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Sex- and age-specific mercury accumulation in a long-lived seabird

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HIGHLIGHTS

- Little is known about within-individual accumulation of mercury (Hg) in birds.
- Studying within-individual processes requires longitudinal data, which are rare.
- We collected 1209 blood samples from 588 common terns across 7 years.
- Hg levels exceeded the admitted low to moderate toxicity threshold.
- Males had higher Hg levels, and accumulated Hg with age at faster rates, than females.

GRAPHICAL ABSTRACT
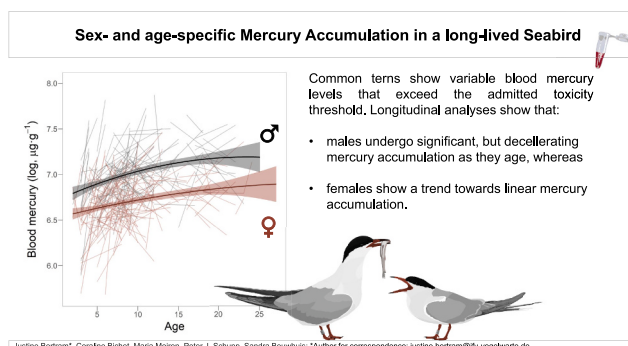
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ABSTRACT

Mercury levels in the environment are increasing, such that they are also expected to accumulate in top-predators, but individual-based longitudinal studies required to investigate this are rare. Between 2017 and 2023, we therefore collected 1314 blood samples from 588 individual common terns (*Sterna hirundo*) to examine how total blood mercury concentration changed with age, and whether this differed between the sexes. Blood mercury concentrations were highly variable, but all exceeded toxicity thresholds above which adverse health effects were previously observed. A global model showed blood mercury to be higher in older birds of both sexes. Subsequent models partitioning the age effect into within- and among-individual components revealed a linear within-individual accumulation with age in females, and a decelerating within-individual accumulation with age in males. Time spent at the (particularly contaminated) breeding grounds prior to sampling, as well as egg laying in females, were also found to affect mercury concentrations. As such, our study provides evidence that male and female common terns differentially accumulate mercury in their blood as they grow older and calls for further studies of the underlying mechanisms as well as its consequences for fitness components, such as reproductive performance and survival.

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1. Introduction

Industrial activities and other drivers of global change, such as ongoing climate warming, increase environmental mercury levels (Fitzgerald et al., 2007; Eagles-Smith et al., 2018; UN Environment, 2019). As mercury enters aquatic ecosystems, microorganisms transform it into its organic and bioavailable form, methylmercury (MeHg). When biota ingest methylmercury, it can accumulate along the food chain (Atwell et al., 1998), exposing long-lived top predators, such as some seabird species, to particularly high levels. As such, seabirds have sometimes been used as bioindicators to monitor environmental mercury contamination and to study its consequences for physiology, behaviour and fitness components (Burger and Gochfeld, 2004). Blood is a tissue regularly used for this purpose, as it enables the measurement of an individual bird's mercury contamination without sacrificing it (Kahle and Becker, 1999). It additionally provides an opportunity for repeated sampling of the same individuals.

Repeated blood sampling of seabirds is most feasible during the breeding season, when individuals are usually highly site faithful and can be caught and/or sampled during nest attendance on land. Blood mercury levels during the breeding season largely reflect a combination of recent mercury uptake, dependent on trophic position and contamination of prey items in the main foraging areas (Monteiro and Furness, 1995), and levels of deposition in internal organs (Eagles-Smith et al., 2008; Fromant et al., 2016). For migratory seabirds, however, there may also be carry-over effects from mercury uptake in the wintering areas or during migration (Lavoie et al., 2014; Quillfeldt et al., 2022; Carravieri et al., 2023), the influence of which could depend on the timing of sampling relative to the timing of these other events along the annual cycle (Eagles-Smith et al., 2009a).

Besides by timing, local uptake and deposition in internal tissues, blood mercury contamination may be affected by sex. Female birds have often been found to exhibit lower mercury levels than males, potentially suggesting a role for egg laying in the ability of females to excrete part of their mercury (Bond and Diamond, 2009; Ackerman et al., 2020; but see Robinson et al., 2012; Hitchcock et al., 2019). To test whether egg laying could explain (part of) an observed sex difference, one would ideally sample birds of both sexes both prior to, and after, female egg laying, or artificially manipulate the number of eggs females produce, for example by conducting an egg removal experiment, inducing females to produce additional eggs. Alternatively, however, one could test whether variation in clutch size explains variation in post-laying levels of mercury contamination in both sexes, whereby one would expect a negative correlation in females only in case of eggs being a pathway for detoxification.

Despite detoxification, potentially via eggs in females (Bond and Diamond, 2009; Ackerman et al., 2020), and via feathers in birds of both sexes (>90 %; Braune and Gaskin, 1987a; Whitney and Cristol, 2017), if incomplete, the relative longevity of seabirds might predispose them to bioaccumulation of mercury as they age (Rowe, 2008; Celik et al., 2021). Studies examining the age-specificity of mercury contamination in seabirds have, however, not yet yielded conclusive evidence for this to occur. Insightful first studies reported the expected positive correlation between age and mercury concentration when comparing birds of different age classes, i.e. chicks vs. adults (Souza et al., 2020; Bighetti et al., 2021). Given that chicks and adults do not only differ in age, but may also differ in diet - with chicks feeding on smaller, potentially less contaminated prey items - and moult pattern - given that chicks are in the process of growing their feathers, potentially depositing mercury - these results, however, do not provide conclusive proof that mercury accumulates as birds grow older. Indeed, among adult seabirds, further studies found either no (Thompson et al., 1993; Becker et al., 2002; Carravieri et al., 2014, 2022; Tartu et al., 2015) or a negative correlation (Agusa et al., 2005; Tavares et al., 2013; Tartu et al., 2014) between age and mercury concentration. Interpretation of the differences in the perhaps surprising results among these latter studies is currently

