Environmental Pollution 323 (2023) 121187

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

A century of mercury: Ecosystem-wide changes drive increasing contamination of a tropical seabird species in the South Atlantic Ocean☆

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ARTICLE INFO

Keywords:
Feathers
Hg
Museum
Sooty tern
Stable isotopes
Temporal monitoring

ABSTRACT

Mercury (Hg) is a highly toxic metal that adversely impacts human and wildlife health. The amount of Hg released globally in the environment has increased steadily since the Industrial Revolution, resulting in growing contamination in biota. Seabirds have been extensively studied to monitor Hg contamination in the world's oceans. Multidecadal increases in seabird Hg contamination have been documented in polar, temperate and subtropical regions, whereas in tropical regions they are largely unknown. Since seabirds accumulate Hg mainly from their diet, their trophic ecology is fundamental in understanding their Hg exposure over time. Here, we used the sooty tern (*Onychoprion fuscatus*), the most abundant tropical seabird, as bioindicator of temporal variations in Hg transfer to marine predators in tropical ecosystems, in response to trophic changes and other potential drivers. Body feathers were sampled from 220 sooty terns, from museum specimens (n = 134) and free-living birds (n = 86) from Ascension Island, in the South Atlantic Ocean, over 145 years (1876–2021). Chemical analyses included (i) total- and methyl-Hg, and (ii) carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotopes, as proxies of foraging habitat and trophic position, respectively, to investigate the relationship between trophic ecology and Hg contamination over time. Despite current regulations on its global emissions, mean Hg concentrations were 58.9% higher in the 2020s (2.0 μg g^{-1}, n = 34) than in the 1920s (1.2 μg g^{-1}, n = 107). Feather Hg concentrations were negatively and positively associated with $\delta^{13}C$ and $\delta^{15}N$ values, respectively. The sharp decline of 2.9 ‰ in $\delta^{13}C$ values over time indicates ecosystem-wide changes (shifting primary productivity) in the tropical South Atlantic Ocean and can help explain the observed increase in terns' feather Hg concentrations. Overall, this study provides invaluable information on how ecosystem-wide changes can increase Hg contamination of tropical marine predators and reinforces the need for long-term regulations of harmful contaminants at the global scale.

☆ This paper has been recommended for acceptance by Michael Bank.

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https://doi.org/10.1016/j.envpol.2023.121187
Received 7 November 2022; Received in revised form 10 January 2023; Accepted 30 January 2023
Available online 31 January 2023

1. Introduction

Mercury (Hg) is a toxic metal and its impacts on human health are a major concern. Globally, the amount of Hg released into the environment has steadily increased since the Industrial Revolution, resulting in a three-to five-fold increase of Hg on land, in the atmosphere and the ocean (Lamborg et al., 2014; Selin, 2009). Since Hg is primarily emitted in the atmosphere in its volatile form (Hg^0) by both natural and human sources (Eagles-Smith et al., 2018) where it remains for up to one year (Streets et al., 2019), it disperses widely before its deposition in all global ecosystems. Consequently, even remote oceanic regions are affected by this global pollutant. Once in the sea, Hg is methylated by microorganisms into methyl-Hg (MeHg), its most toxic and bioavailable form. Because of its high assimilation efficiency and strong affinity for proteins, MeHg bioaccumulates in marine organisms (*i.e.*, concentrations increase over time in their tissues) and biomagnifies in food webs (*i.e.,* concentrations increase at each trophic level). Marine food resources represent the principal source of Hg for humans and wildlife. Mercury can cause severe physical and neurological harm and even mortality at sufficiently high concentrations, as is the case in Minamata Disease (Takeuchi et al., 1962). This has led to new regulations, such as the Minamata Convention on Mercury, to protect human health and the environment, by controlling and reducing anthropogenic releases of Hg globally.

Oceans receive 80% of total atmospheric Hg deposition, 49% of which is restricted to tropical oceans (Horowitz et al., 2017). Despite this, little is known about the interactions between Hg and ecosystem function in the intertropical zone. Yet, it is here that artisanal and small-scale gold mining occurs, resulting in annual Hg emissions >720 tonnes. Gold mining is the largest contributor to Hg emissions and represents >35% of total global anthropogenic Hg emissions (Eagles-Smith et al., 2018). In addition, tropical oceans have recently experienced increased oxygen-depletion (Breitburg et al., 2018), creating favourable conditions for MeHg formation (Cossa, 2013; Mason et al., 2012; Sunderland et al., 2009). Quantifying Hg contamination and its temporal trends in tropical oceans and wildlife is thus urgently needed.

Monitoring tropical oceans is logistically challenging but can be achieved through investigations of bioindicators that feed extensively across their waters and, thus, provide a lens through which to investigate Hg contamination. Seabirds are effective bioindicators (Burger and Gochfeld, 2004; Furness, 1993) because of their high position in marine trophic webs and longevity, allowing them to integrate and reflect Hg contaminations of their food webs (Fort et al., 2016; Furness and Camphuysen, 1997; Piatt et al., 2007). Furthermore, most seabirds are colonial (*i.e.*, they concentrate in high numbers in breeding colonies) and philopatric (*i.e.*, they have high site fidelity). Thus, several individuals can be sampled simultaneously, and repeatedly through time, in particular through non-lethal collection of feathers (Albert et al., 2019). Feathers are a reliable tissue to investigate Hg contamination in seabirds (Thompson et al., 1998), because dietary MeHg accumulates in body tissues between moulting episodes (Furness et al., 1986). Up to 90% of the accumulated MeHg is sequestered into growing feathers after transportation in the blood and then excreted during moult (Honda et al., 1986). In feathers, MeHg binds to sulfhydryl groups of keratin molecules (Crewther et al., 1965) to form strong, stable bonds that withstand rigorous physical treatments over time (Appelquist et al., 1984). Mercury contained in feathers is mostly in the form of MeHg (>90%, Renedo et al., 2017) and thus seabird feathers are commonly used for Hg biomonitoring over the short- and the long-term (*e.g.*, Albert et al., 2019; Burger and Gochfeld, 2004; Cherel et al., 2018).

Seabirds have been collected for exhibitions and museum collections worldwide over the last centuries and, therefore, their specimens are invaluable to study temporal trends of Hg contamination (Vo et al., 2011). However, inorganic Hg salts were used historically by museum curators as preservatives in specimen preparation, resulting in methodological bias when analysing museum *versus* free-living specimens. This bias can be overcome by analysing MeHg specifically (Hogstad et al., 2003). Overall, only a limited number of studies ($n = 14$) have quantified Hg in museum-held seabird specimens over the last century, and only four of these have considered trophic ecology as a factor that could influence Hg contamination (see Supplementary Material, Table S1 for species and temporal comparisons). Yet, bird trophic ecology is key to understanding the mechanisms of Hg contamination and to disentangle whether temporal variations in Hg contamination are linked to dietary shifts and/or to changes in environmental Hg contamination (Bond et al., 2015; Carravieri et al., 2016; Choy et al., 2022; Fort et al., 2016; Vo et al., 2011). Over multidecadal timescales, decreases and stable trends in Hg contamination have been documented in only one temperate Atlantic species (Thompson et al., 1992) and one temperate Pacific species (Choy et al., 2022), respectively. In contrast, increases in Hg contamination have been observed in 12 temperate Atlantic species (Monteiro and Furness, 1997; Thompson et al., 1992; Thompson et al., 1993), one temperate/subtropical Pacific species (Vo et al., 2011) and one Arctic species (Bond et al., 2015). Notably, none of these studies involved long-term investigation of tropical regions.

