Science of the Total Environment 953 (2024) 175891

Contents lists available at ScienceDirect

Science of the Total Environment

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

Levels of trace elements in the blood of chick gulls from the English Channel: Spatial and trophic implications

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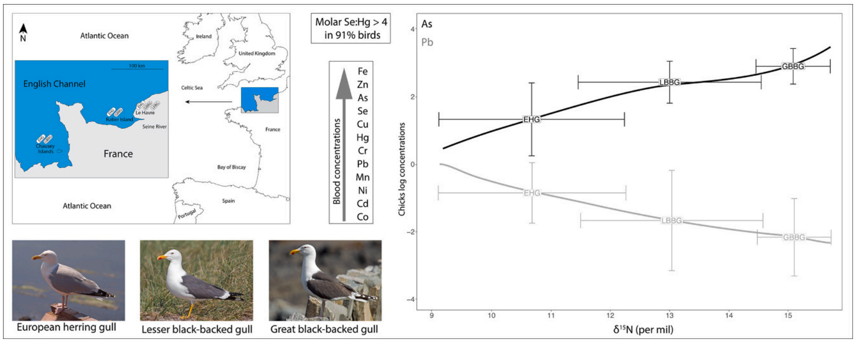
HIGHLIGHTS

- Most elements were trophic or habitat dependent (proxied by $\delta^{15}N$, $\delta^{13}C$ and $\delta^{34}S$).
- As and Hg were biomagnified in the food web studied, while Pb revealed biodilution.
- Cd, Cr and Ni had higher levels in the Chausey Islands than at the Seine estuary.
- A low Se:Hg molar ratio suggests a toxic risk for 8.6 % of great black-backed gulls.
- No relationship between element concentrations and body condition was observed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Rafael Mateo

Keywords:
Trophic tracers
Seabirds
Bioaccumulation
As
Pb
Se:Hg
$\delta^{15}N$
$\delta^{13}C$
$\delta^{34}S$
Stable isotopes
SMI
The Seine

ABSTRACT

Anthropogenic activity has disturbed the natural distribution and circulation of trace elements in the environment. This has led to increased background levels of numerous elements, causing global pollution. In this context, seabirds are relevant bioindicators of environmental contamination. This study focuses on the ecological factors that influence the concentrations of 14 trace elements in the blood of the chicks of three sympatric gull species from the French coast of the English Channel. Between 2015 and 2017, 174 birds were sampled in the industrialised Seine Estuary (in the city of Le Havre and on Ratier Island) and in the remote Chausey Islands, 200 km to the west. We also considered the Se:Hg molar ratio using Hg concentrations in those birds. Ag and V concentrations were below the quantification limit in all cases, while the fraction of non-quantified samples was higher than 30 % for Cd, Cr and Ni. Among the elements quantified in the samples, the lowest concentrations were noted for Co and the highest for Fe, building the following order: Co $<$ Cd $<$ Ni $<$ Mn $\leq$ Pb $<$ Cr $<$ Hg $<$ Cu $<$ Se $<$ As $<$ Zn $<$ Fe. No unanimous scheme of concentrations among elements, species and sites existed. Similarly, different models were fitted and different factors were significant for different species and elements. We observed the biomagnification of As and the biodilution of Pb. Pb concentrations were also highest in the industrial site in the city of Le Havre. Despite the high proportion of non-quantified samples for Cd, Cr and Ni, we

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https://doi.org/10.1016/j.scitotenv.2024.175891
Received 23 April 2024; Received in revised form 31 July 2024; Accepted 28 August 2024

continued to notice higher concentrations in the marine environment of the Chausey Islands. Concentrations of some elements clearly revealed habitat dependence. In some cases the Se:Hg molar ratio was lower than 4, a threshold for diminishing Hg toxicity by Se.

1. Introduction

Trace elements have always been natural components of the environment (Bryan, 1984; Sen and Peucker-Ehrenbrink, 2012). Thousands of scientific papers, however, have discussed the environmental pollution caused by trace elements, which may at first glance seem controversial because of their natural origin (reviewed by Ali et al., 2019; Sahu and Basti, 2021; Edo et al., 2024; Evers et al., 2024). The reason for this lies in the fact that anthropogenic activities, such as people's day-to-day lives, industrial activities and the exploitation of the environment have caused considerable disturbance in the distribution of trace elements throughout the world. This has moved a significant proportion of some trace elements (e.g. As, Cd, Hg, and Pb) from inaccessible deposits to deposits highly accessible to biota and consequently food webs (Nordberg et al., 2007). In this context, there is a need to understand the exposure of organisms to trace elements, the influencing factors, and the distribution of pollution. Seabirds are commonly exposed to trace elements through diet (Furness and Camphuysen, 1997; Barwick and Maher, 2003). They are long-lived animals that usually feed on high trophic levels, which promotes the elemental bioaccumulation in some tissues and exposure to elements that biomagnify along food webs (Blévin et al., 2013; Carravieri et al., 2020). At the same time, they are suitable bioindicator organisms for the study of contamination, since they are common species to many parts of the world, relatively easily accessible during the breeding season and non-lethal sampling of them is relatively easy (Kalisińska et al., 2004; Elliot and Elliot, 2013; Albert et al., 2019). A number of factors, however, influence adult trace element exposure during their life cycle, including age, reproduction and migration patterns (Fort et al., 2014; Albert et al., 2021, 2022; Cherel et al., 2018). In this respect, chicks are more indicative in the study of local chemical contamination and its relationship with environmental factors (e.g. food diversity, feeding habitat), since they are, for most species, fed on prey caught by their parents in the vicinity of the colony (Blévin et al., 2013; Carravieri et al., 2020).

The English Channel forms part of the north-eastern Atlantic Ocean, considerably influenced by anthropogenic contamination. The source of the contamination is at some places local (e.g., the city of Le Havre), but a significant share of contaminants comes from distant anthropogenic sources (e.g., industry in the city of Rouen) that reach the English Channel by large rivers such as the Seine (Meybeck et al., 2007; Flipo et al., 2021; El Haddad et al., 2024). A number of studies have shown high concentrations of trace elements in biota along the French coast of the Channel, but studies have yet to be made for seabirds (e.g. Lahaye et al., 2007; Henry et al., 2012; Larsen and Hjermann, 2022). To the best of our knowledge, only two comprehensive studies were published for this area for mercury (Hg) on seabirds (Binkowski et al., 2021; Lemesle et al., 2024). The fate of contaminants in the environment needs to be better understood and monitored to protect marine ecosystems and biodiversity (Mauffret et al., 2023). Monitoring contaminants such as trace elements is necessary to assess good ecological status under descriptor 8 of the European Water Strategy Framework Directive (MSFD, 2008/56/EC). Bivalves have been used for this purpose in France since the late 1970s, but they only provide information on local and coastal contamination at the base of food webs (Mille et al., 2023). On the other hand, top predators such as birds (consumers at a higher trophic level) provide integrated information on the contaminants transferred into the food webs.

