



# Concentrations of organic pollutants in seabirds from the tropical southwestern Atlantic Ocean are explained by differences in foraging ecology<sup>☆</sup>

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## ABSTRACT

Persistent organic pollutants are a potential threat for marine vertebrates in both coastal and offshore areas. In this study, organic pollutants were evaluated in the blood and feathers of four seabird species that forage in the tropical southwestern Atlantic Ocean. Red-billed tropicbirds (*Phaethon aethereus*) and brown boobies (*Sula leucogaster*) were sampled in the Abrolhos Archipelago, 70 km from the coast, and used as proxies of nearshore contamination. The Trindade petrel (*Pterodroma arminjoniana*) was sampled on Trindade Island, 1200 km offshore, and the Atlantic yellow-nosed albatross (*Thalassarche chlororhynchos*) was sampled at sea, both used as proxies of pelagic contamination. Concentrations of organohalogen pesticides ( $\sum$ OHP) and polychlorinated biphenyls ( $\sum$ PCB) were generally higher in the booby, the most nearshore forager, followed by the tropicbird, petrel and the albatross. Carbon isotope values ( $\delta^{13}\text{C}$ ) were positively associated with  $\sum$ OHP and  $\sum$ PCB in the blood of seabirds and explained 28.6% of the variation in pollutant data, suggesting higher concentrations of pollutants in the nearshore marine habitats, where  $\delta^{13}\text{C}$  is generally higher. Nitrogen isotope values ( $\delta^{15}\text{N}$ ) also had a positive influence over pollutant concentrations and explained 13% of pollutant data, suggesting an influence of trophic level. Variations in polycyclic aromatic hydrocarbon ( $\sum$ PAH) concentrations among species, and relationships with isotopic values were less clear. Furthermore, the concentrations of organic pollutants were substantially higher in 2019 than 2022, which suggests greater environmental pollution in 2019 that could be related to urban and agricultural sources. Results demonstrate relationships between seabird ecology and organic pollutants in the tropical marine environment and highlight the importance of assessing multiple species in monitoring pollutant concentrations in wildlife.

## 1. Introduction

The marine environment is the endpoint of many pollutants released by human activities (Elliott and Elliott, 2013; Weiss et al., 2021; Cong et al., 2022). The rate at which new compounds are implemented by industry and released in the environment is far beyond the understanding of their potential harmful effects on organisms, which represents a challenge to environmental conservation. Particularly relevant in terms of environmental impacts are the persistent organic pollutants,

synthetic halogenated compounds of historical concern due to their high resistance to degradation, capacity to bioaccumulate in organisms and magnify along the food chain (Fry, 1995; Fisk et al., 2001; Finkelstein et al., 2007). Due to these chemical properties and toxicity, persistent organic pollutants started being regulated worldwide after the Stockholm convention in 2004 (UNEP, 2017). Nonetheless, they are still present in the environment, and the long-range transport through ocean currents and the atmosphere makes any region susceptible to their presence (Lohmann et al., 2007; Adrogué et al., 2019; Campioni et al.,

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2024).

Sampling of animal tissues is an effective strategy for obtaining data on organic pollutants in marine food chains (Adrogué et al., 2019). Animals at high trophic levels, such as seabirds, reflect the bottom-up trophic transfer and magnification of contaminants in food webs. Seabirds are particularly useful to assess contamination in the ocean, as they aggregate in large numbers in breeding sites on land, and provide biological tissues that can be sampled non-destructively, their blood and feathers, allowing for long-term monitoring (Furness and Camphuysen, 1997; Adrogué et al., 2019; Campioni et al., 2024). Different time frames can be assessed, as blood has a short turnover (weeks) and feathers generally reflect the contamination over the period of moult (months) (Burger and Gochfeld, 2004), and also external deposition after the plumage is formed (Jaspers et al., 2008). Moreover, seabirds are highly mobile and integrate contaminants from their wide foraging areas through time (Burger and Gochfeld, 2004). Even sympatric seabird species may differ widely in their diets, at-sea distributions, and year-round movement patterns (Petalas et al., 2021; Linhares et al., 2024). These features allow the use of seabirds to track local, regional and biogeographical contamination patterns in the ocean (Jouanneau et al., 2022; Pollet et al., 2023), as well as assessing the influence of ecological traits on contaminant concentrations.

Organohalogen pesticides (OHPs) and polychlorinated biphenils (PCBs) are major persistent organic pollutants. Effects of legacy OHPs such as dichlorodiphenyltrichloroethane (DDT), hexachlorocyclohexane (HCH), endosulfan and aldrin were early demonstrated in wildlife after they started being indiscriminately applied after the World War II for the control of agricultural pests and human disease vectors (Carson, 1962). PCBs, on the other hand, were implemented in the industry as dielectric fluids and flame retardants in a range of materials (Diamond, 2017). These persistent organic pollutants can cause sublethal effects in birds such as eggshell thinning, embryo malformations and impairment of reproductive behaviour (Bryan et al., 1989; Holm et al., 2006; Blus, 2011; Win-Shwe et al., 2024).

Another important class of organic pollutants are the polycyclic aromatic hydrocarbons (PAHs). While PCBs and OHPs are synthetic, PAHs can occur naturally from petrogenic and pyrogenic sources, increasing in association to human activities such as the oil and gas industry, and the combustion of fuels and organic matter (Walker, 2008). Increased PAH concentrations in the marine environment were evidenced in birds after major oil spills or near highly industrialized areas (Troisi et al., 2007; Pérez et al., 2008; Seegar et al., 2015). However, PAHs may be readily metabolized by birds and relationships with trophic ecology are less clear than for PCB/OHPs, highlighting the need for investigation to understand PAH dynamics in wildlife (Seegar et al., 2015; Jodice et al., 2023).

Tropical seabirds have received less attention regarding organic pollution in comparison to those of high-latitude and polar regions, where long-term data is available in some areas (e.g. Mello et al., 2016; Braune et al., 2019; Carravieri et al., 2020; Provencher et al., 2022). Consequently, the hard-to-detect impacts of these compounds in tropical, developing countries, may have been overlooked, although these areas were the last to regulate most persistent organic pollutants (Gevao et al., 2010). Moreover, these regions may represent important sources of OHP, PCBs and PAHs worldwide due to intense agriculture and poor environmental regulations. For instance, less persistent OHPs are still allowed in South America, such as trifluralin, dichlofluanid, and chlorothalonil (current use pesticides – CUPs). In addition, acute contamination events, such as the Fundão Dam rupture that occurred in Brazil in 2015, have caused important changes in the contamination of organisms, including seabirds (Costa et al., 2022; Nunes et al., 2022; Bauer et al., 2024; Linhares et al., 2024). Some studies suggest increases in OHPs in the marine environment after the dam failure, which is probably related to the drag of contaminants deposited along the Doce River (Oliveira-Ferreira et al., 2022; Cabral et al., 2023; Yamamoto et al., 2023). Such a catastrophic event highlights the need of research

targeting organic pollution in tropical systems, such as along the Brazilian coast, in order to subsidize actions to secure wildlife in developing countries.

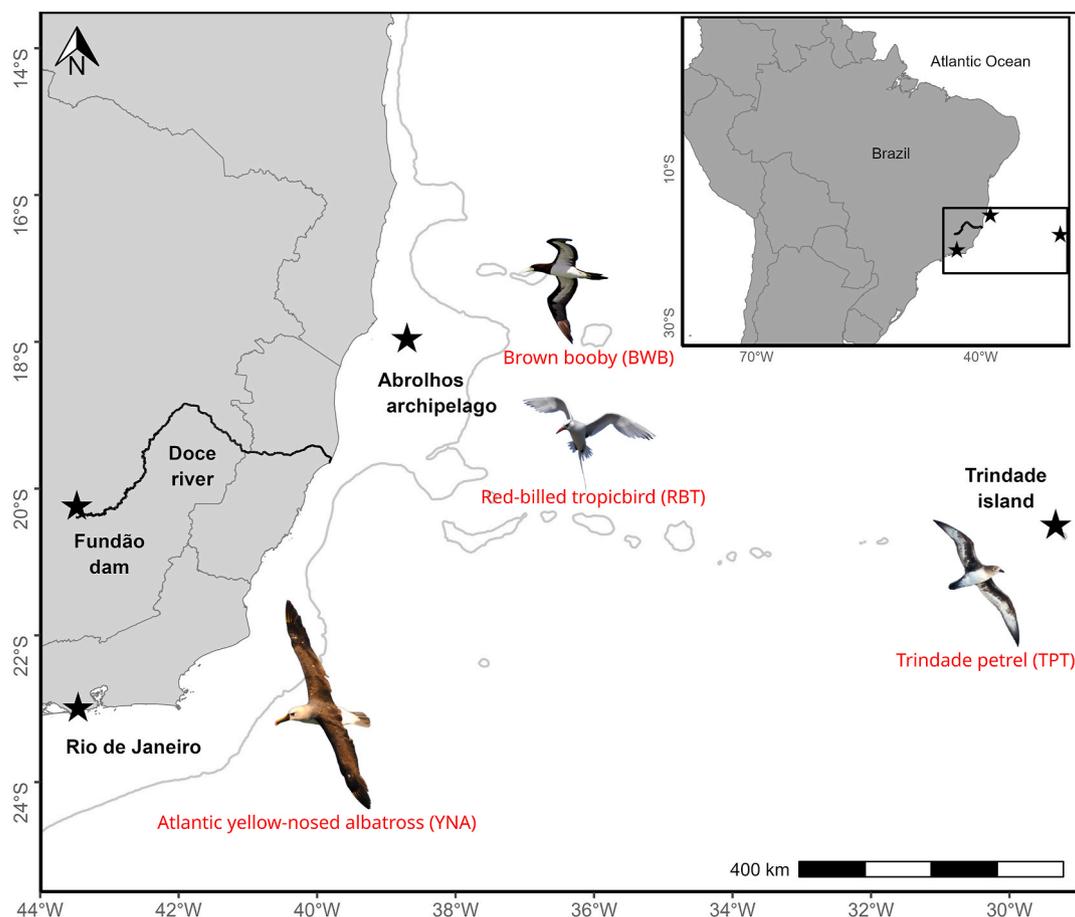
Investigation of organic pollutants in tropical seabirds with differing foraging ecologies could be useful to understand the factors that affect organic pollution. In the southwestern Atlantic Ocean, brown boobies (hereafter booby, *Sula leucogaster*, Suliformes) and red-billed tropicbirds (hereafter tropicbird, *Phaethon aethereus*, Phaethontiformes) breed at 70 km from the coast in the Abrolhos Archipelago. Both species overlap in their foraging range around the archipelago, but the booby mainly forages on shallower coastal areas (50 km from the coast, 80 m of depth; see Linhares et al., 2024), while the tropicbird has a larger range, using also deeper offshore waters (142 km from the coast, 1700 m of depth; see Linhares et al., 2024). Inversely, the Trindade petrel (hereafter petrel, *Pterodroma arminjoniana*, Procellariiformes) breeds 1200 km from the Brazilian coast in the tropical Trindade Island and uses a wide oceanic region for foraging during the breeding season (Leal et al., 2017). Likewise, the Atlantic yellow-nosed albatross (hereafter albatross, *Thalassarche chlororhynchos*, Procellariiformes) breeds in the subtropical Tristan da Cunha archipelago, more than 3500 km from the coast, and uses a very large foraging area in Brazil for foraging during the breeding and non-breeding season (Gabani, 2020). All these seabirds have been used to monitor trace elements in the marine environment after the Fundão Dam failure (Nunes et al., 2022; Bauer et al., 2024; Linhares et al., 2024). The tropicbird is threatened in Brazil (MMA, 2022), the booby has declined severely in Abrolhos over the past years (PMBA, 2023), and the petrel and albatross are globally classified as 'Vulnerable' and 'Endangered' by IUCN, respectively, what makes it important to quantify contaminant exposure, accumulation and potential health effects in these species.

