0.85	0.91	0.95
δ^{13}C	0.22	0.97	1	0.68

concentrations increased with increasing lipid content (Figure S2, SI). Multiple models had a similar support for blood $\sum_{11}$OCPs concentrations, explaining approximately 20% of the total variation (Table 1). δ^{13}C, sex, and lipid were the most important predictor variables (Table 2). Exposure was negatively related to δ^{13}C values (Figure 1a). Males and females had Σ_{11}OCPs concentrations of 18.4 ± 10.7 and 11.1 ± 6.2 ng g^{-1} ww, respectively.

One single model best described blood Hg data, the LM δ^{13}C + sex + δ^{13}C:sex, with a percentage of explained variation of 53% (Table 1). The sum of Akaike's weights across all models confirmed the strong effect of δ^{13}C and sex on Hg concentrations (Table 2). Unlike the Σ_{11}OCPs, Hg concen-

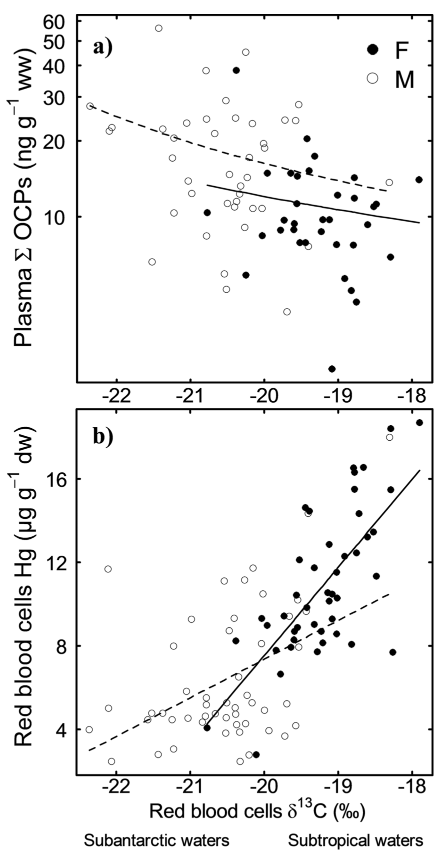

Figure 1. (a) OCP concentrations decrease whereas (b) Hg concentrations increase with decreasing latitude of foraging habitats in blood of breeding wandering albatrosses from the Crozet Islands.

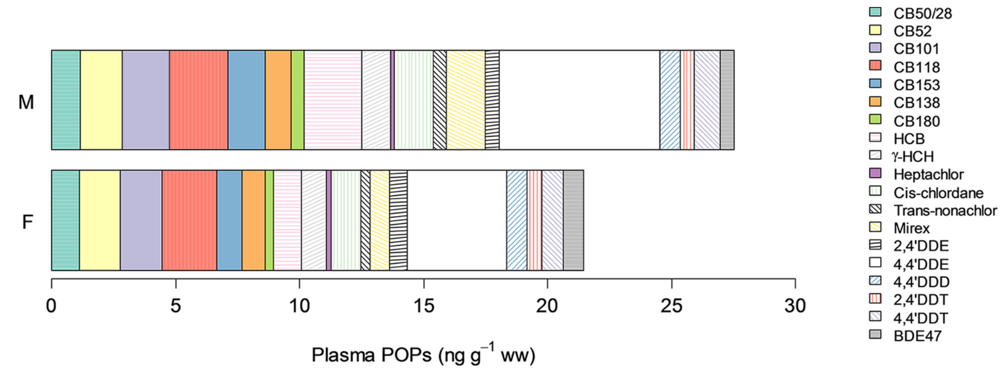

Figure 2. Stacked bar plot of POPs in plasma of male and female wandering albatrosses from the Crozet Islands. Values correspond to median concentrations.

trations were positively related to δ^{13}C values (Figure 1b) and were lower in males than in females (6.4 ± 3.3 and 10.9 ± 3.5 μg g^{-1} dw, respectively). Finally, for blood Cd concentrations, multiple models had a similar support, but explained only 5–7% of the total variation (Table 1). Sex was the factor with the highest sum of Akaike's weights across all models (Table 2). Males had lower Cd concentrations than females (56 ± 28 and 72 ± 38 μg g^{-1} dw, respectively).

Notably, age had no significant effect on contaminant exposure for any of the tested contaminants ($\sum_7$PCBs, $\sum_{11}$OCPs, Hg, and Cd), with the sum of Akaike's weights accounting for age ranging only between 0.26 and 0.31 (Table 2).