The sooty tern (*Onychoprion fuscatus*) is the most abundant tropical seabird, widely distributed in all three ocean basins. This species is therefore an ideal candidate to monitor Hg contamination in tropical food webs. The present work extends the investigation of Reynolds et al. (2019), who highlighted long-term changes in the feeding ecology of sooty terns breeding on Ascension Island (the largest colony in the South Atlantic Ocean), by using carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotopes, which are proxies of feeding habitat (*i.e.,* carbon source) and diet (*i.e.,* trophic position), respectively (Kelly, 2000). Reynolds et al. (2019) found long-term declines in both feather $\delta^{13}C$ and $\delta^{15}N$ values since the 1890s, indicating a fundamental change in their foraging behaviour, including a dietary shift from predominantly teleost fish to squid coinciding with a dramatic decline in the size of the breeding population on Ascension Island. Here, our aim was three-fold to: (i) document the temporal trend of feather Hg concentrations in Ascension sooty terns between the 1890s and the 2020s, (ii) investigate the influence of their trophic ecology (reflected by $\delta^{13}C$ and $\delta^{15}N$ values) on this temporal trend and (iii) compare feather Hg contamination before and after population collapse. As for other remote oceanic regions, we predict that there will be a general increase in feather Hg concentrations to the present day, linked to the previously documented changes in their diet and foraging behaviours.

2. Materials and methods

2.1. Study area and museum specimens

Ascension Island (07°57' S, 14°24' W) is an isolated 97 km^2 volcanic island in the tropical South Atlantic Ocean. It accommodates the largest breeding population of sooty terns in the entire Atlantic Ocean, containing on average > 200,000 pairs (Hughes et al., 2012). Currently, they nest on bare ground across the Wideawake Fairs – an Important Bird Area (IBA SH009) – at two Nature Reserves (NRs) called Mars Bay and Waterside Fairs. As epipelagic seabirds, sooty terns feed on small pelagic fish and squid caught at/ near the ocean surface (Ashmole, 1963a; Reynolds et al., 2019). Globally, sooty terns are listed as of 'Least Concern' in the IUCN Red List of Threatened Species. Yet, the population from Ascension Island has drastically declined by approximately 84% between the 1960s and 1990s from a peak of approximately two million birds (Hughes et al., 2017). Many factors may have contributed to this precipitous decline, including the depletion of food resources and predation by introduced species such as domestic cats (*Felis silvestris catus*), black rats (*Rattus rattus*) and common mynas (*Acridotheres tristis*) (Reynolds et al., 2019 and references therein).

2.2. Feather sampling and preparation

Sooty terns from Ascension Island breed sub-annually (~10 months; Reynolds et al., 2014). While still breeding, they start their post-nuptial moult (*i.e.,* basic moult) and replace all of their feathers (i.e., flight and body) during the following weeks/months (Ashmole, 1963b). Therefore, body feathers collected during the breeding season provide information on bird Hg exposure since the previous moult (*i.e.* for the past 10 months; Albert et al., 2019). This includes different stages of their life cycle: (i) the end of the previous breeding season, (ii) the post-breeding migration, (iii) the non-breeding period, (iv) the pre-breeding migration and (v) the beginning of the current breeding season. Consequently, feathers also provide information at different spatial scales, since sooty terns have a maximum non-breeding range of 2900 km in tropical waters around Ascension Island (Reynolds et al., 2021). Feathers also retain dietary signatures at the time of their synthesis, as carbon and nitrogen isotopes are incorporated into feathers as they grow *(i.e.,* over a few weeks). Consequently, Hg concentrations and stable isotope values of feathers are temporally decoupled (Thompson et al., 1998; Bond, 2010). Nonetheless, stable isotope values can provide invaluable information about seabird foraging ecology across century scales by virtue of their constant integration over time, thereby allowing for long-term understanding of Hg temporal trends.

Historical feather samples were collected from skins held in 10 natural history museum collections (see details in Supplementary Table S2). Details of museum skins are provided in Supplementary Table S3. The complete procedure for historical sampling and preparation is detailed in Reynolds et al. (2019). Only specimens with known collection date (*i.e.*, year or decade) were considered for the current study. When the precise year of specimen collection was unknown ($n = 7$), we attributed the first year of the corresponding decade to it. Sex of birds from which skins were prepared was known for some historical samples (*i.e.*, 43 females and 41 males from the 1920s). Feather sampling of free-living birds was carried out at Mars Bay NR and Waterside Fairs NR breeding colonies in 2006 by BJH ($n = 8$), 2012 by CPW and SJR ($n = 40$) and 2020 ($n = 4$) and 2021 ($n = 30$) by LS. These samples were collected by licenced professionals under environmental research permits issued by Ascension Island Government. Incubating sooty terns were caught during ringing activities using a soft mesh hand-held net. While processing the birds (Redfern and Clark, 2001), contour feathers were removed from across the breast on both sides of the keel ridge. Samples were collected from breeding adults and stored at room temperature prior to preparation and analysis.

To eliminate any external contamination, all feathers were cleaned in the laboratory with a chloroform:methanol mixture (2:1), sonicated for 3 min and rinsed twice in methanol. They were then oven-dried for 48 h at 45°C. For each bird specimen, several feathers (2–4) were pooled to derive the individual mean value (Carravieri et al., 2014; Jaeger et al., 2009), then cut with stainless scissors into a homogeneous powder to be analysed for both stable isotopes and Hg.

2.3. Stable isotope analyses

Carbon and nitrogen stable isotopes were analysed in homogenized subsamples of body feathers. Results are expressed in the δ unit notation as deviations from standards (Vienna Pee Dee Belemnite for $\delta^{13}C$ and atmospheric N_2 for $\delta^{15}N$) following the formula:

$$\delta^{13}C \text{ or } \delta^{15}N = \left(R_{sample}/R_{standard} - 1\right) \times 10^3$$

where R is $^{13}C/^{12}C$ or $^{15}N/^{14}N$, respectively. Analyses of historical samples (*i.e.*, from the 1890s to the 2010s) were performed at the MARE and results were presented in Reynolds et al. (2019). The present study extends this previous investigation by another decade to the present day. Analyses of these contemporary feathers (*i.e.,* from the 2020s) were performed at the LIENSs. Feather subsamples (~0.4 mg) were weighed with a microbalance and loaded into tin cups. Values of $\delta^{13}C$ and $\delta^{15}N$ were determined with a continuous flow mass spectrometer (Delta V Plus with a Conflo IV Interface, Thermo Scientific, Bremen, Germany) coupled to an elemental analyzer (Flash 2000 or EA Isolink, Thermo Scientific, Milan, Italy). The analytical precision was <0.10 ‰ and <0.15 ‰ for $\delta^{13}C$ and $\delta^{15}N$ values, respectively. Ten samples were analysed in both laboratories to compare analytical performances. No difference in $\delta^{13}C$ (ANOVA, $F_{1,18} = 0.04$, $p = 0.80$) nor $\delta^{15}N$ values (ANOVA, $F_{1,18} = 0.008$, $p = 0.90$) was detected between the two laboratories, ensuring data comparability between the older (from Reynolds et al., 2019) and most recent feather samples.