hampered by the fact that all were based on cross-sectional data, which do not facilitate distinguishing between within-individual accumulation of mercury with age and among-individual variation in survival in relation to mercury pollution levels (selective disappearance, sensu van de Pol and Wright, 2009). Mercury levels could, for example, be positively associated with survival due to better foragers both acquiring more resources to aid survival (as a function of individual quality), but also becoming more contaminated via increased dietary mercury uptake (Binkowski et al., 2021). Alternatively, mercury levels could be negatively associated with survival (Ma et al., 2018a) due to detrimental effects on physiology (Hawley et al., 2009; Lewis et al., 2013; Ma et al., 2018a, 2018b; Adams et al., 2020; Grunst et al., 2023a). Such selective disappearance would result in patterns of age-specific mercury contamination at the population level (here: an in- or decrease, respectively) that could either inflate or mask within-individual patterns of accumulation. When data have been collected longitudinally, methods are available to assess the relative importance of within- and among-individual sources of age-related patterns of mercury contamination (van de Pol and Wright, 2009). These methods have been routinely applied in studies of ageing in birds (see Bouwhuis and Vedder, 2017 for a review), and could be useful when adopted in studies of avian physiology as well.

In this study, we assess factors underlying variation in blood mercury concentrations in a long-lived migratory seabird, the common tern (*Sterna hirundo*). Using 1209 blood samples obtained from 588 common terns of known age and sex, and collected across seven years, we investigate whether variation in total mercury levels (THg) can be explained by the birds' (i) timing, assessed as the intervals between arrival at the breeding colony, egg laying and blood sampling, (ii) sex, (iii) clutch size and (iv) age. Importantly, after first running a global linear mixed effect model assessing the effect of age, we adopt a longitudinal analysis approach and run a second linear mixed effect model to test whether age-specificity of mercury concentration is best explained by within-individual processes and/or selective (dis)appearance of birds in relation to, or based on, their mercury levels. Third, we add a within-season perspective to our analyses, enabled by the fact that 88 birds were sampled repeatedly either within a single breeding attempt or among two breeding attempts when birds re-nested after having failed.

2. Methods

2.1. Study species and site

Common terns are long-lived seabirds, mainly feeding on fish of a length up to 15 cm (Becker and Ludwigs, 2004). After migration to their wintering areas along the African coast (Kürten et al., 2022), they return to their breeding colony each spring (Dittmann and Becker, 2003; Dittmann et al., 2005). Starting their first breeding attempt on average at an age of three years, common terns usually produce clutches of one to three eggs, which are incubated by both pair members (Becker and Ludwigs, 2004). Their colonial breeding, longevity and high level of site-fidelity (Szostek and Becker, 2012) make common terns particularly suitable for extensive longitudinal studies.

We studied the mercury concentration in blood of common terns breeding at a monospecific colony located at the Banter See in Wilhelmshaven, at the German North Sea coast (53°30′40″ N, 08°06′20″ E). Since 1992, this colony is monitored three times weekly to, among others, record laying date and clutch size for each nest, and all locally hatched birds are marked with rings, as well as subcutaneously implanted transponders if they reach fledging age (Becker and Wendeln, 1997). The colony site consists of six artificial islands (10.7 × 4.6 m), covered with gravel and surrounded by 60 cm high walls. The walls sustain 44 resting platforms equipped with antennas that allow the automatic and remote detection of arrival date (Moiron et al., 2020) and presence of transponder-marked birds. Additionally, placing mobile antennas around each nest for at least 24 h allows the identification of

breeders and linking them to their nests. The sex of the studied birds is determined molecularly following Becker and Wink (2003).

2.2. Blood collection

Between May and August 2017–2023, we used the antenna system to identify 588 transponder-marked common terns during incubation to collect 1209 blood samples 3–36 days after the first egg of their clutch was laid. We collected 47 further blood samples from 45 birds within the same breeding attempt within 1–24 days after their first blood collection in 2021 and 2023. Despite replacement clutches being infrequent (approximately 9 %), they significantly contribute to the lifetime reproductive success of common terns (Becker and Zhang, 2011) and we made use of their occurrence to additionally collect 58 blood samples from 55 individuals within 19–55 days after their first blood collection in 2017, 2021, 2022 and 2023 (Table 1). In all cases, blood sampling was conducted prior to hatching of the first chick. In total, we collected 1314 blood samples from 588 individuals (Table 1).

For blood sampling, we used starved kissing bugs (*Dipetalogaster maxima*; Becker et al., 2006). We inserted one or two third larval instars into an artificial hollow egg, closely resembling a real common tern egg in size and colour. After the target bird was detected on its nest by the automatic antenna system, we replaced one of its real eggs with the "bug-egg", storing the real egg in a foam-lined box kept warm in an incubator at 37 °C. Birds resumed incubation shortly after nest manipulation. Small holes along the artificial egg allowed the bug(s) to suck blood from the incubating bird. Throughout this process, birds were meticulously observed from observation huts within the colony to prevent sample contamination by switches between partners. After 20–30 min, we visually confirmed whether the bug had successfully obtained a blood meal; if so, we retrieved the artificial egg and returned the real egg. Up to 300 µl blood of the bird was then extracted from the bug's abdomen using a syringe and stored at −18 °C until analysis. To ensure that mercury concentrations in blood sampled this way are not affected by bug physiology, in 2017, we additionally blood sampled 18 birds (7 males and 11 females) from the brachial vein using needles and capillaries. A correlation test, performed in R, showed a highly significant correlation of 0.93 (Fig. S1).