Breeding status had a significant effect only on the $\sum_7$PCBs in males (GLM on log-transformed data, Gaussian distribution, identity link function, taking into account the lipid effect, p = 0.01, N = 79), with concentrations in nonbreeding individuals being higher than in breeding ones. Conversely, $\sum_{11}$OCPs, Hg and Cd concentrations were not influenced by breeding status (GLM, gamma distribution, inverse link function, p = 0.38, 0.14, and 0.55, N = 79, 106, and 100, respectively). There was however a tendency for immature birds to show higher blood Hg concentrations than nonbreeding ones.

DISCUSSION

The present work is one of the most comprehensive evaluations of POP and trace element burdens in free-living seabirds of known life-history traits, because of the large number of both sampled individuals (N = 180) and targeted contaminants (N = 33). Our results document that the wandering albatross was exposed to a wide range of organic and inorganic contaminants during the breeding period, highlighting the extent of global contamination in the remote Southern Ocean. Recent investigations on physiological and demographic consequences of selected contaminants on the same individuals have revealed that blood POP and Hg concentrations are related to increased oxidative damage[43] and to decreased breeding probability and output.[44]

Pattern of Contamination and Comparison to Other Species and Areas. Previous studies in subantarctic seabirds evaluated legacy-POP concentrations in internal tissues of dead individuals,[14−16,66] and emerging-POPs (perfluorinated compounds) in blood.[17] Therefore, no previous data is available for comparing wandering albatross blood concentrations of legacy-POPs to those of neither subantarctic, nor subtropical seabirds from the Southern Hemisphere. Plasma POP concentrations of the wandering albatross were similar to or lower than those of high-Antarctic seabirds. For example, the south polar skua *Catharacta maccormicki* had three- and 15-fold higher plasma HCB and mirex concentrations, respectively, than the wandering albatross, while median PCB concentrations were comparable.[67] The snow petrel *Pterodroma nivea* showed similar OCP, but higher PCB concentrations than wandering albatrosses.[68] When compared to seabirds from the Northern Hemisphere, wandering albatrosses had overall lower plasma POP concentrations. In particular, two- to 200-fold higher median PCB concentrations have been reported in the plasma of Arctic seabirds (e.g., refs 67 and 69) and North Pacific albatrosses.[13,32,70] Conversely, differences in plasma OCP concentrations between the wandering albatross and Northern Hemisphere seabirds are less pronounced, especially for HCB and mirex.[38,69] Overall, the pattern of organic contamination in plasma of the wandering albatross is remarkable for three main reasons: (1) the smaller abundance of PCBs over OCPs (Figure 2) with respect to Northern Hemisphere species, which is probably related to the distance to industrial sources;[67] (2) the lower concentrations of PBDEs than both PCBs and OCPs, as reported in other albatrosses;[13] and (3) the abundance of HCB, mirex and DDT derivatives, which testifies to their use and emissions in the Southern Hemisphere,[71,72] including recent DDT application for disease vector control.[73]

With regards to nonessential trace elements, blood Hg concentrations were remarkably high in the wandering albatross, as previously shown in feathers (e.g., ref 27, 48, and 49) and internal tissues (e.g., ref 74). Similar blood Hg concentrations have recently been reported in the brown skua *Stercorarius lonnbergi* from the subantarctic Kerguelen Islands.[28] Wandering albatrosses had higher blood Hg concentrations than Antarctic seabirds, such as the south polar skua[28] and the snow petrel.[42] Notably, blood Hg concentrations of wandering albatrosses were comparable to those of the great skua *Catharacta skua*,[39] one of the species with the highest blood Hg concentration in the Northern Hemisphere. On the other hand, blood Cd concentrations were lower than expected, given the importance of squid in the diet of the wandering albatross.[75] Squid has been recognized to be an important vector for Cd transfer to top predators.[19] However, similar low Cd concentrations were found in blood of wandering albatrosses at South Georgia, southern Atlantic Ocean.[35] This result suggests that after assimilation Cd is efficiently transported toward target tissues where it is stored, as proved by high concentrations in liver and kidneys.[74,76] Low blood concentrations were reported also for Pb, which is consistent with results of Anderson et al.[35] and likely the consequence of low dietary exposure. Indeed, high blood Pb concentrations have been reported in procellariiform species feeding in neritic waters off Patagonia and Brazil, respectively, thus reflecting the contamination of those coastal waters.[77,78] With regards to essential elements, blood concentrations had relatively low between-individual variation (Table S4, SI) and were overall within the same range of values as other subantarctic procellariiform seabirds,[35,77] suggesting that no apparent deficiencies were present in this population.