Here, OHP, PCB and PAH concentrations were assessed in the blood and feathers of four seabird species with distinct foraging ranges in the southwestern Atlantic Ocean (Fig. 1). Stable isotopes of carbon and nitrogen in blood were used as tracers of foraging habitat ( $\delta^{13}\text{C}$ ) and trophic level ( $\delta^{15}\text{N}$ ), respectively (Bustamante et al., 2023; Elliott et al., 2023). Our main objective was to assess if pollutant concentrations were associated with the nearshore (the booby and tropicbird in Abrolhos) or pelagic (the petrel and albatross) marine habitats. We expected higher pollutant concentrations in the coastal species (booby and tropicbird) due to higher exposure from continental pollutant sources, with the highest concentrations in the booby due to more nearshore foraging habits and higher trophic position than the tropicbird (Linhares et al., 2024). Nonetheless, we expected higher concentrations of pollutants in the albatross than the petrel due to the consumption of high trophic level fish from fishery discards (Bugoni et al., 2010). Furthermore, we expected a relationship between organic pollutant concentrations and  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values, reflecting the difference in the foraging habitat and trophic position of seabirds, respectively.

## 2. Material and methods

### 2.1. Seabird sampling

Seabird blood and feathers were sampled at different sites: in the Abrolhos Archipelago, where the booby and tropicbird breed, at approximately 70 km from the Brazilian coast; in Trindade Island, the main global breeding site of the petrel, at approximately 1200 km from the coast; and during on-board cruises off the Rio de Janeiro state, in southeast Brazil, where albatross uses as a foraging site, mainly during the non-breeding season (Fig. 1). At breeding sites, birds were captured around their nests, by hand or using hand nets, in February 2019 and March 2022 in Abrolhos, and from March to May of 2019 and 2022 in Trindade Island. At sea, the albatrosses were attracted to research vessels using bait and captured with cast nets (Bugoni et al., 2008) in September 2019. Three breast contour feathers were plucked by hand and ~1 mL of blood was obtained from the tarsal vein; then, the seabird



**Fig. 1.** Study area in the southwestern Atlantic Ocean, where seabirds were sampled. Brown booby (*Sula leucogaster*) and red-billed tropicbird (*Phaethon aethereus*) were sampled while breeding in the Abrolhos Archipelago, and the Trindade petrel (*Pterodroma arminjoniana*) on Trindade Island. The Atlantic yellow-nosed albatross (*Thalassarche chlororhynchos*) was sampled at sea off Rio de Janeiro, a foraging area for non-breeding and breeding individuals. Stars represent an indication of sampling sites, as well as the site where the Fundão Dam ruptured in 2015, and tailings further reached the ocean through the Doce River. The 1000 m isobath is shown as a grey line. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

was released near the site of capture. Blood was frozen ( $-20\text{ }^{\circ}\text{C}$ ) in sterilized glass vials and feathers were kept in glass vials at room temperature and protected from light until analysis, which was conducted within three months of collection. All birds sampled were adults. Sampling was approved by SISBIO 64381 and 64234 licenses, and by the ethics committees at the *Universidade Federal do Rio Grande* and *Universidade Federal do Rio Grande do Sul*, Brazil.

A total of 32 blood samples (booby = 10, tropicbird = 10, petrel = 10 and albatross = 2) and 39 feather samples (booby = 10, tropicbird = 10, petrel = 10 and albatross = 9) from 2019 were used. As for 2022, 62 blood samples (booby = 20, tropicbird = 17 and petrel = 25) and 69 feather samples (booby = 20, tropicbird = 19 and petrel = 30) were analysed.

## 2.2. Organic pollutant analysis

Whole blood samples were subjected to solid-phase extraction using Chromabond® C18ec cartridges (Macherey-Nagel, Germany). Blood sample preparation followed Martins et al. (2020). Before the application of the blood samples, the cartridges were cleaned and conditioned with methanol, followed by ultrapure water, and then dried. The adsorbed analytes were eluted with methylene chloride, and then eluted with gentle vacuum. Extracts were then evaporated under a gentle nitrogen stream, and analytes were solubilized in hexane (Camacho et al., 2014). Feathers were washed with deionized water and dried. Then, 0.05 g feather aliquots were homogenized with anhydrous sodium

sulfate and spiked with p-Terphenyl-d14 and PCB 103, used as surrogate standards. Extraction was performed using a Soxhlet apparatus with a 1:1 n-hexane and dichloromethane solution, further subjected to evaporation (see Martins et al., 2023). Extracts of blood and feathers were then separated in two aliquots, used separately for PAH and OHP/PCB determination.

Fractionation of PAHs was carried through liquid chromatography (Martins et al., 2023). Serial elution was carried with n-hexane, n-hexane/dichloromethane (9:1) and n-hexane/dichloromethane (1:1). Quantification of PAH compounds was conducted with gas chromatography (GC-2010 Plus, Shimadzu, Kyoto, Japan) coupled to mass spectrometry (Shimadzu GCMS-QP, 2020; Shimadzu Corporation, Kyoto, Japan). Eighteen PAHs were determined (1-methylnaphthalene, 2-methylnaphthalene, naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene, and benzo[g,h,i]perylene).

The fractionation of PCBs and OHPs was carried out using a Florisil® liquid chromatography column. Serial elutions of n-hexane and an 8:2 mixture of n-hexane and dichloromethane corresponded to PCB and OHP fractions, which were evaporated in a rotary evaporator and then transferred to 2 mL vials for determination. OHPs and PCBs were determined using gas chromatography (GC-2010 Plus, Shimadzu, Kyoto, Japan) with Electron Capture Detector (GC-ECD). All samples were previously spiked with recovery standards (Sigma-Aldrich, St. Louis,

MO, USA) before the chromatographic analysis. Tetrachloro-m-xylene (TCMX) was used as the internal standard, while PCB-30, PCB-103, and PCB-198 were employed as recovery standards (Supelco®). The recovery rate of spiked standards ranged from 81.13% to 94.68% ( $87.72 \pm 4.3\%$ ). Twenty four OHPs were determined:  $\alpha$ -HCH,  $\gamma$ -HCH,  $\beta$ -HCH, d-HCH, heptachlor,  $\beta$ -heptachlor epoxide, aldrin,  $\alpha$ -endosulfan, o,p-DDE, dieldrin, p,p-DDE, p-DDD, endrin, p,p-DDD,  $\beta$ -endosulfan, p,p-DDT, o,p-DDT, endrin aldehyde, endosulfan sulfate, methoxychlor, trifluralin, chlorothalonil, dichlofluanid and endrin ketone. Six PCB congeners were determined: PCB-28, PCB-52, PCB-101, PCB-138, PCB-153, PCB-180.

The limits of detection (LOD) and quantification (LOQ, i.e. 3.3 times the LOD) are defined and reported in Table S1. Individual compounds were above LOD in all samples, and values below LOQ were substituted by LOQ/2, unless were noted. Pollutant concentrations were calculated based on analytical curves using specific standards. Quality control samples included blank, spike and reference samples, and were analysed simultaneously with the target samples to ensure reliability and comparability of quantifications. Blanks (analyte-free) were treated identically to target samples to ensure that the working material was not contaminated, and no blank correction was applied as values were below LOD in blanks. In spike samples, individual analytes were spiked prior to extraction. The recovery of the spiked analytes was used to check the accuracy of the analytical method.