Since the Industrial Revolution, the combustion of human fossil fuel has increased atmospheric CO_2, resulting in an accelerating decrease in biosphere $\delta^{13}C$, mainly influenced by two additive effects: (i) an increase in phytoplankton fractionation (Rau et al., 1992), and (ii) the so-called « Suess-Effect» (Keeling, 1979), whereby anthropogenic carbon has lower $\delta^{13}C$ values than natural background carbon. In our study, raw $\delta^{13}C$ values were thus corrected accordingly, following calculations from previous work (Eide et al., 2017; Hilton et al., 2006; Jaeger and Cherel, 2011; Körtzinger et al., 2003). For further information, see Supplementary Figure S1 for a comparison of isotopic niches of raw and corrected $\delta^{13}C$ values.

2.4. Mercury analyses

Generally, total Hg (THg) includes both inorganic (iHg) and organic Hg (*i.e.,* MeHg). In feathers, MeHg accounts for >90% of THg (Bond and Diamond, 2009; Renedo et al., 2017; Thompson and Furness, 1989). So, THg can be used as a proxy of MeHg in feathers. Here, THg analyses were performed on homogenized feathers using an Advanced Mercury Analyzer spectrophotometer (Altec AMA 254). Each sample was analysed in duplicate-triplicate to guarantee a relative standard deviation <10%. Accuracy was verified by running certified reference material (DOLT-5, Fish liver, National Research Council, Canada: certified Hg concentration = 0.44 ± 0.18 $\mu g\ g^{-1}$ dry weight [dw]). Our measured values were 0.43 ± 0.01 $\mu g\ g^{-1}$ dw ($n = 14$). Blanks were run at the beginning of each set of samples. Detection limit of the AMA was 0.01 ng. Mercury concentrations are expressed in $\mu g\ g^{-1}$ dw.

Historically, inorganic Hg salts were used as preservatives in museum collections until approximately the 1970s, creating a major bias in THg comparison between old and recent specimens (Thompson et al., 1992). Therefore, Hg speciation analyses were performed on the oldest specimens ($n = 131$; until the 1970s) to quantify both MeHg and iHg (Renedo et al., 2017, 2018) from feather homogenates (1.1–12.0 mg), using a GC-ICP-MS Trace Ultra GC equipped with a Triplus RSH autosampler coupled to an ICP-MS XSeries II (Thermo Scientific, Bremen, Germany). Mercury speciation analyses were also performed on a few free-living individuals ($n = 3$, from the 2000s) to determine the precise proportion of MeHg in sooty tern feathers and hence validate the use of THg as proxy of MeHg after the 1970s. Further methodological details are provided in Supplementary Material, including a comparison of speciation results between old and recent specimens (Table S4). Certified reference material (NIES-13, human hair; certified MeHg concentration = 3.80 ± 0.40 $\mu g\ g^{-1}$ dw) was analysed for validation of feather analyses (keratin-based matrices). Our measured concentrations for MeHg were 3.64 ± 0.09 $\mu g\ g^{-1}$ dw ($n = 3$). Method recovery was checked by comparison of THg values measured with AMA and the equivalent Σ MeHg + iHg obtained from speciation analyses. Further details on recovery calculations are shown in Table S5. Average recovery of sooty tern feather Hg was 96.1 ± 4.9% (92.3–101.3%; $n = 3$; see Supplementary Material). Subsequently, feather Hg concentrations reflect MeHg concentrations before the 1970s and THg concentrations thereafter.

2.5. Data analyses

All statistical analyses and graphical representations were carried out in R 4.0.3 (R Core Team, 2020).

2.5.1. Temporal trends of stable isotopes and Hg over 145 years

Year-specific sample sizes were unbalanced (Table 1), with five times more observations on average in 1925 than in other years. Thus, 50 values were randomly extracted from this specific year and used in all models. One outlier was identified in 1915 (*i.e.,* JR11, Table S3) and removed from all analyses. Data exploration revealed that Hg, $\delta^{13}C$ and $\delta^{15}N$ varied non-linearly with time. Therefore, Generalized Additive Models (GAMs) were run to investigate non-linear temporal trends over 145 years, using the « mgcv » package (Wood, 2011). Each variable was modeled independently against year as a smoother. For each model, we adapted the smoothed term (k) to avoid overfitting and over-simplification, and obtain the best and most realistic fit to the data ($k = 5$ for Hg, $\delta^{13}C$ and $\delta^{15}N$ values). Model assumptions were checked *via* residual analysis through the « gam. check » function.

2.5.2. Drivers of Hg contamination over time

Multifactorial analyses were used to test for the effect of trophic ecology on feather Hg concentrations over time. To discard the combined effect of time on both feeding habitat and trophic position, we detrended all variables by extracting residuals from GAM temporal trends of Hg, $\delta^{13}C$ and $\delta^{15}N$ values. The new response variable was time-detrended Hg concentrations and continuous explanatory variables were the time-detrended $\delta^{13}C$ and $\delta^{15}N$ values. Relationships between detrended variables were then investigated with linear models using the « nlme » package (Pinheiro et al., 2020). Prior to model construction, one-way ANOVA was used to test for differences in Hg concentrations between the sexes of terns. As no difference was detected ($F_{1,82} = 2.82$, $p = 0.10$; Supplementary Figure S2), sex was not included in model selection procedures. Models were GAMs with a Gaussian distribution and an identity link function. The initial model was: detrended Hg ~ detrended $\delta^{13}C$ + detrended $\delta^{15}N$. All potential combinations of variables (Table 2) were subjected to model selection based on the Akaike's Information Criterion adjusted for small sample sizes (AIC_c), using the « MuMIn » package (Bartoń, 2022). The model with the lowest AIC_c value, with a difference of AIC_c of two or more compared to the next model (ΔAIC_c), was selected as the best model (Burnham and Anderson, 2002). Model performance was assessed using Akaike weights (w_i) (after Johnson and Omland, 2004) and model fit was checked by residual analysis of the initial model.

To compare temporal trends with the published literature, we calculated the percentage of change (increase or decrease) for evolution phases identified by GAM temporal trends. Details of empirical calculations are provided in the Supplementary Material (Table S6).

Table 2

AIC_c model ranking from statistical analyses of feather Hg concentrations from sooty terns breeding on Ascension Island. Models are Generalized Additive Models (GAMs) with a Gaussian distribution and an identity link function. Variables were all time-detrended (see Section 2.5.2 for further details). Abbreviations: k, number of parameters; AIC_c, Aikaike's Information Criterion adjusted for small sample sizes; w_i AIC_c weights.

Models	k	AIC_c	ΔAIC_c[a]	w_i[b]
Detrended $\delta^{13}C$ + Detrended $\delta^{15}N$	**4**	**184.52**	**0.00**	**0.96**
Detrended $\delta^{13}C$	3	191.49	6.97	0.03
Detrended $\delta^{15}N$	3	193.75	9.23	0.01
NULL	2	206.61	22.09	0.00

[a] A model with $\Delta AIC_c = 0$ is interpreted as the best model among all the selected ones (in bold).