Effect of Lipids and Breeding Status. Since POPs are strongly lipophilic, their concentrations in living organisms are influenced by lipid dynamics.[2] Here, plasma POP concentrations were influenced by lipid content, in particular for PCBs. This is consistent with previous works showing that lipid status is more determinant for some compounds, such as low-chlorinated PCBs.[41,45,46] Breeding status implies particular physiological conditions that could also affect blood contaminant concentrations.[40,79] Again, only plasma PCBs were significantly affected by breeding status, indicating that physiological traits may be more important than extrinsic factors in driving variation for this class of POPs. On the other hand, immature birds had slightly higher blood Hg concentrations than nonbreeding individuals. It has been hypothesized that immatures moult less frequently than adults,[80] which would mean that they have fewer opportunities to excrete Hg into feathers,[49] but there is no conclusive evidence to support this explanation. Other between-individual and between-compounds differences in detoxification capabilities can also be responsible for the high variability in the data, especially for POPs. The whole body biological half-life of some POPs in birds can be long (100−400 days for the herring gull *Larus argentatus*).[81] There is thus a partial temporal uncoupling between POPs and stable isotope ratios in blood,[6] which could imply carry-over effects of past exposure over wintering grounds.[36] Clearly, a better knowledge on the toxicokinetics of contaminants in blood is needed for the complete understanding of between-individual variation in seabird exposure and contaminant burdens.

Effect of Age. Blood concentrations of POPs, Hg and Cd were not age-related in breeding wandering albatrosses, despite the large age range (7−47 years) and sample size. The absence of age-dependent variation in contaminant concentrations has already been reported in blood[41,49,70,77] and feathers[82,83] of other known-age seabirds, with also some possible decreasing trends being observed.[42] While some studies show contrasting results for age-related trends of trace elements in seabird internal organs, especially Cd,[84,85] overall there is no evidence of a clear, significant increase in POP and Hg concentrations with adult age in any tissue. This pattern could be explained by efficient detoxification mechanisms in seabirds. Indeed, feather excretion is a well-known mechanism for Hg elimination (e.g., ref 37), which is significant also for organic contaminants (e.g., ref 86). Moreover, POPs can be excreted in preen oil[87] and undergo biotransformation in internal tissues.[2] This contrasts with results in marine mammals, which have shown clear age-dependent increases in adult internal tissues contamination, including blood.[88−90] Moreover, the absence of confounding age-related variation in contaminant concentrations enhances the value of adult seabirds' blood as a reliable biomonitoring tool of environmental contamination.[79,82] It must be noted, however, that the present work is a cross-sectional study, which does not necessarily reveal information on the contamination of individuals as they age.[91] However, similar contaminant concentrations have been reported in seabirds sampled repeatedly in different years.[41,70] Furthermore, POPs, Hg, and Cd did not affect mortality in the wandering albatross,[44] which excludes potential bias from differential survival of the most contaminated individuals.

Effect of Feeding Habitat and Sex during the Breeding Period. Key findings of the present study are the correlations between blood $\delta^{13}C$ values and blood OCP and Hg concentrations (Figure 1). The Southern Ocean is marked by a well-defined latitudinal baseline $\delta^{13}C$ gradient that is reflected in the tissue of consumers.[50,58,92] The wide range of blood $\delta^{13}C$ values thus indicates that wandering albatrosses foraged over a vast latitudinal area during the incubation period. Based on blood $\delta^{13}C$ isoscapes, wandering albatrosses exploited large domains of the Subantarctic and Subtropical Zones.[50] The inverse relationship between blood $\delta^{13}C$ values and OCP concentrations (Figure 1a) therefore strongly suggests a latitudinal pattern in OCP transfer to predators, which *increased* from warm subtropical (high $\delta^{13}C$ values) to cold subantarctic waters (low $\delta^{13}C$ values). Conversely, the direct correlation between blood values and Hg concentrations (Figure 1b) likely indicates the opposite pattern in Hg transfer to predators, which *decreased* from warm subtropical (high $\delta^{13}C$ values) to cold subantarctic (low $\delta^{13}C$ values) waters. Importantly, sexual differences in contaminant exposure were explained by gender specialization in foraging habitats.[61,93] Indeed, male (vs female) wandering albatrosses had higher exposure to POPs (vs Hg) likely because they used more subantarctic (vs subtropical) waters (Figure 1). Gender differences in uptake, metabolism, storage and excretion of organic and inorganic contaminants cannot be excluded, particularly taking into account that a fraction of both POPs and Hg is eliminated through the egg for females.[69,85] However, since the wandering albatross is a biennial species laying only one egg at each breeding attempt,[94] egg transfer alone could hardly explain sexual differences in contaminant concentrations (e.g., refs 2 and 95). Indeed, previous studies in low-fecundity seabird species have shown that sexual differences in contaminant concentrations are not always significant and are in different directions depending on species.[2,77,95] The observed higher blood concentration of Cd in females than in males was partially related to feeding habitats (Table 2) and is more difficult to explain. Nevertheless, different proportions of particular prey species (e.g., squids) between males and females could account for this pattern.