### 2.3. Stable isotope analysis

Freeze-dried and homogenized blood samples were placed in tin capsules for the stable isotope analysis of carbon ( $\delta^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ). The analysis was carried out at the *Centro Integrado de Análises* (CIA-FURG, Brazil) with an Isotope Ratio Mass Spectrometer (IRMS, ThermoFisher) coupled to an elemental analyser (EA Flash, 2000 Delta V Advantage). Stable isotope ratios were calculated in comparison to the international standards, Vienna Pee Dee belemnite for carbon and atmospheric air for nitrogen (Peterson and Fry, 1987). Relative abundances of stable isotopes are expressed in delta ( $\delta$ ) notation, in parts per mil (‰). Caffeine, acetinilide and glutamic acid were used as laboratory standards and analysed simultaneously. Analysis of laboratory standards corresponded to an accuracy of 0.07‰ ( $\delta^{13}\text{C}$ ) and 0.3‰ ( $\delta^{15}\text{N}$ ).

### 2.4. Data analysis

All analyses were performed in R version 4.3.1 (R Core Team, 2021). Compounds in each sample were summed to obtain  $\Sigma\text{PCB}$ ,  $\Sigma\text{OHP}$  and  $\Sigma\text{PAH}$  concentrations. Then, comparisons of  $\Sigma\text{PCB}$ ,  $\Sigma\text{OHP}$  and  $\Sigma\text{PAH}$  concentrations between seabird species were conducted separately for each tissue (i.e. blood and feathers) and sampling year (2019 and 2022), using a non-parametric Kruskal-Wallis followed by a post-hoc Dunn test, with the package 'dunn.test' (Quinn and Keough, 2002; Dinno, 2024). Non-parametric testing was used due to the low and varying number of samples with quantified pollutants between species and years. P-values lower than 0.05 were considered significant for interspecific pairwise comparisons. Organic pollutant concentrations obtained in other studies were summarized in Table S8 for comparisons.

To calculate the contribution (%) of compounds to the concentration of  $\Sigma\text{PCB}$ ,  $\Sigma\text{OHP}$  and  $\Sigma\text{PAH}$  of each seabird species, tissue and year, we used only the quantified concentrations (values above LOQ). In this analysis, therefore, we focused on the quantified pollutant values, so that the compound contributions were not influenced by the substituted values in the dataset. To enhance visualization, PAHs were classified in accordance to the number of rings (di to hexa-cyclic); and OHPs were separated in HCHs, DDTs, endosulfans, drins (e.g. aldrin, dieldrin), CUPs (dichlofluanid, chlorothalonil and trifluralin), and the rest of compounds (heptachlor,  $\beta$ -heptachlor epoxide and methoxychlor) were grouped as 'other' (Table S1).

Biplots of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values in blood were used to assess variation

in stable isotopes among seabird species and sampling periods, and Kruskal-Wallis post-hoc Dunn test to determine interspecific differences within sampling years. Moreover, to assess potential associations between stable isotopes and log-transformed  $\Sigma\text{OHP}$ ,  $\Sigma\text{PCB}$  and  $\Sigma\text{PAH}$  in blood samples, we computed a Spearman correlation matrix (Legendre and Legendre, 2012), followed by a principal component analysis (PCA) using the FactoMineR package, with scaled values (Husson et al., 2017). Then, a redundancy analysis (RDA) was implemented using  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values as explanatory variables and pollutant classes as response variables, to determine explanatory power of isotopes. Variance inflation factor (VIF) was checked between isotopes and was below 2. A forward model selection was implemented, and the significance of the selected model and variables was assessed.

## 3. Results

### 3.1. Organohalogen pesticides (OHPs)

In 2019, concentrations of  $\Sigma\text{OHPs}$  in the blood and feathers were higher in the booby, followed by the tropicbird, petrel and the albatross; with the exception that for blood, the two samples analysed for the albatross had also high concentrations of  $\Sigma\text{OHP}$  (Table 1, Table S2, S3; Fig. 2). Differences in 2019 were not significant for blood, but for feathers, the booby had higher concentration than the petrel ( $Z = 2.16$ ,  $p = 0.031$ ) and the albatross ( $Z = 3.55$ ,  $p < 0.001$ ). Feather  $\Sigma\text{OHP}$  concentrations were also higher in the tropicbird than the albatross in 2019 ( $Z = 3.27$ ,  $p = 0.001$ ). For blood and feather samples collected in 2022, the tropicbird had lower  $\Sigma\text{OHP}$  concentrations than the booby and the petrel ( $p < 0.05$ ).

Strikingly, mean  $\Sigma\text{OHP}$  in blood samples were approximately five times higher in 2019 than 2022; and 18 to 41 times higher in feathers from 2019 than 2022 (Table 1, Table S2, S3). Regarding the composition of  $\Sigma\text{OHP}$  in 2019,  $\Sigma\text{DDTs}$  were quantified across species and sample types (6.3–35.7% of quantified  $\Sigma\text{OHP}$ ), except in petrel feather samples, that had only values below LOQ (Fig. 2).  $\Sigma\text{HCH}$  and  $\Sigma\text{Drins}$  were detected throughout sample types and species and constituted from 3.7 to 40.3% of quantified  $\Sigma\text{OHP}$  in 2019. In 2019,  $\Sigma\text{Endosulfans}$  were not quantified in the feathers of the petrel and the blood of the albatross in 2019, and constituted up to 15.8% of quantified  $\Sigma\text{OHP}$  in the feather of the albatross. The  $\Sigma\text{CUPs}$  constituted 9.6–47% of quantified  $\Sigma\text{OHP}$  in samples from 2019, and class 'other' contributed with 6–42% to the samples from this year. In 2022, however, a number of OHP compounds were not quantified in seabird samples (all samples below LOQ), such as DDTs and CUPs. HCHs were only quantified ( $>\text{LOQ}$ ) in feathers of the booby in 2022, and drins in the feathers of the booby and blood of the petrel (Tables S2 and S3).  $\Sigma\text{Endosulfan}$  had the greatest overall contribution to quantified  $\Sigma\text{OHP}$  in 2022, constituting between 50 and 100% of the quantified concentrations (Fig. 2). For tropicbirds, OHPs were quantified only in a single blood sample among the 17 analysed in 2022.

### 3.2. Polychlorinated biphenils (PCBs)

For samples collected in 2019, the albatross presented the lowest concentrations of  $\Sigma\text{PCB}$  in both feathers and blood in relation to all other species (Fig. 3, Table 1, Table S4, S5). Samples from the booby had the highest blood  $\Sigma\text{PCB}$  concentrations in 2019, followed by the tropicbird and the petrel. In 2022, blood samples from the tropicbird were the lowest considering  $\Sigma\text{PCB}$ , significantly lower in comparison to the booby and the petrel ( $p < 0.05$ ) (Fig. 3). Feather  $\Sigma\text{PCB}$  in 2022 were the lowest in the petrel, significantly in relation to the booby ( $Z = 2.83$ ,  $p = 0.004$ ).

Remarkably, mean  $\Sigma\text{PCB}$  was 14–20 times higher in 2019 than 2022 for blood, and 45 to 113 times higher in feathers in 2019 than 2022 (Table 1). PCB-180 was the predominant congener in samples of the albatross in 2019, constituting 100% and 38.3% of the quantified  $\Sigma\text{PCB}$  concentration in blood and feathers, respectively. Also in 2019, PCB-101 was the major quantified congener in blood and feathers of the booby

**Table 1**

Concentrations (mean ± 1 standard deviation; ng/mL for blood and ng/g for feathers) of organohalogen pesticides (OHPs), polycyclic aromatic hydrocarbons (PAHs) and total polychlorinated biphenils (PCBs) in the blood and feathers of brown boobies (*Sula leucogaster*), red-billed tropicbirds (*Phaethon aethereus*), Trindade petrels (*Pterodroma arminjoniana*) and Atlantic yellow-nosed albatrosses (*Thalassarche chlororhynchos*) sampled in the southwestern Atlantic Ocean in 2019 and 2022. Values below the limit of quantification (LOQ) were substituted by LOQ/2. When all samples were below LOQ, '<LOQ' is shown for clarity. Summaries of individual compounds are provided in the supplementary material (Tables S2–S7). DT = percentage of samples above LOQ, or the number of samples analysed for isotopes. ns = not sampled.