[b] Weights are cumulative (sum to 1).

2.5.3. Temporal comparison: before and after population collapse

Following Reynolds et al. (2019), we tested for differences in trophic ecology and Hg concentrations of sooty terns relative to their population collapse on Ascension Island (*i.e.,* before: 1890s–1940s; after: 1970s–2020s). First, a Multivariate Analysis of Variance (MANOVA) enabled us to test for differences in isotopic niches (*i.e.,* $\delta^{13}C$ and $\delta^{15}N$ values combined) between periods. Differences between periods for individual isotopes were then identified using the « summary. aov » function. Residual normality (« mvnormtest package »; Jarek, 2012), outliers (« rstatix » package; Kassambara, 2022) and multicollinearity (between $\delta^{13}C$ and $\delta^{15}N$ values; « cor. test » function) were checked prior to interpretation. Secondly, one-way ANOVA was used to test for differences in Hg concentrations between the two periods. Prior to model construction, normality of residuals and homoscedasticity were tested with Shapiro and Breush-Pagan tests (« lmtest » package; Zeileis and Hothorn, 2002), respectively. An alpha threshold of 0.05 was used for all statistical analyses.

3. Results

3.1. Temporal trend of Hg contamination and trophic ecology

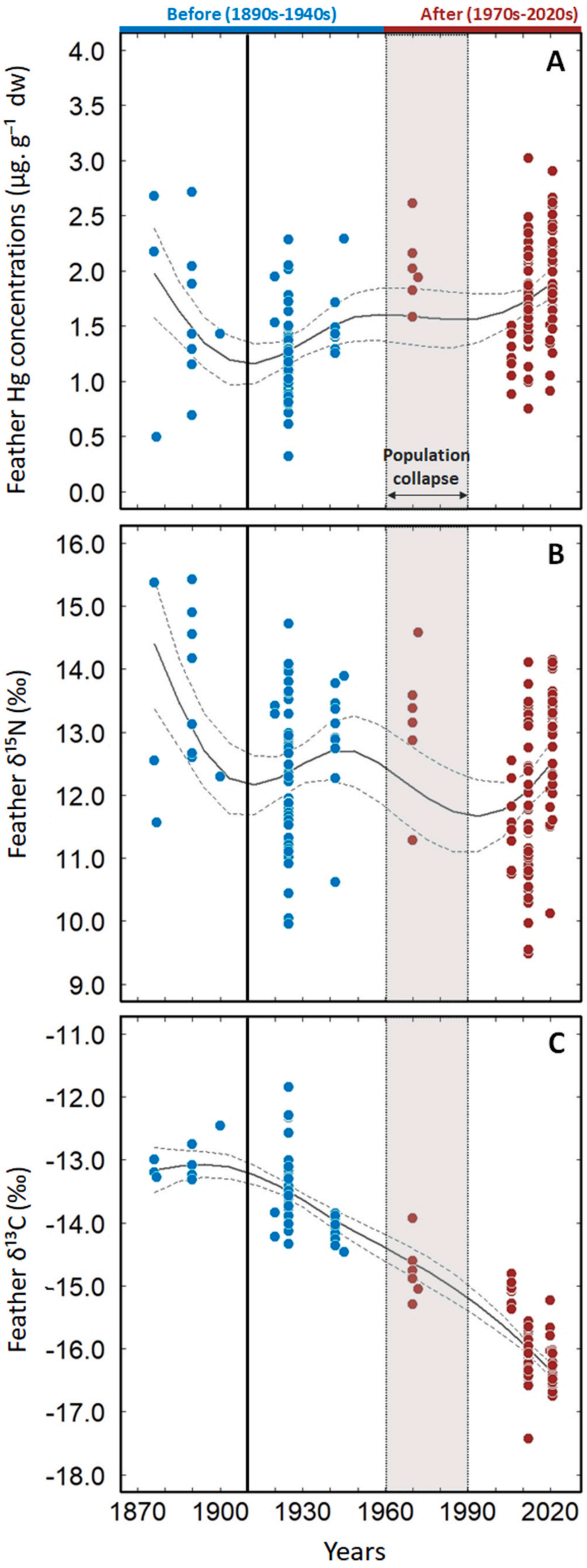
Body feathers were sampled from 220 sooty terns from seven different decades, spanning 145 years (*i.e.,* 1876–2021; Table 1), from museum skins ($n = 134$) and free-living birds ($n = 86$). Few data were available prior to the 1920s, leading to high variability and large uncertainty in temporal trends of Hg concentrations before the 1920s (Fig. 1A). Thus, interpretation of GAM temporal trends could only be carried out from the 1920s onwards. For this reason, results presented here mainly focus on the period between the 1920s and the 2020s. Detailed outputs from GAMs describing temporal variations in Hg, $\delta^{15}N$ and $\delta^{13}C$ values are provided in Table S7.

Since the 1920s, Hg concentrations increased non-linearly with time (GAM, $p < 0.0001$, deviance explained was 0.25), following three

Table 1

Mercury concentrations and stable isotope values (mean ± SD, range) in feathers of sooty terns ($n = 220$) breeding on Ascension Island (South Atlantic Ocean) from the 1870s to the 2020s, before (1840s–1940s) and after (1970s–2020s) their population collapse. Total-Hg (THg) was measured in all samples, whereas methyl-Hg (MeHg) was analysed when inorganic contamination by museum preservatives was detected (*i.e.,* prior to the 1970s). Feather $\delta^{13}C$ (corrected for the Suess Effect) and $\delta^{15}N$ values are proxies for the foraging habitat and trophic position during moult, respectively. N indicates sample size for each decade or period.

Decade	n	Feather THg ($\mu g\ g^{-1}$)	Feather MeHg ($\mu g\ g^{-1}$)	Corrected feather $\delta^{13}C$ (‰)	Feather $\delta^{15}N$ (‰)
1890	11	70.12 ± 87.34 (1.65–264.92)	1.63 ± 0.74 (0.49–2.71)	−13.11 ± 0.27 (−13.32, −12.46)	13.56 ± 1.36 (11.57–15.42)
1920	107	5.25 ± 7.82 (1.65–55.57)	1.24 ± 0.42 (0.32–3.47)	−13.50 ± 0.45 (−14.55, −11.85)	12.15 ± 1.04 (9.96–14.72)
1940	10	9.70 ± 7.51 (3.36–26.12)	1.51 ± 0.30 (1.25–2.29)	−14.10 ± 0.21 (−14.47, −13.85)	12.90 ± 0.94 (10.61–13.89)
1970	6	5.25 ± 6.50 (2.20–18.51)	2.02 ± 0.35 (1.58–2.61)	−14.76 ± 0.47 (−15.30, −13.93)	13.14 ± 1.08 (11.28–14.58)
2000	8	1.21 ± 0.20 (0.88–1.50)	–	−15.11 ± 0.19 (−15.37, −14.81)	11.56 ± 0.64 (10.74–12.55)
2010	44	1.72 ± 0.43 (0.75–3.02)	–	−16.11 ± 0.33 (−17.44, −15.56)	11.75 ± 1.19 (9.49–14.11)
2020	34	1.97 ± 0.49 (0.91–2.90)	–	−16.35 ± 0.35 (−16.75, −15.23)	12.89 ± 0.89 (10.12–14.14)
1890–1940	**128**	–	1.29 ± 0.46 (0.32–3.47)	−13.51 ± 0.47 (−14.55, −11.85)	12.33 ± 1.13 (9.96–15.42)
1970–2020	**92**	1.79 ± 0.48 (0.75–3.02)	–	−16.02 ± 0.58 (−17.44, −13.93)	12.24 ± 1.19 (9.49–14.58)