In this general context this paper aimed to understand exposure of seabird chicks to trace elements under the potential influence of sex, feeding habitat and trophic position. For this purpose, we evaluated the concentrations of 13 other trace elements (silver (Ag), arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), selenium (Se), vanadium (V) and zinc (Zn)), as well as the molar ratio between Se and Hg (Se:Hg) in the blood cells of the chicks of three sympatric gull species: the European herring gull (*Larus argentatus*), the lesser black-backed gull (*L. fuscus graellsii*) and the great black-backed gull (*L. marinus*) from different breeding sites and industrialisation levels along the French coast of the English Channel. To consider the influence of the trophic ecology of birds, stable isotopes of carbon (C) and sulfur (S) were analysed as proxies of their feeding habitat and stable isotopes of nitrogen (N) as a proxy of their trophic position (Post, 2002). We tested the hypothesis that trace element concentrations measured in the chicks are related to 1) the sex of the individuals, 2) the location of the colony, 3) the trophic position, 4) the habitat and origin of their prey (marine vs. terrestrial). We also investigated whether the chicks' body condition (proxied by scaled mass index (SMI)) was related to their level of contamination. We predicted that the location and feeding habitat were significant factors for trace element concentrations, as local emission usually influences the exposure. In contrast, we expected that sex differences were insignificant, as we suspected no substantial discrepancies in physiology between male and female chicks, and that the trophic position would have had a limited effect on trace element concentrations, since biomagnification and biodilution have only been shown for some elements (Sun et al., 2020).

2. Materials and methods

2.1. Sampling sites and study species

Birds were sampled over three years (2015–2017) in the last week of June and the first week of July in the three areas along the French coast of the English Channel (Fig. 1). Two of them, further combined into one study site (the Seine Estuary), were highly urbanised and putatively polluted environments: the city of Le Havre (N49°29′, E0°12′; nests distributed across the city on the roofs of buildings) and 8 km away in

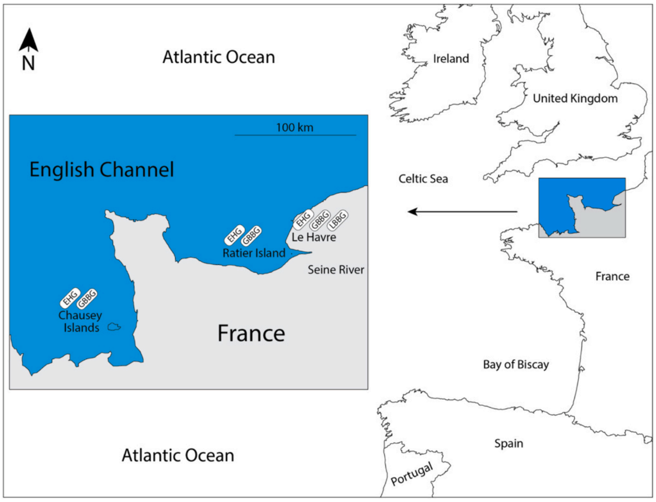

Fig. 1. Sampling sites along the southern part of the English Channel along the French coast, Ratier Island and the city of Le Havre, as parts of the Seine Estuary, and the more remote the Chausey Islands. EHG: European herring gull; GBBG: great black-backed gull; LBBG: lesser black-backed gull.

the centre of the estuary Ratier Island (N49°25′46″, E0°08′7″; nests built on the ground). The third site, 200 km away in the Normand-Breton Gulf, was far from industrial activity and was treated as a reference site: the Chausey Islands (N48°53′, W1°49′; nests built on the ground on a number of islands).

Chicks of the herring gull (hereafter EHG) and great black-backed gull (hereafter GBBG) were sampled in both study sites, while lesser black-backed gull (hereafter LBBG) were sampled only in the city of Le Havre, yielding a total of 174 birds captured (approximately two months old) and sampled. The primary distinction between these sympatric species lies in their foraging ecologies. GBBG occupies the highest trophic position as the most predatory and marine species, preying on fish, invertebrates, refuse (such as anthropogenic food and resources from landfills), and other birds. In contrast, EHG holds the lowest trophic position and exhibits greater dietary flexibility, consuming offshore, onshore and anthropogenic food (Threlfall, 1968; Ewins et al., 1994; Rail and Chapdelaine, 2000). LBBG by this reckoning falls between the GBBG and the EHG, with preferences for onshore and anthropogenic food (Gyimesi et al., 2016). The diet of chick gulls is similar to the adult ones since they rely on parental food provisioning (Inzani et al., 2024).

2.2. Sample collection

For each individual, blood (ca. 2 mL) was sampled from the brachial vein with a 25G needle and heparinised syringe. Immediately upon collection the samples were placed in a cool box. After the sampling session, blood samples were centrifuged at 4000 rpm for 6 min at 4 °C to obtain red blood cells for stable isotope and trace element analyses. The separated fractions were frozen at −80 °C until further laboratory analyses.

2.3. Biological information

The birds were sexed using samples of red blood cells following Fridolfsson and Ellegren (1999) as described in Binkowski et al. (2021).

As body condition index, we used a scaled mass index (SMI) that utilises type II regression (SMA) to account for specimen size to calculate a body mass assuming an equal size of specimens (Peig and Green, 2009). In brief, morphometric parameters were determined for each bird, including the length of the wing, head with bill, culmen and tarsus. As the length of the head with the bill correlates best with the body mass ($r = 0.92$), it was used in the SMI calculations based on the formula:

$$SMI_i = M_i\left(\frac{L_0}{L_i}\right)^{b_{SMA}}$$

where M_i and L_i stand for body mass and length of head with the bill for a given specimen, L_0 represents the mean length of head with the bill for the given sex and species, and b_{SMA} represents the slope of SMA regression between body mass and length of head with the bill (both log-transformed) for the given species and sex (Kucharska et al., 2022). The SMA was calculated using the “lmodel2” package (Legendre, 2018) in the R environment (R Core Team, 2022).