Compound	Tissue/ year	Brown booby			Red-billed tropicbird			Trindade petrel			Atlantic yellow-nosed albatross		
		DT	Mean ± SD	Min-Max	DT	Mean ± SD	Min-Max	DT	Mean ± SD	Min-Max	DT	Mean ± SD	Min-Max
∑HCH	Blood/ 2019	100	1.77 ± 1.04	0.82–4.23	80	3.25 ± 4.98	0.82–4.23	20	0.92 ± 0.89	0.5–2.66	100	2.93 ± 1.96	1.55–4.32
	Blood/ 2022	0	<LOQ	<LOQ	0	<LOQ	<LOQ	0	<LOQ	<LOQ	ns	ns	ns
	Feather/ 2019	100	34.96 ± 37.56	12.62–136.88	100	21.06 ± 12.85	12.62–136.88	100	23.52 ± 22.43	4.92–71.62	89	2.95 ± 1.97	0.5–5.51
	Feather/ 2022	30	0.89 ± 0.7	0.5–3	0	<LOQ	<LOQ	0	<LOQ	<LOQ	ns	ns	ns
∑DDT	Blood/ 2019	60	4.25 ± 4.58	0.49–10.81	50	3.87 ± 4.63	0.49–10.81	70	5.38 ± 12.27	0.49–40.18	100	2.63 ± 0.64	2.18–3.09
	Blood/ 2022	0	<LOQ	<LOQ	0	<LOQ	<LOQ	0	<LOQ	<LOQ	ns	ns	ns
	Feather/ 2019	50	23.61 ± 60.07	0.49–191.9	50	8.04 ± 16.37	0.49–191.9	0	<LOQ	<LOQ	44	2.56 ± 4.2	0.49–13.26
	Feather/ 2022	0	<LOQ	<LOQ	0	<LOQ	<LOQ	0	<LOQ	<LOQ	ns	ns	ns
∑Drins	Blood/ 2019	70	2.27 ± 1.66	0.62–5.62	80	2.63 ± 3.33	0.62–5.62	50	3.87 ± 5.27	0.62–15.57	100	2.63 ± 1.06	1.88–3.38
	Blood/ 2022	0	<LOQ	<LOQ	0	<LOQ	<LOQ	8	0.64 ± 0.06	0.62–0.84	ns	ns	ns
	Feather/ 2019	100	13.11 ± 8.8	2.06–29.99	60	7.8 ± 9.23	2.06–29.99	90	5 ± 3.5	0.62–10.67	67	2.68 ± 2.32	0.62–7.28
	Feather/ 2022	10	0.67 ± 0.14	0.62–1.13	0	<LOQ	<LOQ	0	<LOQ	<LOQ	ns	ns	ns
∑Endosulfans	Blood/ 2019	60	0.95 ± 0.8	0.37–2.86	20	0.46 ± 0.21	0.37–2.86	20	2.4 ± 4.9	0.37–15.61	0	<LOQ	<LOQ
	Blood/ 2022	65	0.63 ± 0.24	0.37–1.07	6	0.4 ± 0.12	0.37–1.07	32	0.49 ± 0.22	0.37–1.17	ns	ns	ns
	Feather/ 2019	50	3.26 ± 4.24	0.37–13.84	40	8 ± 12.02	0.37–13.84	0	<LOQ	<LOQ	44	3.96 ± 5.26	0.37–14.46
	Feather/ 2022	90	0.77 ± 0.28	0.37–1.24	42	0.6 ± 0.3	0.37–1.24	93	0.81 ± 0.26	0.37–1.24	ns	ns	ns
∑CUP	Blood/ 2019	100	2.95 ± 1.8	0.56–6.11	80	1.74 ± 1.16	0.56–6.11	70	2.19 ± 3.25	0.37–9.5	100	7.03 ± 1.61	5.89–8.18
	Blood/ 2022	0	<LOQ	<LOQ	0	<LOQ	<LOQ	0	<LOQ	<LOQ	ns	ns	ns
	Feather/ 2019	100	31.93 ± 31.25	2.43–100.82	100	50.24 ± 58.45	2.43–100.82	100	5.77 ± 4.03	1.56–14.56	100	8.17 ± 5.97	1.48–19.46
	Feather/ 2022	0	<LOQ	<LOQ	0	<LOQ	<LOQ	0	<LOQ	<LOQ	ns	ns	ns
∑Other OHPs	Blood/ 2019	90	2.59 ± 1.11	0.38–3.72	80	0.91 ± 0.49	0.38–3.72	60	1.31 ± 1.6	0.38–5.18	100	1.13 ± 0.07	1.07–1.18
	Blood/ 2022	0	<LOQ	<LOQ	0	<LOQ	<LOQ	4	0.39 ± 0.08	0.38–0.76	ns	ns	ns
	Feather/ 2019	80	25.74 ± 17.28	0.38–50.32	90	27.96 ± 25.12	0.38–50.32	90	24.76 ± 18.76	0.38–61.18	89	4.83 ± 2.49	0.38–9.32
	Feather/ 2022	5	0.39 ± 0.06	0.38–0.64	0	<LOQ	<LOQ	3	0.4 ± 0.11	0.38–1	ns	ns	ns
∑OHP	Blood/ 2019	100	15.14 ± 5.39	5.42–22.22	90	12.98 ± 11.47	5.42–22.22	100	17.48 ± 27.11	3.48–88.14	100	16.85 ± 2.12	15.36–18.35
	Blood/ 2022	65	3.1 ± 0.24	2.85–3.54	6	2.88 ± 0.12	2.85–3.54	40	3 ± 0.24	2.85–3.83	ns	ns	ns
	Feather/ 2019	100	132.73 ± 73.31	23.32–304.84	100	124.92 ± 85.49	23.32–304.84	100	60.05 ± 39.76	19.59–127.6	100	26.06 ± 14.1	5.43–45.47
	Feather/ 2022	90	3.71 ± 0.78	2.85–6.1	42	3.08 ± 0.3	2.85–6.1	93	3.3 ± 0.27	2.85–3.73	ns	ns	ns
∑PAH	Blood/ 2019	100	8.58 ± 2.14	4.42–10.62	100	5.52 ± 2.32	4.42–10.62	60	6.09 ± 5.6	3.36–21.4	100	28.91 ± 7.3	23.75–34.08
	Blood/ 2022	100	8.57 ± 1.07	6.72–11.3	100	7.92 ± 0.97	6.72–11.3	100	7.94 ± 1.02	5.26–9.42	ns	ns	ns
	Feather/ 2019	100	346.8 ± 103.21	136.2–493.88	100	375.26 ± 235.96	136.2–493.88	100	284.44 ± 256.71	82.59–860.46	100	33.74 ± 8.26	22.5–44.44

(continued on next page)

Table 1 (continued)

Compound	Tissue/ year	Brown booby			Red-billed tropicbird			Trindade petrel			Atlantic yellow-nosed albatross		
		DT	Mean $\pm$ SD	Min-Max	DT	Mean $\pm$ SD	Min-Max	DT	Mean $\pm$ SD	Min-Max	DT	Mean $\pm$ SD	Min-Max
$\Sigma$ PCB	Feather/ 2022	100	8.55 $\pm$ 0.99	6.9–11.13	100	8.32 $\pm$ 0.81	6.9–11.13	100	8.54 $\pm$ 0.99	6.41–11.4	ns	ns	ns
	Blood/ 2019	100	4.66 $\pm$ 4.47	1.1–14.42	90	1.88 $\pm$ 1.64	1.1–14.42	100	6.16 $\pm$ 12.37	0.12–37.41	100	0.26 $\pm$ 0.04	0.24–0.29
	Blood/ 2022	45	0.31 $\pm$ 0.37	0.03–0.96	29	0.09 $\pm$ 0.18	0.03–0.96	84	0.45 $\pm$ 0.28	0.03–0.9	ns	ns	ns
	Feather/ 2019	70	21.96 $\pm$ 31.92	0.03–102.72	100	30.89 $\pm$ 40.42	0.03–102.72	100	25.97 $\pm$ 32.41	0.12–110.32	100	4.05 $\pm$ 1.99	0.6–6.57
$\delta^{13}\text{C}$ (‰)	Feather/ 2022	85	0.48 $\pm$ 0.35	0.03–0.98	58	0.4 $\pm$ 0.36	0.03–0.98	37	0.23 $\pm$ 0.32	0.03–0.97	ns	ns	ns
	Blood/ 2019	8	-17.1 $\pm$ 0.6	-18.6- (-16.7)	10	-17.2 $\pm$ 0.4	-18(-16.4)	6	-18.3 $\pm$ 0.2	-18.6- (-18.0)	2	-18.9 $\pm$ 0.1	-19- (-18.9)
$\delta^{15}\text{N}$ (‰)	Blood/ 2022	20	-18.5 $\pm$ 0.4	-19.6- (-17.6)	19	-18.3 $\pm$ 0.4	-19(-17.8)	25	-19.2 $\pm$ 0.1	-19.6- (-18.8)	ns	ns	ns
	Blood/ 2019	8	10 $\pm$ 0.9	8.6–11.2	10	9.4 $\pm$ 1	8.4–11.9	6	11.8 $\pm$ 0.6	11–12.7	2	13.2 $\pm$ 1	12.5–13.8
$\delta^{15}\text{N}$ (‰)	Blood/ 2022	20	10.9 $\pm$ 0.4	10.1–11.6	19	8.7 $\pm$ 1.2	7.4–12.9	25	11.1 $\pm$ 0.4	10.3–11.8	ns	ns	ns

and tropicbird (37.8–62.9% of quantified  $\Sigma$ PCB), whereas PCB-138 (47%) and PCB-52 (46.8%) were dominant in petrel blood and feathers, respectively (Fig. 3; Table 1). In 2022, PCB-28 was the major congener in booby blood and feathers, as well as in tropicbird and petrel feathers (36.2–96.4% of quantified  $\Sigma$ PCB). PCB-101 (67.1% of quantified  $\Sigma$ PCB) and PCB-52 (52.8% of quantified  $\Sigma$ PCB) predominated in tropicbird and petrel blood in 2022, respectively.

### 3.3. Polycyclic aromatic hydrocarbons (PAHs)

For  $\Sigma$ PAH in 2019 blood samples, the highest concentration was recorded in the two samples of the albatross (Table 1, Table S6, S7; Fig. 4). In this year, blood  $\Sigma$ PAH was significantly higher in the booby and the albatross than the petrel and the tropicbird ( $p < 0.05$ ). Interestingly, in feathers from 2019, only the albatross presented significantly lower  $\Sigma$ PAH than all other species ( $p < 0.05$ ). Considering the samples from 2022, both for blood and feathers, concentrations were not significantly different between the species.

Mean  $\Sigma$ PAH in blood were of similar magnitudes between 2019 and 2022, while for feathers, the concentrations were between 33 and 45 times higher in 2019 in comparison to 2022. In 2019, compounds with four and five rings were predominant for booby, tropicbird and petrel, while three and four ring molecules were for the albatross. Compounds with six rings were only detected in 2019. In 2022, three and four ring molecules were the main constituents of quantified  $\Sigma$ PAH in the blood and feather of all seabird species (Fig. 3).