2.3. Mercury analysis

Mercury analysis was performed using a Direct Mercury Analyzer (DMA-80, MLS Mikrowellen-Labor-Systeme GmbH, Germany) to quantify total mercury concentration, which we report on a wet-weight (ww) basis, and which provides a reliable proxy for MeHg levels (Renedo et al., 2021). For each sample, ca. 20 µl of wet whole blood was pipetted into a nickel sample boat and weighted, then dried and thermally decomposed in a stream of oxygen at 750 °C. This process released gaseous mercury that was trapped by gold amalgamation to prevent its combustion along with the remaining compounds of the blood sample.

Table 1
Details regarding the 1314 blood samples collected from 588 incubating common terns in the years 2017–2023 (total (males; females)).

Year	No of 1st blood samples taken in the first clutch	No of 2nd blood samples taken within the same clutch	No of 2nd blood samples taken from a replacement clutch
2017	157 (78; 79)	–	5 (2; 3)
2018	166 (78; 88)	–	–
2019	216 (106; 110)	–	–
2020	107 (51; 56)	–	–
2021	152 (73; 79)	26 (13; 13)	30 (14; 16)
2022	184 (97; 87)	–	11 (7; 4)
2023	227 (114; 113)	21 (8; 13)	12 (7; 5)
total	1209 (597; 612)	47 (21; 26)	58 (30; 28)

By subsequent heating to 850 °C, the mercury was released and determined by atomic absorption spectrometry at 253.7 nm. Each of our 35 measurement batches included an analytical blank to verify the absence of contamination of the device, and for 29 out of the 35 batches we have the values obtained for a certified reference material (DORM-4; fish protein from National Research Council, Canada, 412 µg·kg^{-1} dry weight; with a mean percent recovery ± standard deviation of 109 ± 7 %) to ensure its accuracy as well. The detection limit of THg was 0.0003 µg·g^{-1} and none of the samples showed a value below this limit.

As part of the laboratory's quality assurance and quality control (QA/QC) measures, it engages in the QUASIMEME international laboratory performance study program twice a year with satisfactory results. Nevertheless, to evaluate the reliability of our measurement method, we split 77 sufficiently large blood samples to obtain duplicates, which were measured blindly and across batches. The relative difference among duplicates was 10 ± 11 % and a correlation test, using the "cor.test" function in R, showed a highly significant correlation of 0.88 (Fig. 1), such that all further samples were measured only once.

2.4. Statistical analyses

To assess potential sources of variation in the blood THg levels of common terns, we first used a linear mixed model with 'blood THg concentration' (in µg·kg^{-1} ww) as the dependent variable and 'year' and 'individual identity' as random effects to account for the non-independence of repeated observations on the same individual and potential environmental differences across years. We log$_e$-transformed our dependent variable to meet model assumptions and modelled it assuming a Gaussian error distribution (in this model and hereafter). We included 'age' as a linear and a quadratic covariate, and 'sex' as 2-level and 'clutch size' as 3-level class variables, to not limit ourselves to assuming linear effects. To account for the fact that common terns are migratory, and that the sampled individuals may have taken different amounts of time to transfer from arriving to laying the first egg (Moiron et al., 2023), we added the 'interval between arrival and laying date' as a covariate. Moreover, we added the 'interval between laying and sampling' as another covariate to correct for any potential methodological variation in the speed with which we could sample birds in the field
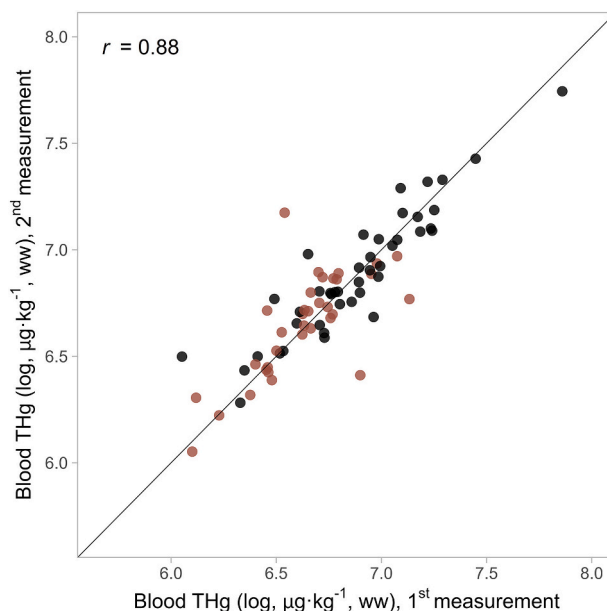
Fig. 1. Correlation (*r*) between two total blood mercury concentration measurements of the same blood sample (*n* = 43 male samples in black, 34 female samples in red). The line represents the x = y line, the slope of the correlation between both values equals 0.82 ± 0.05 SE.

(which mainly depends on the synchrony of the colony's breeding). Finally, we tested for sex-specificity of our candidate variables by including all interactions between 'sex' and the terms introduced above.