2.4. Analysis of trace elements

After being freeze-dried for 48 h, samples of red blood cells (hereafter blood) were ground to powder. Subsamples of a measured weight (mean: 0.21 g, SD: 0.05, min-max: 0.06 to 0.29 g; the differences were due to the varying quantities of blood sampled) were microwave digested in a 6 mL mixture of nitric acid and hydrochloric acid (Fisher Scientific Trace Metal Quality) with a volume ratio of 3:1. For samples weighing <0.10 g the volumes of the acids were divided by two. Samples were further diluted with ultrapure water to 50 mL (25 mL for samples <0.10 g).

Concentrations of all studied elements (Ag, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, V, and Zn) were analysed by Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES; Varian Vista-Pro) and Inductively Coupled Plasma Mass Spectrometer (ICP-MS; X Series II Thermo Fisher Scientific) (Table S1). The concentrations were expressed as $\mu g \cdot g^{-1}$ of dry weight (dw).

The accuracy and reproducibility of the methods used were validated with analytical blanks and Certified Reference Material (CRM) analyses: National Research Council, Canada (NRCC) TORT-3 (Lobster Hepatopancreas) and NRCC DOLT-5 (Dogfish liver). The CRM weights were adjusted to represent quantities of elements similar to those in the samples studied. Recoveries fitted a range between 83.4 and 123.1 % (Table S2). Blanks and CRMs were analysed at the beginning of each batch of 40 samples. To avoid contamination with the elements studied, all glassware and plasticware used in the protocol were thoroughly washed with detergent, rinsed with Milli-Q quality water and soaked in a nitric acid bath (10 mL/L). After at least 48 h utensils were rinsed in ultrapure water (Milli-Q) and dried under a laminar flux hood.

Total Hg (hereafter Hg) concentrations used in the molar ratio Se:Hg were measured in blood using an atomic absorption spectrophotometer (Altec AMA-254). The details of the method and detailed results of Hg concentrations are described in Binkowski et al. (2021).

2.5. Analysis of stable isotopes

The abundance of stable isotopes (C, N, S) was measured with a continuous flow mass spectrometer (Thermo Scientific Delta V Plus) coupled to an elemental analyser (Thermo Scientific EA Flash 2000). The results were expressed as standard delta (δ) notation as parts per thousand (‰) deviation relative to international standards (Vienna PeeDee Belemnite for C, atmospheric N_2 (air) for N and Vienna-Canyon Diablo Troilite for S). The characteristics of the method and detailed results are presented in Binkowski et al. (2021).

2.6. Statistical analysis

Statistical analyses were conducted in R ver. 4.2.1 (R Core Team, 2022) and a set of packages referenced below. The significance level was set at 0.05.

2.7. Design and analyses

The sampling allowed for no reliable temporal comparison, so we pooled the data from three years of study. In order to visualise the data and find relationships between elements, we calculated principal component analysis (PCA). For that, we used “factoextra” package (Kassambara and Mundt, 2020).

Since LBBG does not breed on the Chausey Islands, we split the statistical inference into two approaches: 1) small-scale: three species within the Seine Estuary (city of Le Havre and Ratier Island), and 2) a large-scale approach: two species (GBBG, EHG) along the French coast (Seine Estuary (pooled) vs. the Chausey Islands). Apparent differences between species meant that we built statistical models separately for each species studied. Before building the models, we scrutinised the data of concentrations graphically (histograms, scatter plots and ECD plots) in order to make a preliminary identification of their distribution. The Akaike Information Criterion (AIC) was then calculated for normal, log-normal and Gaussian distributions in order to compare the goodness of fit. That best-fitted distribution was further used in building generalised linear models. A similar approach was carried out to find the model's best link function. In most cases log-normal distribution with identity link function fitted best (Table S3).

The generalised linear models (GLM) were built for each element as the dependent variable, with the site as a discrete explanatory variable, isotopic values ($\delta^{13}C$, $\delta^{15}N$, $\delta^{34}S$) as continuous explanatory variables, and interactions. Since the isotopic signatures were all closely correlated, separate GLMs with particular isotopes were built to limit the collinearity. Model assumptions were verified based on plots of residuals of the initial models (Zuur et al., 2009). A set of models was built and the Akaike Information Criterion corrected for small sample sizes (AICc) was used to identify the best models. AICc was calculated by hand for censored regression models (see below) and with the "AICcmodavg" package (Mazerolle, 2020).

2.8. Dealing with results below LOQ

Some of the results obtained for the trace element concentrations fitted in a range between zero and LOQ of the ICP-MS instrument. Values below LOQ (hereafter NQs for non-quantified values) are less precise than those within the calibration curve's working range, but they still yield essential information. Since substituting NQs with a chosen value is questionable, we used methods adapted from survival analyses to calculate descriptive statistics (with the Kaplan–Meier method) to compare groups and to run explanatory analyses (with censored regression) (Helsel, 2012). Explanatory examinations and significance tests were calculated only for variables whose NQ fraction was <30 %. For calculations of descriptive statistics the threshold fraction of NQs was set as 80 %. For that part of the analysis, we used the "NADA" package (Lee, 2020).

3. Results

Among the elements studied, Cu (overall range 0.71–3.55 $\mu g \cdot g^{-1}$ dw), Fe (1219–2599 $\mu g \cdot g^{-1}$ dw), Mn (0.11–0.61 $\mu g \cdot g^{-1}$ dw), Se (1.02–22.9 $\mu g \cdot g^{-1}$ dw) and Zn (14.6–42.2 $\mu g \cdot g^{-1}$ dw) were quantified in all individuals, while As (<0.16–46.4 $\mu g \cdot g^{-1}$ dw), Co (<0.02–0.10 $\mu g \cdot g^{-1}$ dw) and Pb (<0.02–1.46 $\mu g \cdot g^{-1}$ dw) were quantified in at least 70 % of the individuals. Other elements (Ag, Cd, Cr, Ni, and V) presented quantifiable concentrations in <40 % of the individuals and were not used in further statistical modelling (Table S1). Concentrations of Hg (0.05–4.30 $\mu g \cdot g^{-1}$ dw) were thoroughly described in another paper (Binkowski et al., 2021).