### 3.4. Stable isotopes and relationships with pollutants in blood

In 2019,  $\delta^{13}\text{C}$  values were contrastingly different between the pelagic species (albatross and petrel) and Abrolhos' seabirds (booby and tropicbird). The albatross and petrel had average  $\delta^{13}\text{C}$  values between -18.3‰ and -18.9‰, while the booby and tropicbird had values around -17‰ (Fig. 5; Table 1). The albatross and petrel had also higher  $\delta^{15}\text{N}$  values in 2019, with tropicbird and booby having lower and similar values ( $Z = -0.98$ ,  $p = 0.656$ ). Nonetheless, the petrel and booby  $\delta^{15}\text{N}$  values were non-statistically different in 2019 ( $Z = 2.19$ ,  $p = 0.112$ ), but petrel values were higher than those of tropicbirds ( $Z = -3.20$ ,  $p = 0.008$ ). In 2022, tropicbirds and boobies had  $\delta^{13}\text{C}$  values lower than 2019, but still their values were significantly higher than the petrel ( $Z = 5.87$ ,  $p < 0.001$ ;  $Z = -4.73$ ,  $p < 0.001$ , respectively). Regarding  $\delta^{15}\text{N}$ , the tropicbird had significantly lower values than the booby ( $Z = -4.13$ ,  $p < 0.001$ ) and the petrel ( $Z = -5.50$ ,  $p < 0.001$ ). Booby and petrel  $\delta^{15}\text{N}$

values did not differ in 2022 ( $Z = 1.17$ ,  $p = 0.241$ ).

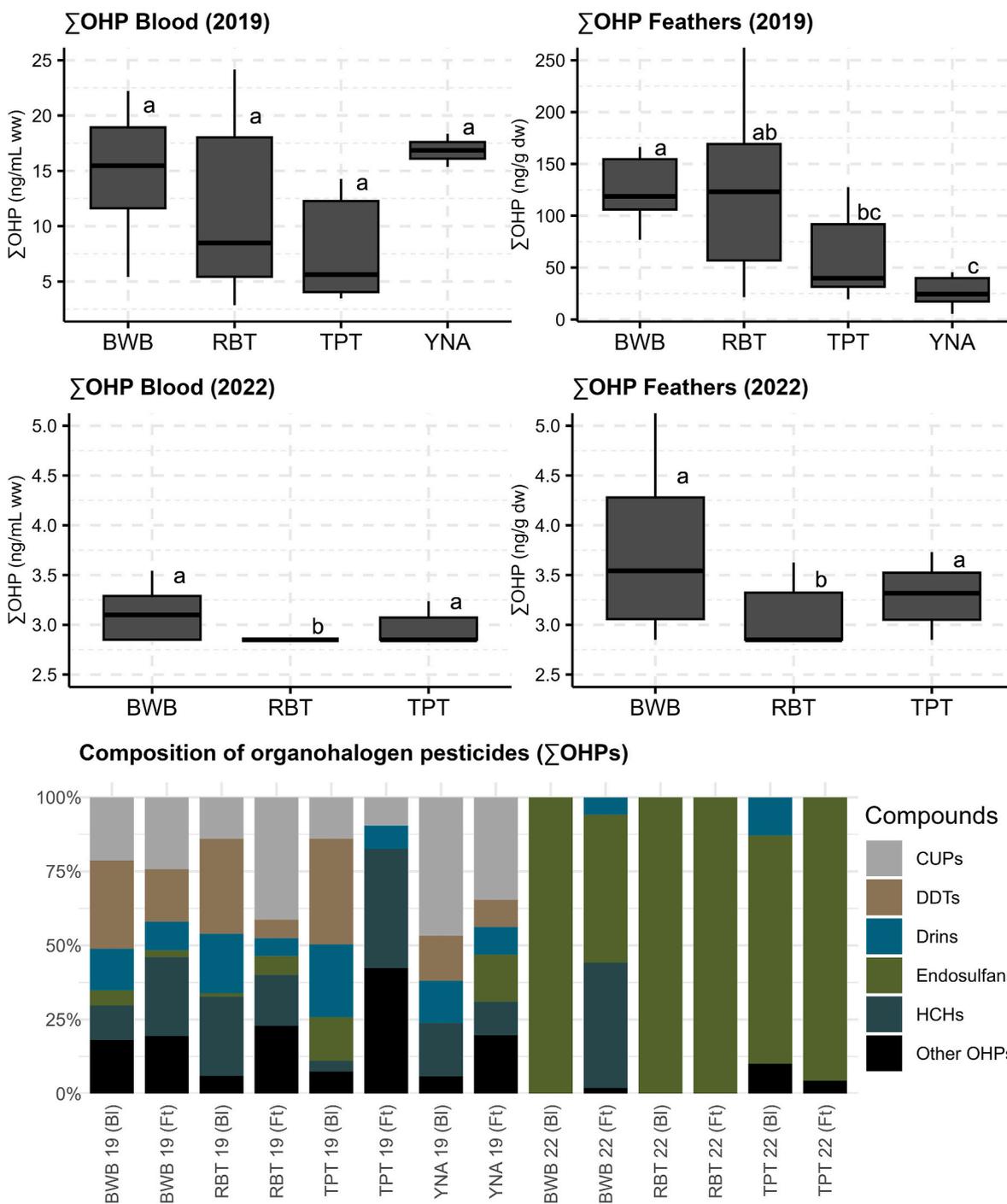
The correlation matrix revealed a negative correlation between  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  ( $r = -0.45$ ,  $p < 0.001$ ). Log-transformed concentrations of  $\Sigma$ OHP and  $\Sigma$ PCB were positively correlated ( $r = 0.58$ ,  $p < 0.001$ ), and both these pollutants were correlated with  $\delta^{13}\text{C}$  ( $r = 0.47$ ,  $p < 0.001$ ;  $r = 0.31$ ,  $p = 0.004$ , respectively). In contrast, correlations of  $\delta^{15}\text{N}$  with pollutants were not significant, and log-transformed PAH did not show significant correlations (Fig. 6). The first component of the PCA captured 41.9% of variation in data, and had the largest contribution from log-OHP (36.6%), logPCB (31.2%) and  $\delta^{13}\text{C}$  (31.1%), with a low influence of logPAH and  $\delta^{15}\text{N}$  (<1% each). The second component had the largest contributions from  $\delta^{15}\text{N}$  (45.6%) and logPAH (30.1%), followed by  $\delta^{13}\text{C}$  (12.2%) (Fig. 6). The selected RDA model included both isotopes as explanatory variables (adjusted  $R^2 = 0.30$ ,  $F = 18.19$ ,  $p = 0.001$ ). Both isotopes were significant predictors ( $p < 0.05$ ), with total variance explained of 28.6% for  $\delta^{13}\text{C}$  and 13.3% for  $\delta^{15}\text{N}$ . The first axis (RDA1) was significant in the model ( $F = 36.03$ ,  $p = 0.001$ ) and had a stronger influence from  $\delta^{13}\text{C}$  (score = 0.74) compared to  $\delta^{15}\text{N}$  (score = 0.26). Conversely, the second axis (RDA2) was not significant ( $F = 0.36$ ,  $p = 0.61$ ), and had a high positive influence from  $\delta^{15}\text{N}$  (score = 0.96) and a negative influence from  $\delta^{13}\text{C}$  (score = -0.67).

## 4. Discussion

Organic pollutants in blood and feathers of the four seabird species varied according to their different foraging strategies in the tropical southwestern Atlantic Ocean. In general, higher pollutant concentrations appear to be associated with nearshore foraging, with higher concentrations observed in the booby, decreasing in more pelagic species. This pattern was generally consistent between blood and feathers. The findings suggest an increased continental input of organic pollutants, which poses a potential higher risk to seabirds along this region of the Brazilian coast. Relationships of pollutants in blood and isotopic carbon contribute to the assumption of higher pollutant concentrations in coastal foragers. Nonetheless, results from the different organic pollutant classes varied by tissue and sampling year, both in concentration and chemical profile, which suggests that exposure is highly dynamic in the tropical seabird community.

### 4.1. Interspecific variation of organic pollutants in seabirds

Persistent organic pollutants in the blood and feathers of tropical seabirds showed interspecific and temporal differences. Concentrations