Given our finding of age-specificity of blood THg levels (see Results and Table S1), we ran a second linear mixed model where used a within-subject age standardization to separate within- and among-individual age effects. Hereto, we partitioned the age of each individual into an 'average age' and 'delta age' component (following van de Pol and Wright, 2009). An individual's average age was calculated as the average of all ages at which the bird was sampled, while delta age was calculated as the difference between an individual's actual age at sampling and its average age (i.e., 'delta age' = 'actual age' – 'average age', see Table S2 for an example). When including both 'average age' and 'delta age' as covariates in the model, 'average age' quantifies the among-individual age effect, whereas 'delta age' quantifies the within-individual change in THg concentration as birds grow older (van de Pol and Wright, 2009). If the among- and within-individual age effects were to be significantly different from each other, this would indicate that the effect of age among individuals cannot be explained by changes within individuals, meaning that there is age-specific selective (dis)appearance of

Table 2

Results from three models testing whether selected candidate factors explain variation in (log$_e$-transformed) total blood mercury concentrations ($\mu g \cdot kg^{-1}$ ww) of 1209 samples taken from 588 common terns for (a) both sexes combined, (b) females, and (c) males.

(a) Both sexes (n = 1209)	Estimate ± SE	t-Value	p-Value
Intercept	6.577 ± 0.1097	59.977	**<0.001**
Sex (female)	−0.137 ± 0.105	−1.306	0.192
Clutch size (2)	−0.107 ± 0.066	−1.613	0.107
Clutch size (3)	−0.095 ± 0.066	−1.449	0.148
Interval AD – LD	0.005 ± 0.001	4.611	**<0.001**
Interval LD - SD	0.005 ± 0.002	2.119	**0.034**
Average age	0.027 ± 0.003	9.821	**<0.001**
Delta age	0.070 ± 0.020	3.523	**<0.001**
Sex (female) * clutch size (2)	0.010 ± 0.084	0.119	0.905
Sex (female) * clutch size (3)	−0.015 ± 0.083	−0.186	0.853
Sex (female) * interval AD – LD	−0.006 ± 0.002	−3.661	**<0.001**
Sex (female) * interval LD – SD	0.006 ± 0.003	1.877	0.061
Sex (female) * average age	−0.007 ± 0.004	−1.853	0.064
Sex (female) * delta age	−0.043 ± 0.026	−1.654	0.099
Average age * delta age	−0.005 ± 0.002	−3.322	**<0.001**
Sex (female) * average age * delta age	0.004 ± 0.002	1.862	0.063

Random effects			Variance ± SD
Year			0.038 ± 0.196
Identity			0.020 ± 0.144
Residual			0.044 ± 0.211

(b) Females (n = 612)	Estimate ± SE	t-Value	p-Value
Intercept	6.459 ± 0.098	66.211	**<0.001**
Clutch size (2)	−0.092 ± 0.050	−1.850	0.065
Clutch size (3)	−0.106 ± 0.050	−2.137	**0.033**
Interval AD – LD	−0.001 ± 0.001	−0.740	0.460
Interval LD – SD	0.011 ± 0.002	4.589	**<0.001**
Average age	0.020 ± 0.003	7.520	**<0.001**
Delta age	0.033 ± 0.018	1.799	0.073
Average age * delta age	−0.002 ± 0.002	−1.017	0.310
Average age (sel. d. app.)	0.022 ± 0.011	0.211	0.833
Actual age (sel. d. app.)	0.017 ± 0.010	1.748	0.081

Random effects			Variance ± SD
Year (r)			0.034 ± 0.183
Identity (r)			0.016 ± 0.125
Residual (r)			0.043 ± 0.207

(c) Males (n = 597)	Estimate ± SE	t-Value	p-Value
Intercept	6.564 ± 0.116	56.629	**<0.001**
Clutch size (2)	−0.116 ± 0.068	−1.699	0.090
Clutch size (3)	−0.107 ± 0.068	−1.578	0.115
Interval AD – LD	0.005 ± 0.001	4.507	**<0.001**
Interval LD – SD	0.006 ± 0.003	2.232	**0.026**
Average age	0.028 ± 0.003	9.184	**<0.001**
Delta age	0.065 ± 0.021	3.144	**0.002**
Average age * delta age	−0.005 ± 0.002	−3.131	**0.002**
Average age (sel. d. app.)	0.018 ± 0.011	1.595	0.111
Actual age (sel. d. app.)	0.009 ± 0.011	0.874	0.382

Random effect			Variance ± SD
Year (r)			0.044 ± 0.210
Identity (r)			0.027 ± 0.164
Residual (r)			0.045 ± 0.213

Significant results (p < 0.05) are presented in bold. 'Average age (sel. d. app.)' and 'Actual age (sel. d. app.)', both in years, report the results for the analysis in which we test whether the average and delta age estimates obtained in the main model are significantly different from one another; all other results reported are obtained from the main model in which 'average age' and 'delta age', also in years, and their interaction are included. 'Interval AD – LD' represents the interval between arrival and laying date, 'Interval LD – SD' the interval between laying and sampling date, both in days. Clutch size refers to the size of the clutch the bird was incubating while its blood sample was collected.

individuals with certain levels of THg concentrations from the sampled pool of individuals (van de Pol and Wright, 2009). In addition to the main effects of 'average age' and 'delta age', we added the interaction between them to test for non-linearity of any within-individual change in THg concentration with age, such as accelerating or decelerating effects as birds grow older. Finally, as in the first model, we tested for sex-specificity of our candidate variables by including all interactions between 'sex' and the introduced terms, as well as the three-way interaction between 'average age', 'delta age' and 'sex'. Candidate variables were not highly correlated (Fig. S2), such that we were able to enter them simultaneously.