There being no significant differences in concentrations between the sexes (with the exception of Zn in LBBG), we pooled male and female data in further analyses (Tables 1 and S4). We similarly failed to identify a well-fitted model for body condition (proxied by SMI) for GBBG and LBBG for the Seine Estuary and EHG for the French coast of the English Channel (Tables 2 and 3). The SMI of EHG from the Seine Estuary was, however, negatively linked with δ^{34}S, while the SMI of GBBG from the Chausey Islands was positively related to δ^{13}C.

3.1. Description of trace element concentrations and Se:Hg ratio

Mean concentrations in quantified blood samples were lowest for Co and highest for Fe, building the following order: Co < Cd < Ni < Mn ≤ Pb < Cr < Hg < Cu < Se < As < Zn < Fe (Table 3). The projection of all the elements (including Hg) by the PCA revealed that the first two PCs explained 49 % (31 % and 18 %, respectively) of the variance. With PC1 a strong correlation was observed for Fe, Pb, Se, and Zn, while PC2's affinity with As, Co, Cu, Hg, and Mn was observed. Biplot clearly distinguishes GBBG from the two other species (Fig. 2).

The Se:Hg molar ratio ranged from 2.2 in GBBG from the Chausey Islands to 215 in EHG for the city of Le Havre. A ratio lower than 4 occurred only in 15 specimens of GBBG. Visibly higher values were

Table 2

GLM models best explaining element blood concentrations and body condition (SMI – Scaled Mass Index) in gull chicks studied (according to the AICc values) within the Seine Estuary (Le Havre city and Ratier Island combined).

Metal	EHG	GBBG	LBBG
As[b]	δ^{15}N↑ + site + interactions	δ^{13}C↑ + site + interactions	δ^{13}C↑
Co[b]	δ^{34}S + site + interactions	δ^{13}C↑ + site + interactions	δ^{13}C↓
Cu	δ^{34}S↑ + site	δ^{34}S↑ + site + interactions	δ^{13}C↓
Fe	Null	δ^{13}C + site + interactions	Null
Mn	Null	δ^{34}S↑ + site	Null
Se	δ^{34}S↑ + site	δ^{34}S↑ + site + interactions	δ^{13}C↑
Pb[b]	δ^{15}N↓ + site + interactions	δ^{15}N + site + interactions	δ^{15}N↓
Zn	site	δ^{13}C↑	Null
SMI[a]	δ^{34}S↓	Null	Null

Grey indicates insignificant model components. EHG: European herring gull; GBBG: great black-backed gull; LBBG: lesser black-backed gull.

[a] In SMI modelling, PC1 and PC2 were used as potential explanatory variables.

[b] A censored regression was run for elements with non-quantified cases (As, Co and Pb).

Table 1

Concentrations of elements (mean ± SD, µg·g-1 dw) and Se:Hg molar ratios in the blood of gull chicks studied along the French coast of the English Channel. Concentrations of Hg have been thoroughly discussed by Binkowski et al. (2021).

	EHG			GBBG			LBBG
	City of Le Havre	Ratier Island	Chausey Islands	City of Le Havre	Ratier Island	Chausey Islands	City of Le Havre
Ag[a]	NQ (100 %)	NQ (100 %)	NQ (100 %)	NQ (100 %)	NQ (100 %)	NQ (100 %)	NQ (100 %)
As[a]	3.48 ± 3.68 (5.1 %)	4.83 ± 2.55	2.73 ± 1.45 (0 %)	18.1 ± 9.36	13.4 ± 5.98	24.8 ± 9.91	11.1 ± 5.70
Cd[a]	NQ (100 %)	NQ (100 %)	NQ (81.2 %)	NQ (97 %)	NQ (100 %)	0.04 ± 0.03 (13.3 %)	NQ 100 %)
Co[a]	0.03 ± 0.02 (25.6 %)	0.03 ± 0.01 (0%)	0.04 ± 0.02 (6.2 %)	0.02 ± 0.01 (56.8 %)	0.02 ± 0.01 (46.2 %)	0.05 ± 0.02 (13.3 %)	0.02 ± 0.01 (16.7 %)
Cr[a]	NQ (89.7 %)	NQ (100 %)	0.17 ± 0.48 (75 %)	0.05 ± 0.04 (78.4 %)	NQ (100 %)	0.23+0.53 (66.7 %)	NQ (87.5 %)
Cu	1.45 ± 0.47	1.07 ± 0.17	2.03 ± 0.54	1.56 ± 0.58	1.31 ± 0.23	2.15 ± 0.35	1.24 ± 0.48
Fe	2083 ± 361	2044 ± 359	1690 ± 120	1968 ± 215	1894 ± 337	1967 ± 108	1967 ± 241
Hg	0.32 ± 0.25	0.47 ± 0.32	0.37 ± 0.18	1.56 ± 0.74	1.38 ± 0.27	3.10 ± 0.92	0.61 ± 0.18
Mn	0.23 ± 0.06	0.23 ± 0.08	0.23 ± 0.08	0.30 ± 0.12	0.21 ± 0.09	0.41 ± 0.14	0.30 ± 0.07
Ni[a]	0.04 ± 0.09 (69.2 %)	NQ (100 %)	0.09 ± 0.14 (0 %)	0.05 ± 0.03 (67.6 %)	NQ (96.2 %)	0.11 ± 0.17 (0 %)	0.04 ± 0.04 (70.8 %)
Pb[a]	0.53 ± 0.33	0.47 ± 0.34	0.14 ± 0.09	0.20 ± 0.22	0.02 ± 0.02 (30.8 %)	0.07 ± 0.02	0.19 ± 0.35 (8.3 %)
Se	3.24 ± 1.74	6.48 ± 4.77	9.15 ± 6.18	3.98 ± 1.36	4.61 ± 1.56	6.02 ± 2.56	5.58 ± 2.69
V	NQ (100 %)	NQ (100 %)	NQ (100 %)	NQ (100 %)	NQ (100 %)	NQ (100 %)	NQ (100 %)
Zn	25.9 ± 3.42	28.5 ± 2.60	26.0 ± 3.28	29.4 ± 3.64	28.1 ± 4.11	27.5 ± 4.18	26.1 ± 2.77
Se:Hg	40.4 ± 37.1	44.7 ± 34.3	70.7 ± 50.8	7.8 ± 3.9	9.0 ± 4.3	6.1 ± 5.5	23.6 ± 9.4
N	39	17	16	37	26	15	24

[a] Descriptive statistics for variables, including values below LOQ, were calculated with the Kaplan-Meier method. A proportion of non-quantified samples (NQ) is also reported in brackets for those cases. Variables marked against grey background had a proportion of NQs too high to be used in statistical analyses (higher than 30 %).