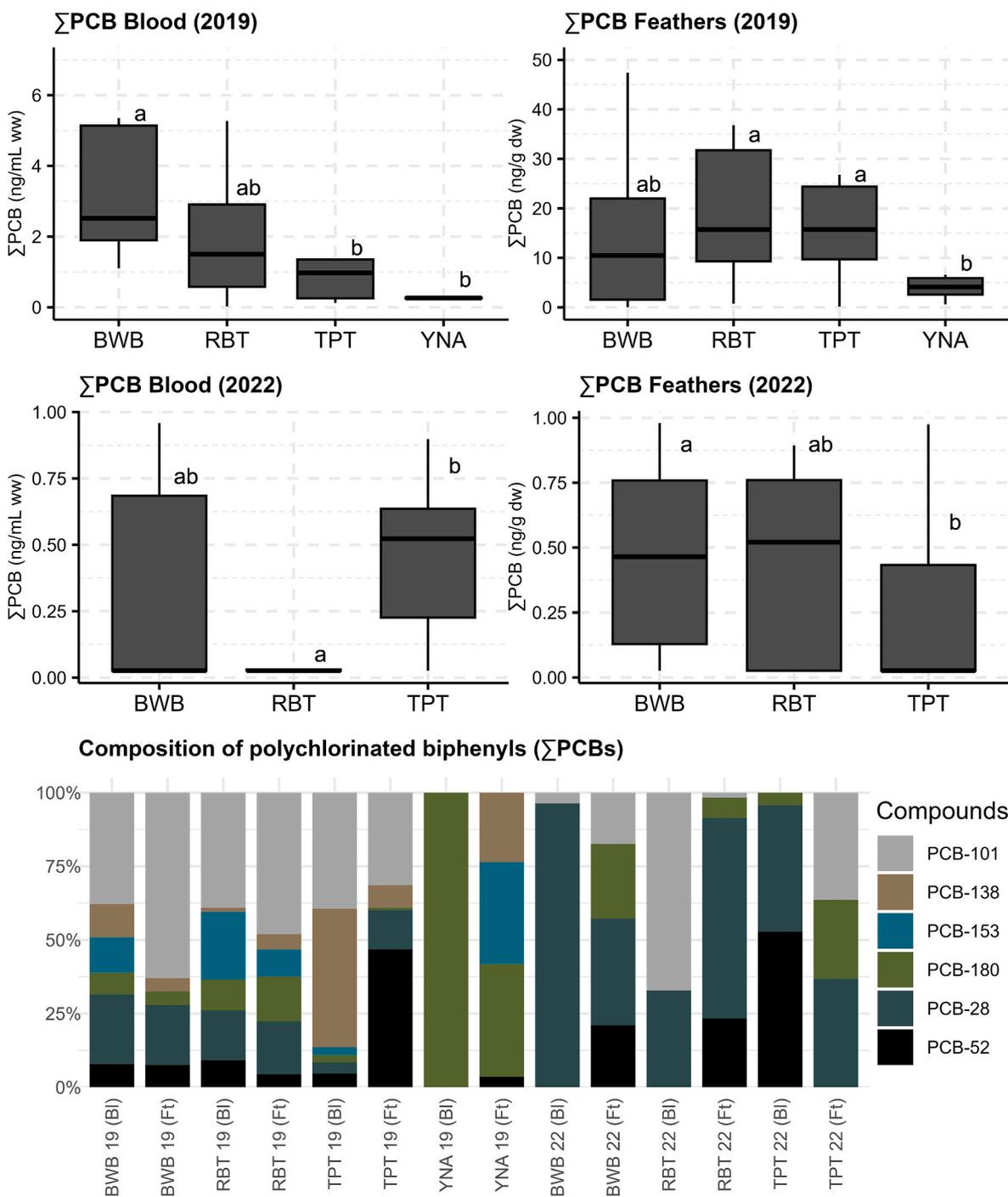


**Fig. 2.** Concentration (ng/mL for blood and ng/g for feathers) and composition of organohalogen pesticides ( $\Sigma$ OHP) in the blood and feathers of the brown booby (BWB; *Sula leucogaster*), red-billed tropicbird (RBT; *Phaethon aethereus*), Trindade petrel (TPT; *Pterodroma arminjoniana*) and Atlantic yellow-nosed albatross (YNA; *Thalassarche chlororhynchos*). Boxplots (graphic panels above) represent the median, first and third quartile. Different letters above boxes represent significant differences ( $p < 0.05$ ) after pairwise Dunn's test. Stacked barplots represent the percentage of contribution of OHP groups to the total quantified (above LOQ) concentration of  $\Sigma$ OHP in the blood (BI) and feather (Ft) of seabirds sampled in 2019 and 2022. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

of  $\Sigma$ OHP and  $\Sigma$ PCB in both sampling years were generally the highest in the booby (Figs. 2–4), which is the seabird foraging closest to the coast, suggesting association with the land-based pollutant sources, such as agriculture and industrial activities. In 2019, the tropicbird had also high  $\Sigma$ PCB and  $\Sigma$ OHP, but concentrations in 2022 blood samples were low in the tropicbirds in relation to the booby and the petrel. Although the tropicbird breeds near the coast (70 km), it relies on offshore waters near the continental shelf break, approximately 140 km from the coast

and in 1750 m of depth (Linhares et al., 2024). Lower concentrations in 2022 blood samples from tropicbirds may be related to a lower exposure due to offshore foraging, in relation to the sympatric booby, or having a lower trophic position than the booby and petrel, as suggested by the  $\delta^{15}\text{N}$  values (Linhares et al., 2024).

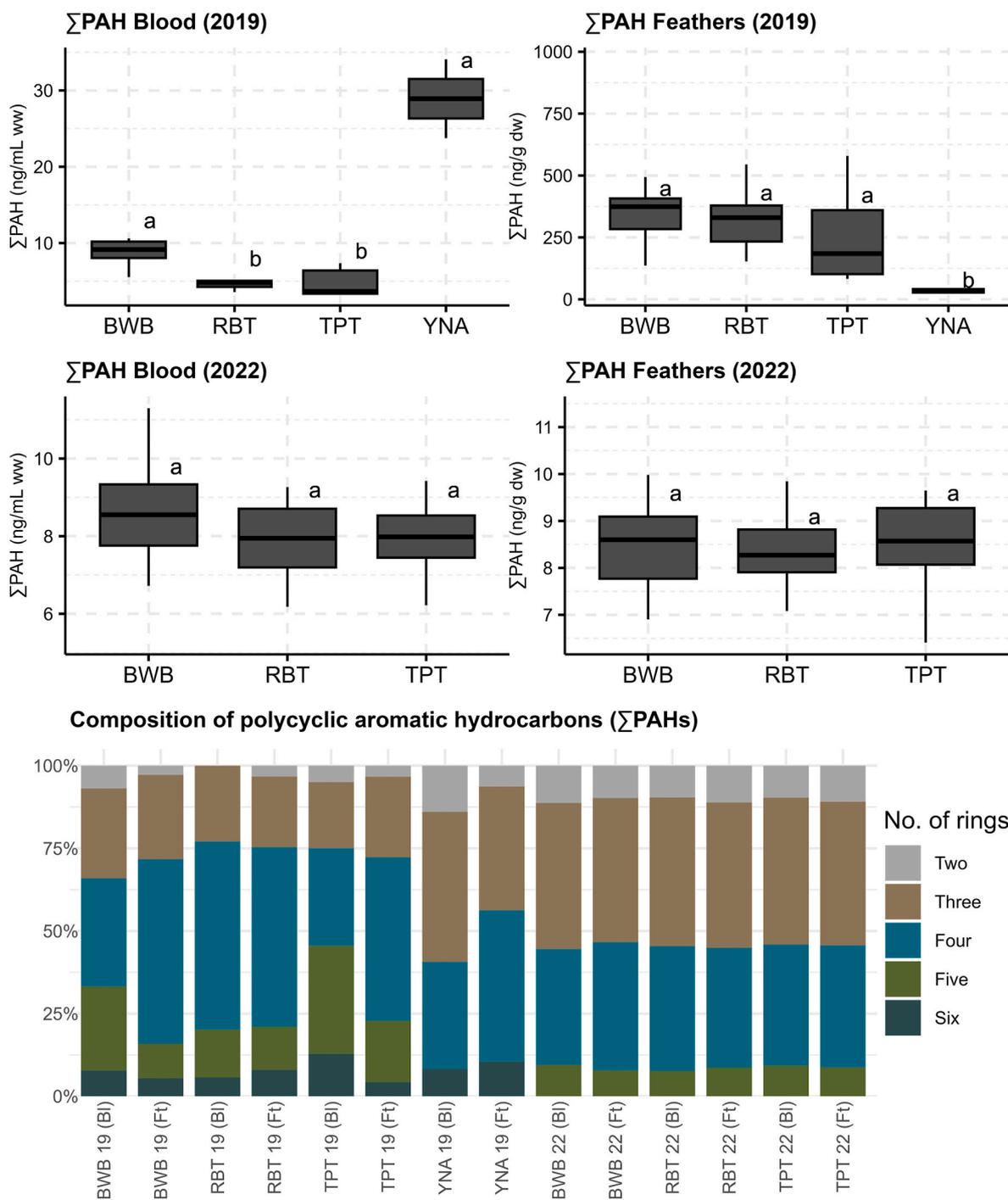
As for the pelagic species, the petrel and albatross had generally lower concentrations of  $\Sigma$ PCB and  $\Sigma$ OHP than the Abrolhos seabirds in 2019, but the concentrations of these pollutants in samples from 2022



**Fig. 3.** Concentration (ng/mL for blood and ng/g for feathers) and composition of polychlorinated biphenyls ( $\Sigma$ PCB) in the blood and feathers of the brown booby (BWB; *Sula leucogaster*), red-billed tropicbird (RBT; *Phaethon aethereus*), Trindade petrel (TPT; *Pterodroma arminjoniana*) and Atlantic yellow-nosed albatross (YNA; *Thalassarche chlororhynchus*). Boxplots (graphic panels above) represent the median, first and third quartile. Different letters above boxes represent significant differences ( $p < 0.05$ ) after pairwise Dunn's test. Stacked barplots represent the percentage of contribution of PCB congeners to the total quantified (above LOQ) concentration of  $\Sigma$ PCB in the blood (BI) and feather (Ft) of seabirds sampled in 2019 and 2022. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

were similar in the petrel in relation to the booby, and higher than the tropicbird in most cases, except for  $\Sigma$ PCB in feathers. Although the petrel breeds ~1200 km from the coast in Trindade Island, it forages over a large oceanic region and migrates to the North Atlantic during the non-breeding period (Leal et al., 2017). Exposure of petrel to pollutants, therefore, may be highly variable, depending on individual-specific foraging sites (Leal and Bugoni, 2021), especially considering that feathers may have been grown during the non-breeding period. The

petrel also occupies a high trophic position in the food web, therefore susceptible to trophic magnification (Mancini et al., 2014; Elliott et al., 2021). Likewise, the albatross breeds in the middle of the subtropical South Atlantic, forages along a large pelagic area (Gabani, 2020; del Hoyo et al., 2023), and occupies a high trophic position from feeding on discards of longline fisheries targeting tuna and sharks (Bugoni et al., 2010). Nonetheless, feathers from the albatross in 2019 had the lowest concentrations of all organic pollutants among seabirds, which suggests