(caption on next column)

Fig. 1. Long-term trends in feather **(A)** Hg concentrations, **(B)** nitrogen (δ^{15}N) and **(C)** carbon (δ^{13}C) isotopic values of sooty terns breeding on Ascension Island (South Atlantic Ocean) between 1876 and 2021 (*i.e.*, over 145 years). The δ^{13}C values are corrected for the Suess Effect and phytoplankton fractionation (cf. see Section 2.3 for further details). The grey area represents the transition between the period before (blue) and after (red) population collapse that occurred between the 1960s and the 1990s (see Reynolds et al., 2019 for further details). Solid and dotted curves represent the best-fitting model detected from construction of GAMs and the confidence interval, respectively. To the right of the vertical solid line at 1910 is the period of realistic interpretation (see **Material and methods** and **Discussion** for further details). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

distinct phases. First, Hg concentrations increased by 62.9% between the 1920s and the 1970s. Then, Hg concentrations exhibited an unclear pattern of change between the 1970s and the 2000s with high variability and large confidence intervals. Finally, Hg concentrations increased again by 62.8% from the 2000s to the present day. Overall, Hg concentrations were 58.9% higher in the 2020s than in the 1920s.

The δ^{15}N values ranged from 9.49 ‰ in the 2010s to 15.42 ‰ in the 1890s (Fig. 1B), with values varying non-linearly over time (GAM, $p < 0.0001$, deviance explained was 0.13), following three distinct phases. Values of δ^{15}N increased by 5.8% between 1920s and the 1940s (*i.e.*, +0.75 ‰), decreased by 10.4% thereafter until the 2000s (*i.e.*, −1.34 ‰) and increased by 11.5% from 2000s (i.e., +1.33 ‰) to the present day (Table 1 and S6).

Corrected δ^{13}C isotopic values ranged between −17.4 ‰ in the 2010s and −11.9 ‰ in the 1920s (Fig. 1C), with values decreasing dramatically over time (GAM, $p < 0.0001$, deviance explained was 0.91). There was a quasi-linear 17.4% change over the century between the 1920s and the 2020s (*i.e.*, +2.85 ‰).

Feather Hg concentrations were 38.8% higher after (1.79 ± 0.48 µg g^{-1} dw) than before (1.29 ± 0.46 µg g^{-1} dw) (ANOVA; $F_{1,218} = 59.2$, $p < 0.001$) the population collapse that occurred between the 1960s and the 1990s (Fig. 2A). Isotopic niches were statistically different before and after the population collapse (MANOVA; $F_{1,218} = 649.8$, $p < 0.0001$; Fig. 2B), with no significant difference in δ^{15}N values (−0.7%; $p = 0.39$) and ~2.5 ‰ lower δ^{13}C values after than before the population collapse (−18.6%; ANOVA; $F_{1,218} = 1271.8$, $p < 0.001$).

3.2. *Drivers of Hg contamination over time*

Results from model selection are provided in Table 2. The best model to explain time-detrended Hg concentrations included both time-detrended δ^{13}C and δ^{15}N values (linear models, $R^2 = 0.14$, $p < 0.0001$). Specifically, Hg concentrations decreased with increasing δ^{13}C values (β ± SE: −0.279 ± 0.082, 95% CI: −0.442, −0.117) and increased with increasing δ^{15}N values (β ± SE: 0.090 ± 0.030, 95% CI: 0.031, 0.149) (Fig. 3).

4. Discussion

In this study, sooty terns act as bioindicators of temporal trends in Hg contamination of food webs in the tropical South Atlantic Ocean. We have built on previous long-term investigations of the trophic ecology of breeding sooty terns on Ascension Island (1890s–2010s; Reynolds et al., 2019), by extending the recent time series by an additional decade and providing feather-derived Hg concentrations. To the best of our knowledge, this is the first study to determine Hg contamination over such an extensive multidecadal timescale in the tropical oceans. Our results revealed a global non-linear increase in Hg contamination of sooty terns over the last 145 years, because of both trophic and environmental changes over time.

Museum specimens represent invaluable archives that enable both dietary and ecotoxicological investigations on multidecadal timescales.

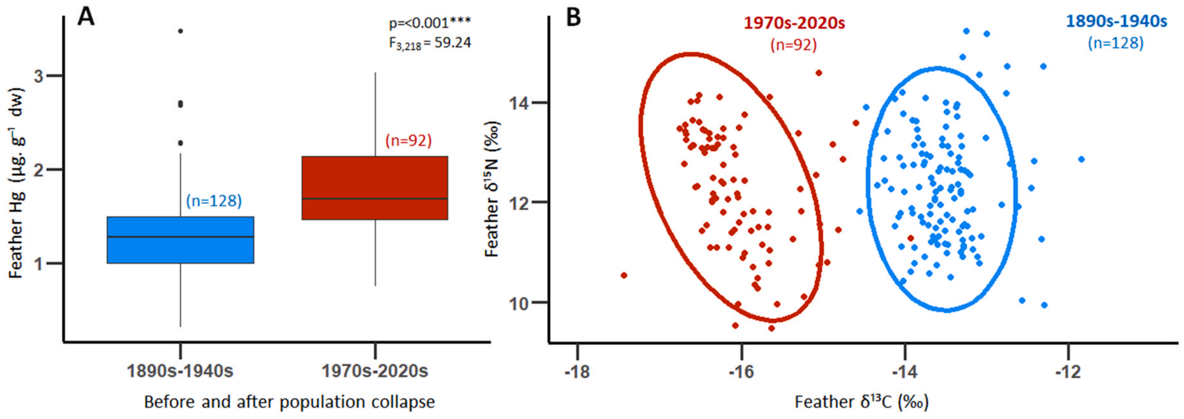


Fig. 2. Difference in **(A)** Hg concentrations and **(B)** isotopic niches (δ^{13}C and δ^{15}N values) from feathers of adult sooty terns relative to the population collapse on Ascension Island between the 1960s and the 1990s. Points, ellipses (including 90% of the data) and bars in blue and red are before (n = 128) and after (n = 92) the population collapse, respectively. n indicates the sample sizes. Boxplots indicate median values (midlines), errors bars (whiskers) and outliers (black dots). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