Table 3
GLM models best explaining element blood concentrations and body condition (SMI – Scaled Mass Index) in gull chicks studied (according to the AICc values) along the French coast of the English Channel (all three sites combined).

Metal	EHG	GBBG
As[b]	δ^{15}N↑ + site + interactions	δ^{13}C↑ + site + interactions
Co[b]	δ^{34}S↓ + site + interactions	δ^{13}C↑ + site + interactions
Cu	δ^{34}S↑ + site	δ^{34}S↑
Fe	δ^{13}C↑ + site	δ^{13}C↑
Mn	Null	δ^{34}S↑ + site
Se	δ^{34}S↑	δ^{13}C↓ + site + interactions
Pb[b]	δ^{34}N↓ + site + interactions	δ^{15}N + site + interactions
Zn	Null	δ^{13}C↑ + site
SMI[a]	Null	δ^{13}C↑

Grey indicates insignificant model components. EHG: European herring gull; GBBG: Great black-backed gull.

[a] In SMI modelling, PC1 and PC2 were used as potential explanatory variables.

[b] A censored regression was also run for elements with non-quantified cases (As, Co and Pb).

noted in EHG, followed by LBBG and GBBG (Table 1, Fig. 2).

3.2. Comparisons within the Seine Estuary

In the Seine estuary, we examined inter-specific variations at both sampling sites (the city of Le Havre and Ratier Island) and intra-specific variations for each site separately. With the exception of Pb, there was no unanimous trend of model fit across the species. For EHG, models with δ^{13}C fitted poorly to data. Models with δ^{15}N (for As and Pb) and δ^{34}S (for Co, Cu, and Se) were well-fitted (Figs. 3 and 4). For GBBG, models including all the isotopes were best fitted and isotopes were positively correlated with the outcome. For LBBG, δ^{13}C (in models for As, Co, Cu, and Se), δ^{15}N (only for Pb) and null (for Fe, Mn, Zn, and SMI) models were best fitted (Tables 1 and S5, Figs. 5 and 6).

In the Seine estuary, the highest As concentrations were observed in GBBG (higher in the city of Le Havre) followed by LBBG and EHG (higher on Ratier Island). Concentrations of Co were similar between sites and species, albeit a higher fraction of NQs was observed in the city of Le Havre for GBBG. Cd concentrations were below the LOQ in all birds sampled in the Seine estuary except for 3 % of GBBG from the city of Le Havre. A lower fraction of NQs was similarly noted in EHG and GBBG from the city of Le Havre for Cr (10.3 % and 21.6 %, respectively) and Ni (30.8 % and 32.4 %, respectively). For Pb, the highest values were recorded for EHG followed by LBBG and GBBG. Pb concentrations in the city of Le Havre were slightly higher for EHG and noticeably higher for GBBG on Ratier Island (for this species, a fraction of 30.8 % NQs was noted). Concentrations of Se were generally higher in LBBG than in the other two species, except for EHG on Ratier Island (Se levels in EHG and GBBG on Ratier Island were higher than in the city of Le Havre; Table 3).

3.3. Comparisons along the French coast of the English Channel

Models including different isotopes were well-fitted to explain concentrations found in EHG and GBBG (Table 3). In models for GBBG, with two exceptions (Se and Pb), positive relationships with δ^{13}C or δ^{34}S were noted. The null model was the best at explaining Mn and Zn concentrations in EHG. For EHG Pb concentrations were negatively linked to δ^{15}N while for GBBG the influence of δ^{15}N was insignificant (Tables 3 and S6, Fig. 4).

Concentrations of As were higher in GBBG, for which higher values were also noted in the Chausey Islands (Fig. 3). Concentrations of Cd, Co, Cr, Cu, Ni, and Se were similar in both GBBG and EHG but were systematically higher in birds from the Chausey Islands. Lower concentrations of Pb were reported for GBBG with significant variation

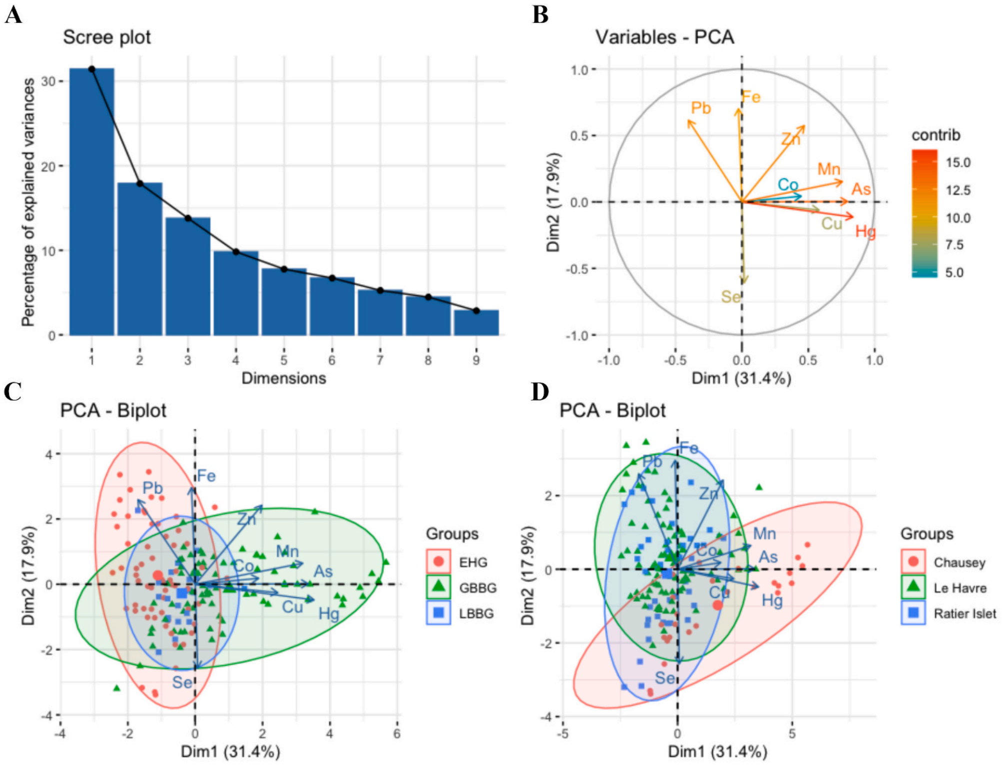

Fig. 2. Principal component analysis (PCA) of the birds studied based on the concentrations of elements in blood. A) Scree plot B) projection of variables with their contribution to the first two components, C)–D) bi-plots with the grouping factors of species and sites.