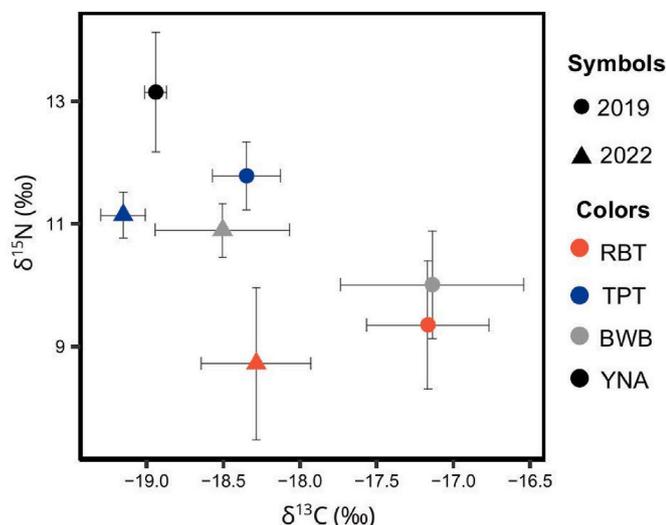


**Fig. 4.** Concentration (ng/mL for blood and ng/g for feathers) and composition of polycyclic aromatic hydrocarbons ( $\Sigma$ PAH) in the blood and feathers of the brown booby (BWB; *Sula leucogaster*), red-billed tropicbird (RBT; *Phaethon aethereus*), Trindade petrel (TPT; *Pterodroma arminjoniana*) and Atlantic yellow-nosed albatross (YNA; *Thalassarche chlororhynchos*). Boxplots (graphic panels above) represent the median, first and third quartile. Different letters above boxes represent significant differences ( $p < 0.05$ ) after pairwise Dunn's test. Stacked barplots represent the percentage of contribution of PAHs with different number of rings to the total quantified (above LOQ) concentration of  $\Sigma$ PAH in the blood (Bl) and feather (Ft) of seabirds sampled in 2019 and 2022. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

lower long-term exposure from its broad offshore foraging areas, or alternatively, different detoxification mechanisms or metabolic rates for this species.

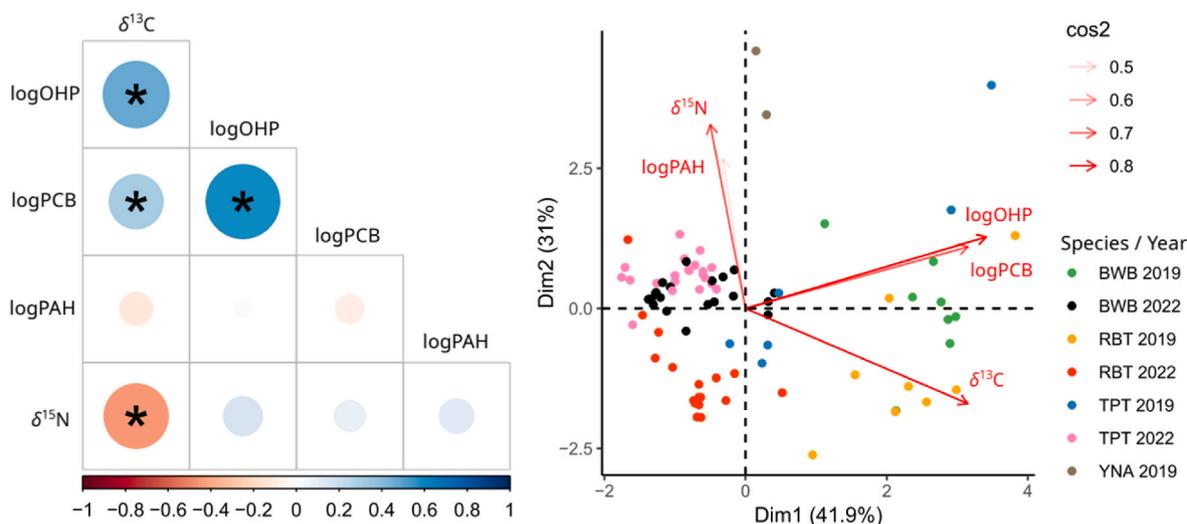
As for  $\Sigma$ PAH, in general, interspecific differences appear to be less consistent than for  $\Sigma$ PCB and  $\Sigma$ OHP, which is in line with some PAHs being metabolized and excreted (Shilla and Routh, 2018; Provencher et al., 2020, 2022; Jodice et al., 2023). Nonetheless, in both year and tissues, the booby had similar or higher  $\Sigma$ PAH than the tropicbird and

the petrel, and higher than the albatross considering the feathers, which may still suggest increased nearshore contamination. However, although the albatross had the lowest  $\Sigma$ PAH in feathers, its blood concentration was the highest, with a mean value three to five times those of the other sampled species. Besides the low number of samples precludes strong inferences, such marked difference in blood concentrations suggest an increased short-term PAH exposure of the albatross in the study area. These albatrosses were sampled in the Campos Basin, offshore Rio



**Fig. 5.** Carbon ( $\delta^{13}\text{C}$ ) and nitrogen ( $\delta^{15}\text{N}$ ) stable isotope values (mean  $\pm$  standard deviation) in blood of red-billed tropicbird (RBT; *Phaethon aethereus*) and brown booby (BWB; *Sula leucogaster*) breeding on Abrolhos Archipelago, Trindade petrel (TPT; *Pterodroma arminjoniana*) breeding on Trindade Island, and Atlantic yellow-nosed albatross (YNA; *Thalassarche chlororhynchos*) sampled at sea in the southwestern Atlantic Ocean in 2019 and 2022. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

de Janeiro, which is a major oil extraction area in Brazil. Magellanic penguins (*Spheniscus magellanicus*) sampled in the Campos Basin have been suggested to have high PAH concentrations in liver potentially associated with the oil industry activities in the area (Quinete et al., 2020). As the blood represents short-term exposure, we assume that albatross blood samples reflect contamination from the sampling area in the Campos Basin. However, a larger sample size of the albatross over a longer period would have benefited our study to support inferences related to the increased PAH concentrations in the blood of this species. We suggest further investigation of PAH contamination in seabirds foraging in oil extraction areas in Brazil, in order to evaluate the impact of this activity related to marine pollution (Quinete et al., 2020).



**Fig. 6.** Relationships among log-transformed concentrations of polychlorinated biphenyls ( $\Sigma\text{PCB}$ ), organohalogen pesticides ( $\Sigma\text{OHP}$ ), polycyclic aromatic hydrocarbons ( $\Sigma\text{PAH}$ ) and stable nitrogen ( $\delta^{15}\text{N}$ ) and carbon ( $\delta^{13}\text{C}$ ) isotope values in the blood of tropical seabirds. Spearman correlation (left) showing the correlation coefficients (colored circles) and asterisks representing significant associations ( $p < 0.05$ ). Principal Component Analysis (right) showing seabird data points in the first (Dim1) and second (Dim2) components, according to the species and year of collection.

#### 4.2. Composition of organic pollutants and comparison with other studies

There was substantial variation in  $\Sigma\text{OHP}$  concentration and composition between years, with the  $\Sigma\text{OHP}$  being four to 40 times higher in 2019 than 2022. Composition of quantified  $\Sigma\text{OHP}$  was more diverse in 2019, with  $\Sigma\text{CUP}$ ,  $\Sigma\text{DDT}$  and  $\Sigma\text{HCH}$  showing contributions of four to 40.3%, while in 2022  $\Sigma\text{endosulfan}$  predominated (70–100%) quantified  $\Sigma\text{OHP}$ . In blood,  $\Sigma\text{DDT}$  was the highest in the petrel in 2019 ( $5.38 \pm 12.27$  ng/mL), similar to that obtained by Silva et al. (2023) in the same species sampled in Trindade in 2017 ( $7.27$  ng/mL). This  $\Sigma\text{DDT}$  concentration was greater than reported by Gilmour et al. (2019) in frigatebirds (*Fregata magnificens* and *F. minor*) and boobies (*S. leucogaster*, *S. dactylatra* and *S. sula*) in the Pacific and Caribbean ( $2.33$ – $3.73$  ng/mL), and for Bermuda gadfly petrels (*Pterodroma cahow*,  $2.92 \pm 1.99$  ng/mL, Campioni et al., 2024). Conversely, DDTs were not quantified in the petrel feathers in our study, but reached up to  $23.61 \pm 60.07$  ng/g in the booby and decreased to  $2.56 \pm 4.2$  ng/g in the albatross, while in 2022 values were below LOQ in both blood and feathers. Adrogué et al. (2019) analysed DDT in feathers of the black-browed albatross (*Thalassarche melanophris*) and the Cape petrel (*Daption capense*) in Argentina and obtained DDT concentrations of  $2.6$ – $4$  ng/g. DDT was banned in Brazil in 2002, but Dicofol (synthesized from DDT) continued to be used until it was banned in 2015 and could have been a continued source of DDT to the environment. In comparison to the above-mentioned studies, values reported in this study for tropical seabird feathers in 2019 could be considered high, but there are not many assessments of organic pollutants in seabird feathers and studies vary in their quantification units, which makes comparisons difficult.