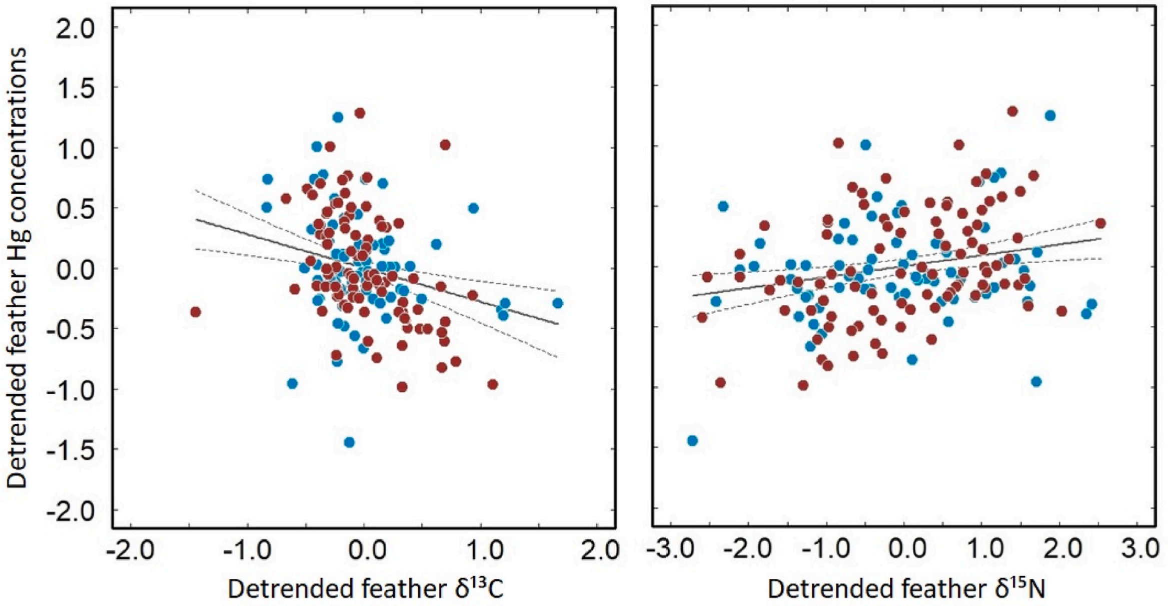


Fig. 3. Relationship between time-detrended Hg concentrations and time-detrended stable isotope values in feathers of adult sooty terns breeding on Ascension Island in the South Atlantic Ocean between 1876 and 2021 (*i.e.,* over 145 years). The tern population collapsed between the 1960s and the 1990s (see Reynolds et al., 2019 for further details), with data before- (in blue) and after (in red) the collapse. All data were scaled. Solid and dashed lines represent the best-fitting linear model and the 95% confidence interval, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

However, any retrospective investigation at a single location is challenging because it is highly dependent on sampling effort each year over a large time scale. Gaps in retrospective time-series are thus very common and consequently, analytical statistical approaches that were non-linear in nature were most relevant. Because the oldest specimens in our study were also the least numerous, temporal trends were characterized by high uncertainty between the 1890s and the 1920s (Fig. 1A). For this reason, interpretations of our findings mainly focussed on the 1920s to the 2020s.

4.1. Long-term trend in foraging ecology: an update

Using feathers as a record of sooty tern foraging ecology on the island, Reynolds et al. (2019) found a significant decrease in feather δ^{15}N values, suggesting a change in their trophic position over time. By extending the time series in our study with the inclusion of feathers obtained in 2020 and 2021, we did not detect any further significant linear trend in δ^{15}N values. Instead, we detected a non-linear trend (Fig. 1B): initially, there was an increase between the 1900s and the 1940s, followed by a decrease between the 1940s and the 2000s, and a second increase from the 2000s onwards (Table 1 and S6). The addition of the most recent decade in the time series clearly highlights the importance of such long-term monitoring, by revealing recent and unpredictable patterns in the foraging ecology of sooty terns. These long-term variations in feather δ^{15}N values could reflect variations in seabird diet over time (Reynolds et al., 2019), but also changes in the isotopic baseline (McMahon et al., 2013). Isoscapes of δ^{15}N can vary seasonally due to changes in primary productivity which stem from variation and cycling of nutrient sources, as well as changes in species composition and growth rates of phytoplankton (Cifuentes et al., 1988; Goering et al., 1990; Ostrom et al., 1997; Vizzini and Mazzola, 2003). The changing δ^{15}N isoscape could then have cascading effects on feather isotopic values of seabird consumers at upper trophic levels and hence, drive the temporal variations in their trophic position. One way to disentangle these hypotheses would be to perform compound-specific stable isotope analysis of amino acids (CSIA-AA) (McMahon and Newsome, 2019), which would help distinguish the specific signatures of the baseline («source» δ^{15}N-AAs) and the diet-consumer transfer («trophic» δ^{15}N-AAs), and hence elucidate the «true » trophic position of sooty terns over time.

Extending the results of Reynolds et al. (2019), corrected δ^{13}C values exhibited a similar and even stronger decreasing trend to the present day (Fig. 1C), with a 2.85 ‰ decline between the 1920s and the 2020s. This represents a 3.24 ‰ decline when considering the trend between the 1890s and the 2020s. A comparable decrease in δ^{13}C values over time was previously described in thin-billed prions (*Pachyptila belcheri*) in the Southern Ocean (Cherel et al., 2014; Quillfeldt et al., 2010), and was interpreted as a result of latitudinal change in their moulting area, switching from sub-Antarctic to Antarctic waters over more than 90 years (1913–2005). Similarly, moulting areas of Ascension sooty terns could have changed spatially over time but, to date, their post-breeding migrations have only been studied using geolocators between 2011 and 2015 (Reynolds et al., 2021) and far more research is required. Continuing such tracking work over the next few decades would allow trophic ecology of sooty terns to be directly related to long-term changes in their spatial distribution in the South Atlantic Ocean. Alternatively and similar to the hypothesis generated above for changes in feather δ^{15}N values, changes in δ^{13}C values might reflect baseline changes in the study area. Baseline δ^{13}C values vary both spatially (latitudinal gradient influenced by temperature; Sackett et al., 1965) and temporally (seasonal variability) in the marine environment (McMahon et al., 2013). The temporal decline in δ^{13}C values of sooty terns could thus reflect fluctuations in baseline δ^{13}C values. However, the observed trend appears too marked to be explained solely by a change in δ^{13}C baseline values. Carbon isotope values reflect phytoplankton productivity in aquatic environments (DeNiro and Epstein, 1978; France, 1995; Hobson, 1999). Declines in δ^{13}C values have been associated with those in primary productivity, and hence with the carrying capacity of ecosystems (Hirons et al., 2001; O'Reilly et al., 2003; Schell, 2000), and ultimately with declines of penguin (Spheniscidae) populations, for example, in the sub-Antarctic region (Hilton et al., 2006; Jaeger and Cherel, 2011). In the South Atlantic Ocean, a 1.38 ‰ decline in δ^{13}C values was observed in muscle of tuna species between 2000 and 2015, suggesting a global shift in phytoplankton community structure (Lorrain et al., 2020). This coincides with the 1.24 ‰ decline we observed in feather δ^{13}C values of Ascension sooty terns between the 2000s and the 2020s in the same region. So, the global 2.85 ‰ decline in feather δ^{13}C values in our study (between the 1920s and the 2020s) could likely result from a change in phytoplankton productivity in the surrounding tropical marine ecosystem.