Latitudinal Variation in POP and Hg Transfer to Predators. The observed latitudinal pattern in POP transfer to the albatrosses, increasing from warm subtropical to cold subantarctic waters, is consistent with the cold condensation and fractionation theory. The latter predicts that increasing quantities of POPs are deposited in polar environments by repeated air-surface exchange and atmospheric transport, with the mixture shifting to more volatile compounds.[9] Our results thus suggest that OCPs enter readily trophic webs after atmospheric deposition, likely through adsorption on organic matter particles and uptake by phytoplankton.[10] Global distillation seems thus to significantly affect predators' exposure to OCPs in the Southern Ocean. This was not verified for PCBs, most likely due to the extremely high between-individual variation in blood (see second section of the discussion). Increases in both OCP and PCB exposure at high latitudes have already been observed in different species and populations of seabirds from Antarctica[71] and the Norwegian Arctic,[38,96] respectively, but we are not aware of previous evidence of such a latitudinal variation within a single seabird population.

Unlike POPs and contrary to our prediction, Hg transfer to the albatrosses decreased from warm subtropical to cold subantarctic waters. Previously, high concentrations of atmospheric Hg have been observed close to the Antarctic continent when compared to lower latitudes.[97] Moreover, the only biogeochemical investigation on Hg speciation and distribution in Southern Ocean waters has shown high concentrations of Me-Hg in Antarctic rather than subantarctic and subtropical waters.[30] Methyl-Hg is the bioavailable and most toxic form of Hg that is readily assimilated by low-trophic level organisms and then biomagnifies up the food web.[7] The heavy Hg burden of birds feeding in warm subtropical waters is therefore puzzling. However, a significant higher exposure to Hg in chicks fed by parents foraging in subtropical rather than subantarctic waters was previously underlined in different oceanic seabird species from the Kerguelen Islands.[24] A similar trend appears also from between-species comparisons. Indeed, Antarctic top predators present lower Hg concentrations than subantarctic ones (see first section of the discussion). Antarctic food webs are simpler that those found at subtropical latitudes.[50,98] Since food web structure influences Hg transfer,[99,100] the complexity of subtropical food webs could explain the higher Hg exposure in northern than southern foraging predators of the Southern Ocean. In addition, Hg dynamics within food webs is affected by many other factors, such as primary productivity, temperature and solar radiation.[8] Clearly, our results call for in-depth investigations of Hg speciation and food web dynamics in waters of the Southern Ocean.

Despite the presence of several confounding factors, our data documents a clear latitudinal trend in both POP and Hg transfer to predators in the southern Indian Ocean. Since breeding wandering albatrosses feed *all along* their foraging trips, their blood isotopic signature integrates prey taken in different water masses, thus diluting δ^{13}C values.[61] Such a "'dilution effect"' reduces differences among individuals, further emphasizing the strength of the habitat-related contaminant exposure depicted here.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Full details on the analytical techniques and Quality Control/Quality Assessment used for persistent organic pollutant (POP), trace element and stable isotope analyses. Results on the effect of heparin on blood mercury (Hg) concentrations. Tables detailing the limits of detection (LoD) and quantification (LoQ), descriptive statistics and principal component (PC) loadings of all the contaminants measured in blood of wandering albatrosses. Figures of the circle of correlations of the PC analyses, and of the relationship between blood Σ_7PCBs concentrations and plasma lipid content in breeding individuals. This material is available free of charge via the Internet at http://pubs.acs.org/.

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Notes

The authors declare no competing financial interest.

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