Along with DDTs, HCHs were one of the most applied pesticides in South America primarily through the use of Lindane, which was banned in Brazil in 2006 (IBAMA, 2006). The highest blood  $\Sigma\text{HCH}$  occurred in the tropicbird in 2019 ( $3.3 \pm 4.98$  ng/mL), which is higher than reported for other tropical seabirds ( $0$ – $3.03$  ng/mL; Gilmour et al., 2019) and from Trindade Island in 2017 ( $0$ – $0.06$  ng/mL; Silva et al., 2023). In liver samples from tropical seabirds offshore Brazil (including *S. leucogaster*), mean values of  $1.78$ – $4.44$  ng/mL of  $\Sigma\text{HCH}$  were reported (Dias et al., 2018), while for Magellanic penguins, mean values were from  $7.75$  ng/mL in muscle and  $17.9$  ng/mL in liver (Quinete et al., 2020). In feather samples, mean values of  $\Sigma\text{HCH}$  in our study were from  $3$  ng/g in the albatross to  $35$  ng/g in the booby in 2019, while values from  $5$  to  $12$  ng/g were reported in Procellariiformes from Argentina

(Adrogué et al., 2019). In 2022, HCHs, DDTs and drins were largely not quantified in seabird samples in our study, apart from the booby and the petrel. Interestingly, endosulfans were the major class of OHPs quantified in seabird samples from 2022, comprising 100% in some cases. This insecticide was intensively used in Brazil in the coffee and soybean culture, banned in 2013 in Brazil and Argentina; while it is still allowed in other countries like China and India (Fang et al., 2016; Yadav et al., 2016; Montory et al., 2017). Composition of endosulfan isomers can help inform on their historical use, as  $\alpha$ - and  $\beta$ -endosulfan are converted to endosulfan sulfate (Weber et al., 2010) and, in our study, endosulfan sulfate was the only quantified isomer in 2022 (6–93% of values above LOQ), which suggests exposure from past input (Tables S2 and S3). Inversely, Adrogué et al. (2019) in Argentina found higher concentrations of  $\alpha$  endosulfan and suggested that this might have resulted from exposure to fresh inputs of endosulfan.

PCB concentration and composition showed substantial variation between years, with heavier congeners with higher chlorination number prevailing in the quantified  $\Sigma$ PCB in 2019, and lighter and less chlorinated congeners prevailing in 2022. Heavier congeners are more lipophilic and tend to be biomagnified, while lighter congeners are more favourable for excretion and more volatile (Jaspers et al., 2007; Cipro et al., 2012). Moreover, mean  $\Sigma$ PCB in samples from 2019 were 14–112 times higher than in samples from 2022. Concentrations of  $\Sigma$ PCB in blood in 2019 (0.3–6.2 ng/mL mean) were within the range quantified for tropical seabirds in the Pacific Ocean (1.62–7.81 ng/mL; Gilmour et al., 2019), in Bermuda petrels (7.87 ng/mL; Campioni et al., 2024). These  $\Sigma$ PCB values were higher than most species studied in offshore islands in Brazil (0.12–1.14 ng/mL), with the exception of the petrel samples from 2017 analysed by Silva et al. (2023) (24.29 ng/mL). Considering feather concentrations, apart from the lower values in the albatross ( $4.0 \pm 2.0$  ng/g), the mean for birds in 2019 (22–30.9 ng/g) were higher than reported for the black-browed albatross and the Cape petrel in Argentina (7.3–9.6 ng/g; Adrogué et al., 2019). In 2022, the low mean values from our study (0.09–0.5 ng/mL or ng/g for blood and feathers) suggests temporal variation in PCB exposure in seabirds from the Brazilian coast.

Blood  $\Sigma$ PAH did not show substantial temporal variation, while feather concentrations were between 33 and 45 times higher in 2019 than 2022. Apart from the blood samples of the albatross ( $28.9 \pm 7.3$  ng/mL), all other seabirds had mean values ranging between 5.5 and 8.6 ng/mL. Bermuda petrels had  $3.95 \pm 2.88$  ng/mL mean blood  $\Sigma$ PAH (Campioni et al., 2024), the brown pelican (*Pelecanus occidentalis*) had a range between 42.44 and 245.47 ng/mL in the Gulf of Mexico (Jodice et al., 2023), and the yellow-legged gull (*Larus michahellis*) 75.15–101.17 ng/mL in non-oiled areas in Spain (Pérez et al., 2008; Seegar et al., 2015). Compared with results from these studies,  $\Sigma$ PAH observed in blood do not appear to represent a concern for seabird health in our study. Nonetheless, feather mean  $\Sigma$ PAH in our study ranged from 8.3 in 2022 to 375.3 ng/g in 2019. There is paucity of information of PAHs in bird feathers to enable a comparison of these values, but Jodice et al. (2023) reports between 32.40 and 166.70 ng/g in brown pelicans. Measured in liver, seabirds in Canada had values between 0.52 and 36.91 ng/mL (Provencher et al., 2020; Provencher et al., 2022), while 8.52–32.5 ng/mL was recorded in Procellariiformes from the North Atlantic and Mediterranean (Roscales et al., 2011). In contrast, in the Campos Basin, Magellanic penguins had  $141 \pm 273$  ng/mL measured in muscle and  $1711 \pm 2859$  ng/mL in liver. It is unclear if the high concentrations of PAHs recorded in feathers from 2019 are causes of concern. Furthermore, it appeared to have occurred a shift from a higher contribution of hexa and hepta-cyclic hydrocarbons in 2019 to di, three and tetra-cyclic in 2022. PAHs with fewer rings have low molecular weight, are generally associated with petrogenic origin, while high molecular weight PAHs are associated with pyrogenic sources (Eisler, 1987).

Temporal variation of all pollutants reported here point to higher contamination in tropical seabirds in 2019 than 2022. Remarkably,

quantified OHP constitution in 2019 included several legacy pesticides, as well as CUPs that are still applied in agriculture. Although the lack of data across multiple years obscures strong conclusions, the higher concentrations of pollutants in the first year could have been influenced by the variation in the discharge from the Doce River to the ocean (PMBA, 2021), which is the largest river in the eastern coast of Brazil. In 2015, the tailings from the Fundão Dam rupture reached the Doce River and potentially remobilized organic pollutants deposited along the river margins, which was suggested in previous studies to have caused an increase of these pollutants in the continental shelf, including pesticides (Oliveira-Ferreira et al., 2022; Cabral et al., 2023; Yamamoto et al., 2023). Studies that monitored the impact of the dam failure have shown temporal fluctuations in trace metal concentrations in the environment and wildlife, including Abrolhos seabirds, following the climatic cycles of the Doce River, which influence contaminant bioavailability (e.g. Costa et al., 2022; Bauer et al., 2024). Our results raise questions about the potential influence of the Fundão Dam failure in the organic contaminants quantified in seabirds in the samples from 2019. Unfortunately, monitoring of the environmental impacts of the Fundão Dam failure started only in 2018, therefore samples from these species have not been analysed during the first three years after the disaster. We advocate for faster action of the environmental government sector to respond to such catastrophic events, in order to achieve effective impact assessment.

#### 4.3. Relationship with isotopic values

The species analysed here had distinct  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values, which are generally attributed to differences in foraging habitat and trophic position, respectively (Fry, 2006; Tatsch et al., 2024). The albatross had the highest  $\delta^{15}\text{N}$  values, which is in line with the species foraging on discards of the longline fishery in Brazil (Bugoni et al., 2010). In a previous isotopic study of tropical seabirds in offshore islands in Brazil (Mancini et al., 2014), the petrel had high  $\delta^{15}\text{N}$  values, as it had also here, followed by the booby and tropicbird. Moreover, both Abrolhos seabirds had higher  $\delta^{13}\text{C}$  values in 2019 than 2022, and also higher than the petrel and the albatross, which suggests a greater influence from the neritic ocean zone for these species breeding near the coast in Abrolhos (Tatsch et al., 2024), especially in the first sampling year.

Multivariate analyses elucidated some potential relationships of seabird ecology, based on isotopic tracers, with pollutant concentrations. Log-transformed  $\Sigma$ PCB and  $\Sigma$ OHP were positively associated to  $\delta^{13}\text{C}$  values, which suggests higher contamination near the coast, where  $\delta^{13}\text{C}$  values are generally higher. Seabird trophic position (indicated by  $\delta^{15}\text{N}$ ) also had a marginal influence on pollutants, as indicated by the RDA, and the lack of a stronger effect can likely be explained by the higher concentrations of pollutants in Abrolhos seabirds in 2019, when  $\delta^{15}\text{N}$  values were lower. This result, nonetheless, suggests a greater influence of foraging habitat than trophic position in pollutant concentrations in our study. For PAH, relationships with isotopic tracers or other pollutants were not significant, which is consistent with PAHs not having clear relationships with trophic position and being readily metabolized and excreted by seabirds (Eisler, 1987; Roscales et al., 2011). A larger sample size of different species over greater temporal and spatial scales could be useful to further explore the potential relationships between isotopic markers and pollutant concentrations in tropical seabirds.

#### 4.4. Conclusions

Organic pollutant concentrations in the four seabird species varied in relation to foraging habitat (nearshore vs. pelagic), trophic position and time. The species associated with nearshore foraging and higher trophic position had generally higher pollutant concentrations, suggesting that organic pollution in these seabirds is influenced by continental sources to the ocean. Furthermore, the higher pollutant concentrations in 2019

suggests drastic temporal variation in marine organic pollution, which could be associated to variation in the input from land-based sources to the ocean. Although it is unclear if the organic pollutant concentrations reported here are solely a cause of concern for seabird health, previous studies have demonstrated high concentrations and drastic temporal variation of toxic metal(loid)s in the booby and tropicbird from Abrolhos (Bauer et al., 2024). These alarming results may point to continuous contaminant exposure of seabirds foraging along this portion of the Brazilian coast, and highlights the need to understand the temporal variation and the potential impacts of marine pollutants on seabirds. For this, a more frequent sampling and association with physiological response biomarkers could be relevant. Monitoring demographic and health parameters, associated with variations in contamination in seabirds, particularly those species threatened by extinction, is highly recommended.

### CRedit authorship contribution statement

**Bruno de Andrade Linhares:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Patrícia Gomes Costa:** Writing – original draft, Resources, Methodology, Data curation. **Leandro Bugoni:** Writing – review & editing, Supervision, Project administration, Investigation, Funding acquisition, Conceptualization. **Guilherme Tavares Nunes:** Writing – review & editing, Supervision, Project administration, Methodology, Investigation, Conceptualization. **Adalto Bianchini:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Investigation, Funding acquisition, Conceptualization.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2025.125928>.

### Data availability

Data will be made available on request.

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