Given that we found a significant interaction between 'sex' and 'interval between arrival and laying date' (in both the first and second model, see Results and Table S1), we proceeded to run the second model for males and females separately, excluding the interactions with sex as well as the main effect of sex (Table 2b and c). In addition, we repeated these sex-specific models while replacing 'delta age' by the 'actual age' of an individual, to test whether the 'average age' and 'delta age' effects were significantly different from one another (van de Pol and Wright, 2009), which would indicate selective (dis)appearance of birds with low or high THg levels (see above, Tables 2b and c).

To determine how two THg concentrations obtained from the same bird in the same season correlate, we ran a final linear mixed model with the $\log_e$-transformed 'second blood THg concentration' as the dependent variable and the 'first blood THg concentration' as a covariate. As above, 'individual identity' and 'year' were included as random effects. 'Clutch' of the second sample (same or replacement) and 'sex' were included as 2-level class variables. Additionally, we added the interaction between (i) 'clutch' and 'sex' to test whether the fact that females laid eggs in between the two measurements in cases where the second sample was taken while the birds were incubating their replacement clutch made a difference, as well as (ii) 'clutch' and 'first blood THg concentration' to account for the fact that the temporal interval between the two measurements was larger in cases where the second sample was taken while the birds were incubating a replacement clutch. Further, we included the three-way interaction between 'clutch', 'sex' and 'first blood THg concentration' because we expected the correlation to be stronger for males than females in cases when the clutch was different, but not when the clutch was the same.

All analyses were performed, and figures created, using R (version 4.3.1, R Core Team, 2022). Models were run with the R package "lme4" (Bates et al., 2015). *P*-values were obtained using the package "lmerTest" (Kuznetsova et al., 2017) with the level of significance set at $p < 0.05$. Data were visualized using the R packages "ggplot2" (Wickham, 2016) or "interactions" (Long, 2022).

3. Results

The total mercury concentration in 1209 blood samples collected from 588 individual common terns ranged from 0.29 to 3.90 $\mu g \cdot g^{-1}$ ww, with a mean $\pm$ SE of 1.00 $\pm$ 0.11. Part of this variation was explained by significant positive linear and negative quadratic age effects, with older birds exhibiting higher THg concentrations in their blood than younger birds (Table S1, Fig. 2).

To distinguishing between within-individual accumulation of mercury with age and among-individual variation in survival in relation to mercury pollution level driving the observed age effect, we repeated the model splitting age into average and delta age components. This model showed that part of the variation in the terns' blood THg concentrations was explained by a significant interaction between sex and 'interval between arrival and laying date', with the interaction between sex and 'interval between laying and sampling date' and the three-way-interaction between sex, average age and delta age also being close to significance (Table 2a), such that we subsequently ran sex-specific models to assess effects for males and females separately.

The sex-specific model for females showed that there was no
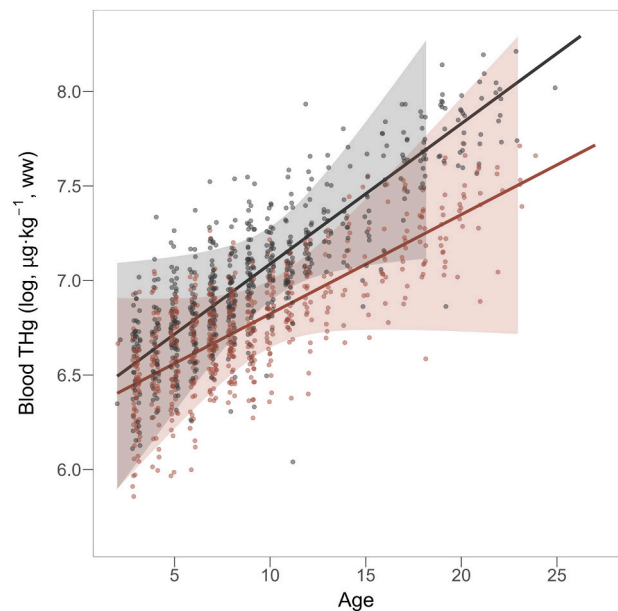
Fig. 2. Total blood mercury concentrations of common terns in relation to age. Each point represents the partial residual of a blood sample, lines the sex-specific model predictions and shaded areas the confidence intervals (black = males, red = females).
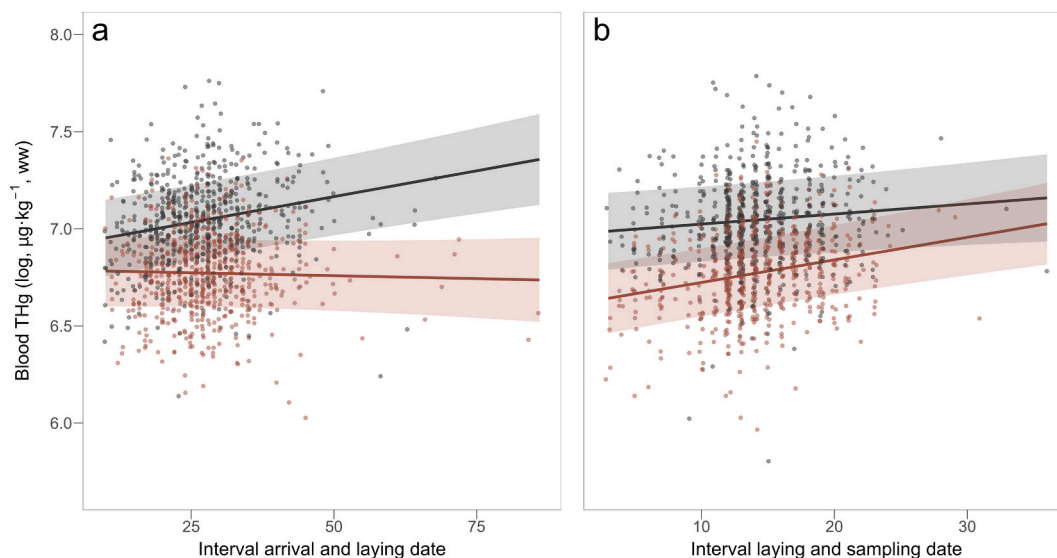
correlation between the interval between arrival and laying of the first egg and female THg concentration (Fig. 3a, Table 2b), but that THg levels were higher in females with a longer interval between first egg laying and blood sampling (Fig. 3b). In addition, females that laid three eggs had significantly lower THg levels than those that laid one egg, with females laying two eggs showing an intermediate THg level that was not significantly different from either (Fig. S3). Females with a higher average age had higher THg levels than those with a lower average age (Fig. 2), which most likely can be explained by within-individual accumulation of THg with age, given that the within-individual term for delta age showed a trend towards being positive (Fig. 4) and the within- and among-individual estimates not differing from one another, such that selective (dis)appearance in relation to THg should not be underlying variation among females.