4.2. Long-term trend in Hg contamination and its drivers

Overall, our results (Table 1) are consistent with the range of average Hg concentrations observed for sooty terns elsewhere, such as in the Mozambique Channel (0.2 μg g^{-1}; Jaquemet et al., 2008; Kojadinovic et al., 2007), Pacific Islands (0.8 μg g^{-1}; Burger et al., 1992), the Seychelles (1.2 μg g^{-1}; Ramos and Tavares, 2010) and Puerto Rico (2.6 μg g^{-1}; Burger and Gochfeld, 1991). However, Ascension sooty terns exhibit Hg concentrations in the upper part of this range (1.5 μg g^{-1}). Among tropical seabirds, feather Hg values were intermediate compared with other sternids at other locations, such as brown noddies (*Anous stolidus*) in Hawaii (0.6 μg g^{-1}; Burger et al., 2001) and bridled terns (*Onychoprion anaethetus*) (1.4 μg g^{-1}) and roseate terns (*Sterna dougallii*) in Puerto Rico (2.3 μg g^{-1}; Burger and Gochfeld, 1991). Low Hg contamination in sooty terns is consistent with their foraging ecology that largely depends on small epipelagic fish and invertebrates from surface waters (Ashmole, 1963a). Epipelagic prey exhibit lower Hg concentrations than mesopelagic and benthic prey (Chouvelon et al., 2012; Monteiro and Furness, 1997; Ochoa-Acuña et al., 2002), mainly because Hg methylation occurs in mesopelagic waters (Blum et al., 2013; Bowman et al., 2020; Cossa et al., 2009; Wang et al., 2018).

Few studies (n = 14) have investigated long-term Hg trends in seabirds by using feathers as archives (Table S1). Most have focussed on temperate (*e.g.,* Appelquist et al., 1984; Thompson et al., 1993, 1992) and polar regions (*e.g.,* Bond et al., 2015; Carravieri et al., 2016; Scheifler et al., 2005). Only two have focussed on subtropical regions, in the Azores (Monteiro and Furness, 1997) and islands of the Pacific (Vo et al., 2011). Monteiro and Furness (1997) measured a 1.1% increase in Hg contamination *per* year between the 1880s and the 1990s in epipelagic Cory's shearwaters (*Calonectris borealis*) that share similar habitat with sooty terns, albeit in the North Atlantic Ocean. This was a stronger annual increase than that of 0.37% that we found if considering simply a linear trend. Despite different temporal coverage between studies, it appears that Hg transfer to predators has increased more slowly in the South than in the North Atlantic Ocean. Similar spatial disparities in Hg contamination were observed in tunas (Médieu et al., 2022), probably as a result of spatial differences in seawater Hg concentrations at the base of the food web, and inorganic Hg deposition among distinct oceanic regions. Mercury deposition could be higher in the subtropical than in the tropical Atlantic Ocean for two reasons. First, anthropogenic Hg was historically mainly emitted by industrialized countries from the Northern Hemisphere (Pacyna et al., 2006). Secondly, Hg is globally distributed by atmospheric circulation (Selin, 2009), which includes successive atmospheric circulation cells from the pole to the tropics, namely the Polar, the Ferrel and the Hadley Cells. As such, Hg emitted in Europe, for example, could be more easily deposited in the subtropical North Atlantic Ocean (*i.e.*, one circulation cell away) than in the tropical South Atlantic Ocean (*i.e.*, two circulation cells away).

Despite the temporal mismatch between feather Hg and stable isotopes (Bond, 2010), the trophic ecology of sooty terns influenced significantly their feather Hg contamination over time. Indeed, the best model explaining feather Hg contamination included both δ^{13}C and δ^{15}N values (Table 2), indicating that the temporal increase in feather Hg concentrations was associated with the strong decrease in δ^{13}C values and fluctuating δ^{15}N values observed over the past century (see **Discussion** above). However, the best model including both isotopic values explained only 14% of the total variation in Hg contamination, which suggests that other unknown factors (discussed below) also influenced long-term Hg contamination in sooty terns from Ascension Island. This is consistent with previous studies on other species (Bond et al., 2015; Carravieri et al., 2016; Vo et al., 2011). In addition, isotopic variations at the century scale are not simply or solely driven by changes in feeding habitat and trophic position, but likely by changes at the base of the food web, which are driven by several environmental and ecological processes (see Section 4.1). Therefore, our results suggest that the temporal trend in Hg transfer to predators in the South Atlantic Ocean was driven by trophic factors linked to ecosystem-wide changes, and other unknown factors. For instance, the increase in feather Hg concentrations observed from the 1920s to the 1970s was likely facilitated by growing Hg emissions during the Industrial Era with developing human activities, such as the Gold Rush, the burning of coal and intensifying manufacturing efforts to meet the military demands of World War II (Thompson et al., 1993). Between the 1970s and the 2000s, Hg concentrations appeared to stabilize. However, this phase included a very low number of available samples *per* year with high variability, implying large uncertainty in the generated models. Thus, whether Hg concentrations really remained stable between the 1970s and the 2000s is difficult to confirm. Mercury concentrations increased again from the 2000s to the present day, despite strong regulations restricting Hg emissions since the 1990s in the Northern Hemisphere and 2017 globally (Minamata Convention on Mercury). This, together with other recently detected increases (*e.g.,* Bond et al., 2015; Carravieri et al., 2016; Tartu et al., 2022), raises concerns for potential consequences on wildlife health.

4.3. Did Hg drive sooty tern population collapse on Ascension Island?

On Ascension Island, sooty terns suffered a 80% population collapse between the 1960s and the 1990s (Hughes et al., 2017). Reynolds et al. (2019) attributed this in part to a dietary shift from predominantly fish in the 1890s to the 1940s prior to the decline, to predominantly squid in the 1970s to the 2010s after it. With additional feather samples from the 2020s, our results revealed that isotopic niches were different between the two periods, with a significant difference in foraging habitat ($\delta^{13}C$) (agreeing with $\delta^{13}C$ values documented in Reynolds et al., 2019) but with an equivalent trophic position ($\delta^{15}N$) (Fig. 2B).

Mercury concentrations were 38.8% higher post-compared to pre-population collapse (Fig. 2A). These results are consistent with previous temporal comparisons, in which Hg concentrations have increased between historical and contemporary time periods in marine environments worldwide (Carravieri et al., 2016; Furness et al., 1995; Vo et al., 2011). During the period of population collapse, all Hg concentrations were relatively high with low variability, likely reflecting a generally high contamination in sooty terns. In comparison, there was more variability before and after the period of population collapse, probably because of slight differences in either diet or the proportions of different prey species constituting the diet. Given measured levels of Hg in sooty tern feathers – although elevated after the population collapse – it seems unlikely that Hg contamination drove the population decline of sooty terns on Ascension Island. Indeed, the extent of Hg contamination detected was below severe toxicity thresholds for seabird feathers (4–10 μg g^{-1}; Ackerman et al., 2016; Chastel et al., 2022). Investigating contamination by other pollutants (such as pesticides or plastics) would undoubtedly allow further insights into the risks and demographic stressors for this population, but our results reinforce the conclusions of Reynolds et al. (2019), that overharvesting of large predatory fish and climate change likely negatively impacted sooty tern trophic ecology, thereby resulting in elevated Hg contamination.