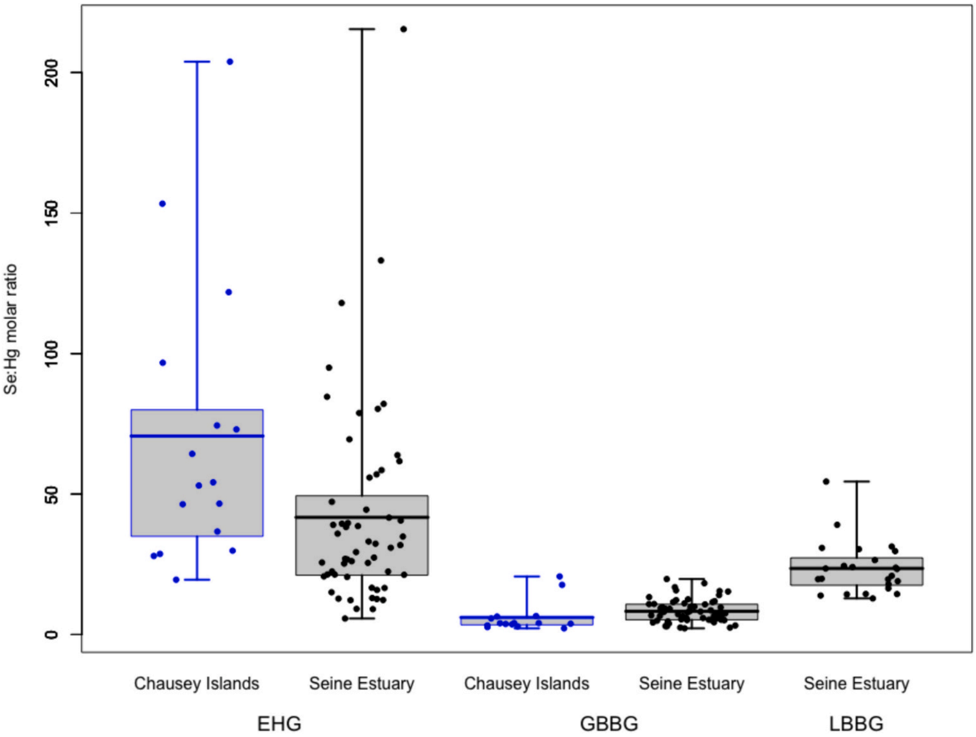

Fig. 3. The Se:Hg molar ratio (mean, SD, min–max) in the blood of specimens studied by species (EHG: European herring gull; GBBG: great black-backed gull; LBBG: lesser black-backed gull) and sites (blue – the Chausey Islands, black - the Seine Estuary (i.e. city of Le Have and Ratier Island combined)).

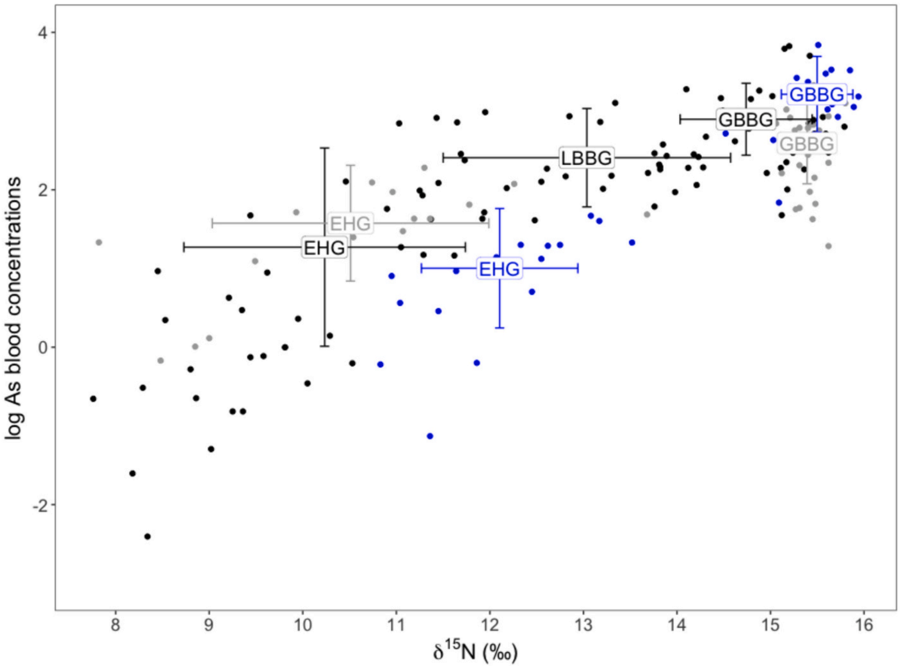

Fig. 4. As concentrations in blood as a function of δ^{15}N values in gull chicks from the French coast of the English Channel (EHG – European herring gull, GBBG – great black-backed gull, LBBG – lesser black-backed gull; blue – the Chausey Islands, black – the city of Le Havre, grey – Ratier Island).

within the Seine Estuary (including 12.7 % of NQs and at the same time reaching a higher mean; Table S6).

4. Discussion

We found that more than one-third of the elements studied had concentrations lower than the limit of quantification. Overall, there was no unanimous scheme of concentrations between species and sites. Variations of the concentrations were undoubtedly element-, species- and site-specific. In many cases, however, trace element concentrations were related to the feeding habitat and to the trophic position of birds (proxied by the relative abundance of C, N and S isotopes). The Se:Hg molar ratio falls below 4 in some specimens of GBBG, suggesting they may be at risk of Hg toxicity. Sex and body condition (calculated by SMI)