The sex-specific model for males showed significantly positive correlations between the interval between arrival and laying (by their partner) and the interval between laying (by their partner) and blood sampling and male THg levels (Fig. 3a and b, Table 2c). Additionally and interestingly, THg levels were lower in males whose partner laid a larger clutch, although not significantly so (Fig. S2). Similar to females, THg levels in the blood of older males were higher than those in younger ones (Fig. 2), which can be attributed to a significant within-individual accumulation, although one that in males decelerated with age (Fig. 4), given that the within- and among-individual patterns did not differ from one another, but did show a significant interaction.

Our within-season analysis revealed that variation in the THg concentration at the second measurement within a season can be explained by a significant interaction between clutch and THg concentration at the first measurement, with there being a trend towards this interaction differing between the sexes (Table 3). These interactions show that the correlation between the two THg concentrations is stronger when measured within the same clutch, especially for females (Fig. 5).

4. Discussion

In this study, we used seven years of longitudinal data to assess factors underlying variation in total mercury concentration (THg) in the blood of a long-lived seabird, the common tern. THg concentrations

Fig. 3. Total blood mercury concentrations of male (black) and female (red) common terns in relation to their (a) interval between arrival and laying date and (b) interval between laying and sampling date, both in days. Each point represents the partial residual of a blood sample, lines the sex-specific model predictions and shaded areas the confidence intervals.
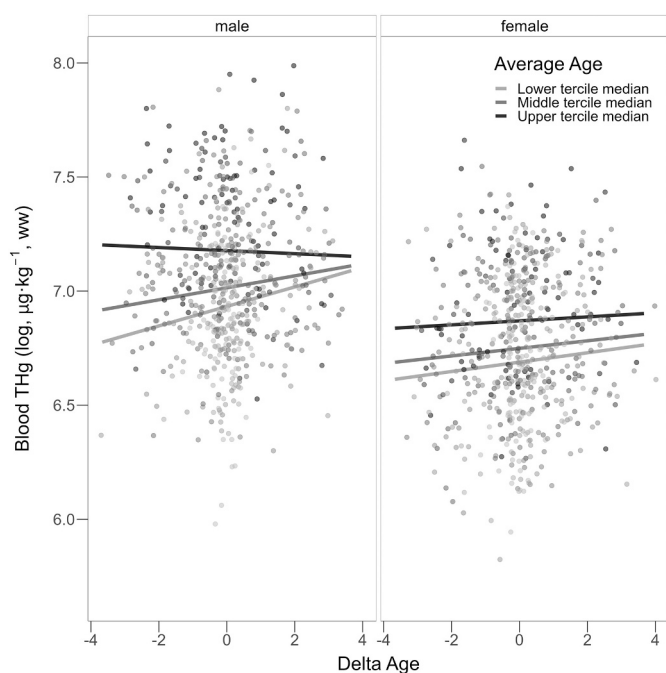
Fig. 4. Total blood mercury concentrations of male (left) and female (right) common terns in relation to the age, in years, at which they were sampled relative to the average age at which they were sampled ('delta age'). Each point represents the partial residual of a blood sample; lines the model predictions for the within-individual change across seven years for birds sampled at an average age at the lower (5 years), middle (8 years) and upper (14 years) tercile median (from light grey to black, respectively).

were highly variable, ranging from 0.29 to 1.80 μg·g⁻¹ ww in females and from 0.35 to 3.90 μg·g⁻¹ ww in males, showing an overall mean of 1.00 ± 0.11 μg·g⁻¹ ww.

Apart from the overall difference in blood THg concentrations between male and female common terns, we observed differences between the sexes in the correlations between some of our variables of interest and THg concentrations (Table 2a). Consequently, we ran sex-specific analyses. With respect to phenology, for males, the sex-specific

Table 3

Results from a model testing whether clutch (same or replacement), sex and/or $\log_e$-transformed first total blood mercury concentration explain variation in the ($\log_e$-transformed) second total blood mercury concentrations (μg·kg⁻¹ ww) of 88 common terns sampled multiple times within a season.