4.4. Influence of climate change and fisheries on tropical Hg contamination

Reynolds et al. (2019) highlighted the importance of increasing sea surface temperatures (SST) and tuna extraction by fisheries in explaining long-term changes in the trophic ecology of sooty terns breeding on Ascension Island. Sooty terns are heavily reliant on large schools of surface-swimming tunas, such as yellowfin (*Thunnus albacares*) and skipjack (*Katsuwonus pelamis*) tuna, to drive their common prey to the ocean surface, where many tropical seabirds forage in so-called « facilitated foraging » (Ashmole, 1963a; Au and Pitman, 1986; Ballance and Pitman, 1999; Maxwell and Morgan, 2013). Sooty terns are therefore susceptible to any over-exploitation and major declines in these associated species (Cullis-Suzuki and Pauly, 2010; Juan-Jordá et al., 2011). Nonetheless, under climate change, and specifically with changes in temperatures and salinity, global abundances of tunas are predicted to increase in tropical waters (Erauskin-Extramiana et al., 2019). Specifically, Ascension Island is predicted to become more suitable for several tuna species, including sooty tern associates (Townhill et al., 2021). Climate change and fishing pressure are therefore likely to impact Hg contamination in sooty terns, and hence in tropical marine ecosystems, even though the direction and the extent of these impacts are difficult to predict precisely.

Since seabird Hg contamination is heavily associated with their foraging ecology, climate change and fishing pressure also have indirect effects on Hg contamination through dietary changes (prey switching), species' interactions and transfer through marine food webs (biotic factors). For example, MeHg concentrations increased in marine fish as a result of dietary shifts initiated by overfishing and with increasing SST (Schartup et al., 2019). Other environmental factors can also influence seabird Hg contamination, such as Hg transport, deposition, uptake and methylation rates in the ocean (Krabbenhoft and Sunderland, 2013). Mercury is mainly deposited in oceans by atmospheric fallouts (Eagles-Smith et al., 2018). Although Ascension Island is isolated, with its nearest neighbour, St Helena, located more than 1300 km away, it is highly unlikely that Hg contamination results from local anthropogenic sources. As a volcanic island, Hg contamination might alternatively result from local geological activities, but there is no record of volcanic activity of Ascension Island over the last 500–1000 years (Preece et al., 2018). Instead, Hg contamination in sooty terns likely results from global Hg transport. In the ocean, sources of MeHg are multiple: methylation can occur in the sediments of continental shelves (Hammerschmidt and Fitzgerald, 2006) and estuaries (Heyes et al., 2006), within the water column (Cossa, 2013; Hammerschmidt and Bowman, 2012) and at deep-ocean hydrothermal vents (Bowman et al., 2016, 2015). Mercury methylated through these processes can then be widely distributed by ocean currents, both spatially (global ocean circulation) and vertically (vertical mixing and upwellings) (Mason et al., 2012). Climate change will likely influence all of these processes and could ultimately influence sooty tern contamination on different temporal and spatial scales. Methylation rate and bioavailability of Hg are also affected by other processes such as acidification, eutrophication and/or deoxygenation (Clayden et al., 2013; Gong et al., 2021; Jardine et al., 2013; Zhang et al., 2021). For instance, oxygen-depleted zones (*i.e.*, «dead zones») are increasing with climate change (Breitburg et al., 2018; Watson, 2016), both in number and size. Mercury methylation is substantially enhanced in low-oxygen zones in the ocean, that at low latitudes (*i.e.,* tropics) are the most vulnerable to further deoxygenation (Gruber, 2011). Tropical oceans should thus be a priority for further studies of Hg contamination, especially in the context of climate change. Further research should focus on Hg stable isotopes to disentangle the different environmental and ecological factors involved, by identifying Hg sources and exploring the associated biogeochemical and trophic processes in the different compartments of tropical marine ecosystems (Renedo et al., 2020).

Mercury contamination of marine environments results from long-term processes, influenced by Hg transport, methylation and bioavailability, as well as from ecological interactions, both globally and locally. Undoubtedly, there is a time lag of years, decades, or even centuries between Hg emissions and deposition in the ocean and its resulting contamination of marine food webs (Driscoll et al., 2013; Foster et al., 2019; Sunderland and Mason, 2007, UN Environment, 2019). Long-term monitoring of Hg is therefore crucial to: (i) assess the ongoing health status of tropical marine ecosystems and its variability over time, and (ii) ultimately minimize the risks of Hg exposure to wildlife and human populations alike, the latter relying heavily on marine food resources worldwide.

Credit author statement

Fanny Cusset: Conceptualization, Methodology, Software, Formal analyses, Investigation, Data curation, Visualization, Writing – Original draft, review and editing, Project administration; **S. James Reynolds:** Conceptualization, Resources, Writing – Reviewing and editing; **Alice Carravieri:** Formal analyses, Investigation, Visualization, Writing – Reviewing and editing; **David Amouroux:** Writing – Reviewing and editing; **Océane Asensio:** Investigation, Writing – Reviewing and editing; **Roger C. Dickey:** Resources, Writing – Reviewing and editing; **Jérôme Fort:** Supervision, Investigation, Validation, Writing – Reviewing and editing; **B. John Hughes:** Resources, Writing – Reviewing and editing; **Vitor H. Paiva:** Conceptualization, Resources, Writing – Reviewing and editing; **Jaime A. Ramos:** Conceptualization, Resources, Writing – Reviewing and editing; **Laura Shearer:** Resources, Writing – Reviewing and editing; **Emmanuel Tessier:** Investigation, Writing – Reviewing and editing; **Colin P. Wearn:** Resources, Writing – Reviewing and editing; **Yves Cherel:** Supervision, Investigation,

Validation, Writing – Reviewing and editing; **Paco Bustamante:** Conceptualization, Supervision, Investigation, Validation, Funding acquisition, Writing – Reviewing and editing.

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.

Data availability

Data will be made available on request.

Acknowledgments

First, we thank all people involved in the original study of Reynolds et al. (2019), for bird sampling, both in museums and in the field. Funding for sample collections was provided by SJR. We are thankful to the Ascension Island Government for granting research permits to sample live birds in the 2000s, the 2010s and the 2020s. The authors are grateful to Carine Churlaud and Maud Brault-Favrou from the Analyses Elémentaires platform (LIENSs) for their support during Hg analysis and to Gaël Guillou from the Analyses Isotopiques platform (LIENSs) for running the stable isotope analyses. Special thanks are due to Sabrina Tartu and Céline Albert for statistical guidance during data exploration. We are grateful to Lucie Machin for the sooty tern drawing on the graphical abstract. Thanks are due to the CPER (Contrat de Projet Etat-Région) and the FEDER (Fonds Européen de Développement Régional) for funding the AMA and IRMS of LIENSs laboratory. The Institut Universitaire de France (IUF) is acknowledged for its support to PB as a Senior Member. This work benefitted from the French GDR "Aquatic Ecotoxicology" framework, that aims to foster stimulating scientific discussions and collaborations for more integrative approaches.

Appendix A. Supplementary data

Supplementary data to accompany this article can be found online at https://doi.org/10.1016/j.envpol.2023.121187.

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