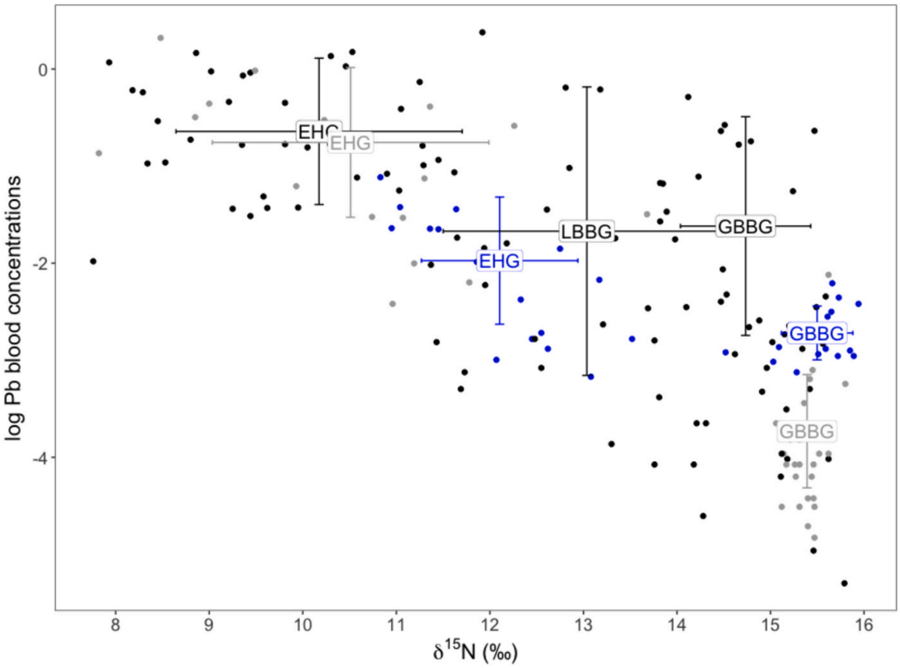

Fig. 5. Pb concentrations in blood as a function of $\delta^{15}N$ values in gull chicks from the French coast of the English Channel (EHG – European herring gull, GBBG – great black-backed gull, LBBG – lesser black-backed gull; blue – the Chausey Islands, black – the city of Le Havre, grey – Ratier Island).

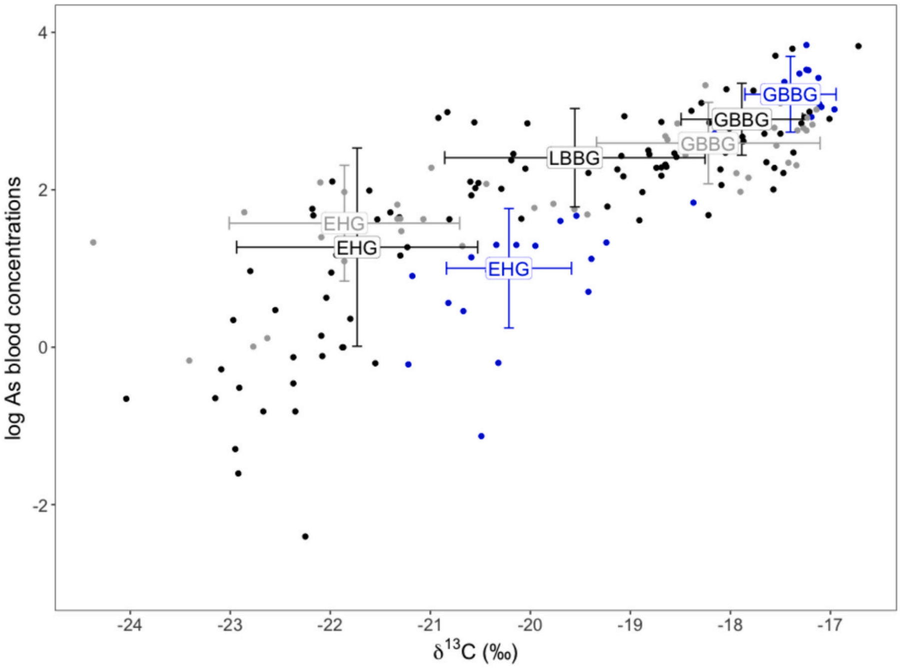

Fig. 6. As concentrations in blood as a function of $\delta^{13}C$ values in gull chicks from the French coast of the English Channel (EHG – European herring gull, GBBG – great black-backed gull, LBBG – lesser black-backed gull; blue – the Chausey Islands, black – the city of Le Havre, grey – Ratier Island).

revealed no relationship with the concentrations of the elements studied.

4.1. *Essential elements (Co, Cu, Fe, Mn, Se, Zn)*

Among the trace elements analysed, Co, Cu, Fe, Mn, Se and Zn are essential for maintaining physiological functions and good health (Nordberg et al., 2007). They often play the roles of enzyme cofactors or catalysts, are components of metalloproteins, stabilising their structure or making them functional, and they may counteract the toxicity of pollutants, including non-essential elements, which promote oxidative stress (Brown and Chatel, 1978; Mertz, 1981; Scheuhammer, 1987; Nordberg et al., 2007; Manceau et al., 2021). Levels of essential elements are therefore regulated by physiological processes (Braune and Simon, 2004), but their accumulation or deficiency may promote alternative pathways that might produce metabolic disorders or even pathologies. Understanding their levels and the factor contributing to their variation is, therefore, a matter of importance in wild animals such as seabirds (dos Santos et al., 2024).

In this study, we failed in numerous cases (i.e., Fe, Mn, and Zn in different species and sites) to fit explaining models, which is probably linked to physiological regulation and lower dependence on external factors (Tables 2 and 3). In the Seine Estuary we found a positive relationship between concentrations of Cu (EHG, GBBG), Mn (GBBG) and Se (GBBG) with δ^{34}S (Table 1). We observed a similar pattern, albeit with lower frequency, in the large-scale approach (Table 2). The increase of δ^{34}S in the blood of birds is linked with a higher proportion of marine prey ingested (Hobson, 1999). This suggests that Cu, Mn and Se concentrations and/or availability are higher in marine habitats. We should note that, despite the fact that some models for essential elements suggested the influence of the sampling site on their concentrations (Tables 2 and 3), the difference was slight, and means were increased by a few individuals (Table 1).