Independent variable	Estimate ± SE	t-Value	p-Value
Intercept	2.466 ± 0.832	2.964	**0.004**
Clutch (replacement)	2.387 ± 0.878	2.718	**0.009**
First Hg concentration	0.649 ± 0.118	5.488	**<0.001**
Sex (female)	0.171 ± 1.086	0.158	0.875
Clutch (replacement) * first THg concentration	−0.324 ± 0.124	−2.604	**0.013**
Clutch (replacement) * sex (female)	1.147 ± 1.443	0.795	0.430
Clutch (replacement) * first THg concentration * sex (female)	−0.028 ± 0.159	−0.173	0.863
Clutch (replacement) * first THg concentration * sex (female)	−0.253 ± 0.151	−1.677	0.099

Random effect	Variance ± SD
Year	0.017 ± 0.131
Identity	0.026 ± 0.161
Residual	0.013 ± 0.115

Significant results (p < 0.05) are presented in bold.

analysis showed that males with longer intervals between arrival and the onset of laying by their female, and between laying and sampling had higher blood THg levels (Table 2c). This may suggest that a longer time spent at the breeding area results in males being exposed to a higher dietary uptake of methylmercury (MeHg) compared to the uptake in their foraging areas prior to arrival. This seems supported by a recent study showing an increasing temporal trend in mercury contamination of fish in this area (Kammann et al., 2023). Alternatively or additionally, however, the causality may be reversed, such that males with higher THg levels might have arrived in a poorer condition, for example due to MeHg exacerbating oxidative stress (Hoffman et al., 2011; Costantini et al., 2014; Eckbo et al., 2019; Soldatini et al., 2020) and/or hormone levels (Varian-Ramos et al., 2014), resulting in delayed breeding and/or reduced incubation behaviour, leading to us needing longer to sample them successfully.

In females, there was no correlation between the interval between arrival and egg laying and THg concentrations, while the correlation
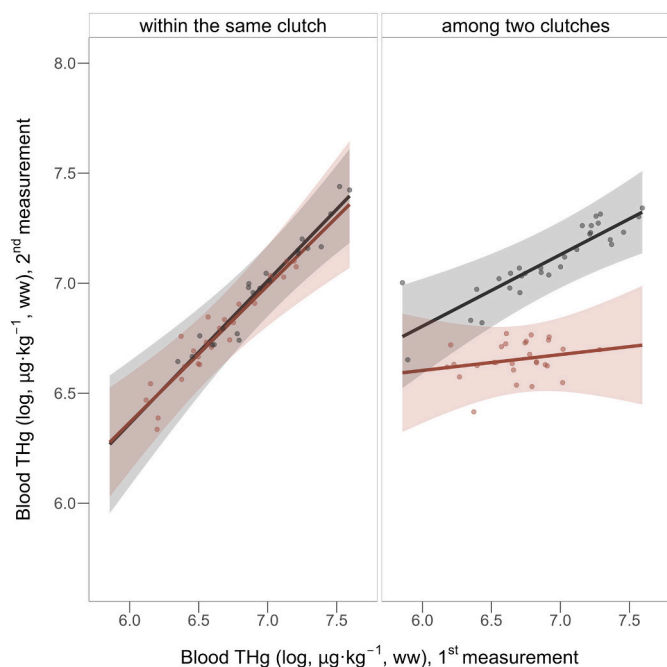
Fig. 5. Total blood mercury concentration at the second sampling event within a season, either within the same clutch ($n = 47$, left) or among two clutches ($n = 58$, right). Each point represents the partial residual of a second blood sample, lines the model predictions for each sex and shaded areas the confidence intervals (black = males, red = females).

between the interval between laying and sampling and THg concentrations was significantly positive (Table 2b) and showed a trend towards being slightly stronger than in males (Table 2a). This difference between the sexes might be explained by females' ability to reduce mercury concentrations via egg laying (Bond and Diamond, 2009; Ackerman et al., 2020). If a longer time spent at the breeding area indeed results in higher blood THg concentrations due to high environmental contamination, egg laying could initially offset an increase in blood THg concentrations in females, but not males. However, after clutch completion, this additional excretion mechanism would cease, leading to an increase in female THg levels over time as well. This hypothesis is supported by our results obtained from the repeated THg measurements within breeding seasons. Whereas the within-individual correlation between THg concentrations is similarly positive for the sexes if both measurements were obtained from blood samples taken within a single clutch, it is somewhat weaker for females than males if the second blood sample was taken during incubation of a replacement clutch, most likely due to a repeated reduction of THg levels by females through egg laying.

A considerable role of egg laying for female THg detoxification is additionally suggested by our finding that the larger the number of eggs in the clutch, the lower the blood THg concentrations. Similar evidence of egg laying being an efficient route of mercury excretion for female terns is provided by studies showing that mercury deposition in eggs decreases across the laying sequence, likely due to a reduced 'availability' of mercury in females for excretion into subsequent eggs after the first one is laid (Becker, 1992; Akearok et al., 2010; Ackerman et al., 2016). Previous studies, however, also indicated that other factors may additionally contribute to sex-differences in THg contamination (Robinson et al., 2012; Hitchcock et al., 2019). In double-crested cormorants (*Phalacrocorax auritus*), females, for example, were shown to have greater capacity to demethylate mercury in the liver than males (Robinson et al., 2011). Additionally or alternatively, males and females may differ in their levels of mercury excretion (Braune and Gaskin, 1987b). Given that moult does not take place during the early breeding season,

excretion through faeces seems the only potential pathway in this respect. We, however, deem this unlikely to be important in our case, because faecal mercury exists only in inorganic form (Morrissey et al., 2005), and the uptake of inorganic mercury is minimal. However, further studies should address the potential impact of excretion mechanisms on sex-specific blood THg levels, as well as the effects of mercury deposited in eggs for the offspring (Ackerman et al., 2020).

Older birds of both sexes had significantly higher THg levels in their blood compared to young birds. We found no evidence for selective (dis) appearance of either weakly or heavily contaminated individuals, and our longitudinal data suggest that the higher THg levels in older individuals result from within-individual THg accumulation with age, the shape and extent of which differed somewhat between the sexes. Specifically, within-individual THg concentrations of males exhibited significant decelerating accumulation, whereas in females there was a trend towards a lesser but linear within-individual increase. Again, the ability of females to excrete MeHg through egg laying could explain why we observe a weaker accumulation with age in females than in males. However, this does not explain the trend towards the non-linear component of the within-individual age effect differing between males and females, given that female terns of our population increase their clutch size as they grow older (Zhang et al., 2015) and therefore would, if anything, be expected to show a stronger deceleration with age than males. Sex-specificity of mercury uptake, via the use of differential foraging areas (Carravieri et al., 2014; Militão et al., 2023) or differential prey choice (De Felipe et al., 2019), which additionally might be age-specific, is an alternative explanation that deserves empirical attention.

As also alluded to above, birds possess mechanisms that should mitigate the MeHg concentrations in their bodies. It is well-established that a substantial portion of ingested MeHg is deposited in feathers (Braune and Gaskin, 1987a), a phenomenon that may explain why in some bird species, such as wandering albatrosses, adults, undergoing frequent moulting, exhibit lower, rather than higher THg levels in blood compared to immature birds undergoing sporadic moults (Tavares et al., 2013; Bustamante et al., 2016). Moreover, mercury that is not excreted, such as that ingested during non-moulting periods, is typically demethylated in the liver to inorganic mercury by binding with selenium at a high rate (e.g. Eagles-Smith et al., 2009b; Manceau et al., 2021). These highly efficient mechanisms could account for the absence of mercury accumulation in blood observed in previous (but cross-sectional) studies on birds (Thompson et al., 1993; Becker et al., 2002; Carravieri et al., 2014, 2022; Tartu et al., 2015). In addition, they could explain the rather slow rate of accumulation found in this study, although the fact that accumulation still occurs suggests that these mechanisms are limited.

The difference in the average THg levels between young (first quartile of age, $0.90 \mu g \cdot g^{-1}$ ww, $n = 318$) and old (fourth quartile of age, $1.17 \mu g \cdot g^{-1}$ ww, $n = 308$) birds equals 30 %, which combined with a relatively low individual repeatability of 20 % (i.e. total phenotypic variance explained by individual identity as a random effect in the model described in Table 2a), raises concerns about the suitability of blood from birds of unknown age as an accurate bioindicator of environmental mercury levels, at least for common terns. The influence of age on mercury levels might, however, be less pronounced in other study systems, especially when considering additional variables such as isotopic data (Polito et al., 2016; Renedo et al., 2018a, 2018b; Jouanneau et al., 2022), different foraging locations (e.g., Ackerman et al., 2016), or species preying at lower trophic levels (Ma et al., 2021) or having shorter lifespans (Hallinger et al., 2011). Assessing the generality of the age-specificity of blood mercury contamination we found therefore is important. Moreover, further studies are also needed to investigate the age-specificity of (maternal) mercury levels in other commonly used bioindicator tissues, such as feathers (Sun et al., 2020) or eggs (Evers et al., 2003; Goodale et al., 2008).

It is noteworthy that the THg values of all of our 1314 blood samples

collected from 588 individual common terns exceeded the toxicity thresholds at which adverse health effects have been observed, with 57 % being at low (0.2–1.0 µg·g^{-1} ww) and 43 % being at moderate (1.0–3.0 µg·g^{-1} ww) risk levels, following the toxicity benchmarks published by Ackerman et al. (2016). As an endocrine disruptor, MeHg has been reported to decrease breeding probability (Tartu et al., 2013; Goutte et al., 2015), to alter risk-taking (Kobiela et al., 2015) or parental behaviour (Tartu et al., 2015) and oxidative stress status (Costantini et al., 2014; Eckbo et al., 2019; Soldatini et al., 2020). Such effects could potentially impair the ability to adapt to environmental changes (Jenssen, 2006; Grunst et al., 2023a) or to mount an effective immune response against disease (Hawley et al., 2009; Lewis et al., 2013). Moreover, mercury has been associated with a reduced body condition (Amélineau et al., 2019; Eckbo et al., 2019; but see Carravieri et al., 2022), which can be particularly harmful for migratory birds (Seewagen, 2020) when their flight performance (Ma et al., 2018a) or fat deposition and muscle development (Adams et al., 2020) would be compromised.

Overall, our study suggests that the risk of suffering from the detrimental effects of mercury pollution differs between the sexes and increases as adult birds age. Although we so far found no evidence of selective (dis)appearance and previous studies on adult seabirds in wild populations mostly also reported no direct correlation between survival and mercury contamination (Thompson et al., 1991; Goutte et al., 2015; Bustamante et al., 2016; Pollet et al., 2017; Amélineau et al., 2019; Chastel et al., 2022; but see Calvert et al., 2024), interactions between mercury pollution and other stressors may have synergistic effects (Costantini et al., 2014; Grunst et al., 2023b; Smith et al., 2023) and further studies are needed to assess how mercury pollution interacts with such stressors and whether effects may be sex- and/or age-specific as well.

CRediT authorship contribution statement

Justine Bertram: Writing – original draft, Visualization, Resources, Investigation, Formal analysis. **Coraline Bichet:** Writing – review & editing, Resources. **Maria Moiron:** Writing – review & editing, Formal analysis. **Peter J. Schupp:** Writing – review & editing, Resources. **Sandra Bouwhuis:** Writing – review & editing, Supervision, Project administration, 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.

Data availability

Data will be made available on request.

Acknowledgments

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.172330.

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