As top predators in aquatic ecosystems, seabirds are particularly exposed to methylmercury (MeHg), which accumulates in their tissues (Manceau et al., 2021). Its higher affinity for Se than for S means that Hg binds to this essential element with great efficiency (Cuvin-Aralar and Furness, 1991; Gailer et al., 2000; Melnick et al., 2010). This causes antagonism, the primary detoxification mechanism of MeHg (contrarily, inorganic Hg is detoxified by metallothionein (Yasutake and Nakamura, 2011)). It depends on the quantity of Se available for association with Hg, which is evaluated by the molar ratio of Se:Hg (Ralston et al., 2008). The ratio is clearly influenced by diet, so it varies among species and according to feeding habitats (Cruz-Flores et al., 2024), and the value >1 is treated as Se-level reducing MeHg toxicity. This value, however, has been questioned and assumed to be underestimated because Se is present in the cells not only in the HgSe complex, except in the liver of very old animals (Manceau et al., 2021). Such a low value may also suggest a reduced Se bioavailability for other biological functions (Gajdosechova et al., 2016). It was recently suggested that the most important compound in reducing MeHg toxicity in seabirds is a four-co-ordinate selenocysteinate complex (Manceau et al., 2021). A minimal safe value of Se:Hg ratio would therefore be 4. In our study, a lower molar Se:Hg ratio was noted only in GBBG for 8 birds from the Seine Estuary and 7 birds from the Chausey Islands (8.6 % of the total birds analysed). Those birds revealed relatively high blood Hg concentrations (mean 3.16 $\mu g \cdot g^{-1}$ dw), which may be the reason for the low Se:Hg ratio. GBBG is the most predatory species (revealed by δ^{15}N) and its higher exposure to Hg than other species has already been confirmed. Additionally, the positive and strong relationship between δ^{15}N and Hg concentrations confirming biomagnification has already been observed among gulls from the English Channel (Binkowski et al., 2021; Lemesle et al., 2024) and other places along the French Atlantic coast (Jouanneau et al., 2022).

4.2. *Other elements (As, Cd, Cr, Ni, Pb)*

Among these elements, As, Cr, and Ni may be considered essential (Nordberg et al., 2007; Kalisińska, 2019), but to the best of our knowledge, there is no information regarding any positive role played by Cd and Pb in vertebrates (and Hg, but that was discussed in Binkowski et al. (2021)).

The concentrations of elements, when compared to other studies, should be evaluated in corresponding age groups because adult birds tend to reveal higher concentrations due to bioaccumulation over time (e.g. Binkowski and Sawicka-Kapusta, 2015; Sebastiano et al., 2017; Carravieri et al., 2020). In our study, we observed visible differences in As concentrations among the species studied. Those concentrations seem to be high, especially in GBBG (3.61–46.45 $\mu g \cdot g^{-1}$ dw), compared to LBBG (1.16–26.48 $\mu g \cdot g^{-1}$ dw) and EHG (<0.16–17.39 $\mu g \cdot g^{-1}$ dw). Lower values were reported in the literature, for example, near the coast of French Guiana, where As blood concentrations did not exceed 4.6 $\mu g \cdot g^{-1}$ dw (Sebastiano et al., 2017), or on the southern Indian Ocean, where concentrations did not exceed 0.81 $\mu g \cdot g^{-1}$ dw (Carravieri et al., 2020). The concentrations in our study were driven by δ^{15}N in EHG and δ^{13}C in GBBG and LBBG (Tables 2 and 3, Figs. 3 and 5), which suggests biomagnification in EHG and habitat trophic dependence in other species (Hobson, 1999). That could be suspected because the biomagnification of As in aquatic food webs (marine and freshwater around the globe) has already been confirmed in the literature but with lower efficiency than for Hg (reviewed by Córdoba-Tovar et al., 2022).

The concentrations of Pb we found were higher, especially in the Seine Estuary, where the city of Le Havre revealed a higher exposure than Ratier Island, than in the majority of species studied in the southern Indian Ocean (Table 3; Carravieri et al., 2020). In the Seine Estuary, the concentrations were negatively driven by δ^{15}N, confirming that Pb does not biomagnify along food webs (Cardwell et al., 2013). The correlation slope even suggests biodilution, which has already been confirmed for marine food webs (Fig. 4; Sun et al., 2020). Concentrations found in the Chausey Islands were lower than in the Seine Estuary, which suggest the impact of industrialisation/urbanisation on the exposure of gulls to Pb (Table S5, Fig. 4). A recent study on yellow-legged gulls from the Portuguese coast also showed higher Pb concentrations in the blood of gulls from the city of Porto than those from Natural colonies (dos Santos et al., 2024). Kucharska et al. (2023) observed no visible impact of industrialisation on Pb concentrations in the chicks of partially waterbird species, such as the black stork. It should be noted that data on trace elements (other than Hg) in the blood of seabird chicks is limited to several geographical locations.

We were unable fully to interpret the concentrations of Cd, Cr, and Ni because most of the values were below LQs. Results suggest that birds in the marine environment (the Chausey Islands) were nevertheless more exposed to all those elements. We may suspect that prey in the Chausey Islands habitat contained higher concentrations than food in the Seine Estuary (Carvalho et al., 2013). Studies on the prey of gulls also support our suspicion. During the breeding season, gulls may feed intensively on cuttlefish which reproduce importantly in this area and their carcasses are abundant since they die immediately after reproduction (Laptikhovsky et al., 2023). Cuttlefish are recognised as an efficient vector of Cd and several other elements in the ecosystem (Bustamante et al., 1998; Miramand et al., 2006). In remote environments such as the Chausey Islands, birds forage mainly on natural food such as cuttlefish, unlike birds in the Seine Estuary, which also consume anthropogenic food containing lower amounts of trace elements.

Our study did not aim to verify the potential interaction between elements experimentally, but it should be mentioned that the additive or synergistic influence of detected elements (especially As, Cd, Hg, and Pb) on seabirds is possible (Lucia et al., 2016).

5. Conclusions

This work contributes to an understanding of marine bird chicks' exposure to trace elements under the potential influence of several factors (sex, feeding habitat and trophic position). The most important results were that As biomagnified, whereas Pb biodiluted along the food web of the English Channel. Greater bioaccumulation of Pb was also observed for those chicks born in the industrialised site of the city of Le Havre. The trace element concentrations found did not influence birds' condition as expressed by the SMI. Five of the 13 elements studied (Ag, Cd, Cr, Ni, and V) had a significant number of unquantified samples due to low concentrations. These data provided some provisional information but allowed only limited inference. Further perspectives should include studies on these species concerning As, Hg, Pb, and Se concentrations, their trophic dependence, and their potential influence on seabird health.

CRediT authorship contribution statement

Lukasz J. Binkowski: Writing – review & editing, Writing – original draft, Visualization, Formal analysis, Conceptualization. **Jérôme Fort:** Writing – review & editing, Validation, Supervision, Investigation, Conceptualization. **Carine Churlaud:** Writing – review & editing, Methodology, Investigation. **Fabrice Gallien:** Writing – review & editing, Investigation. **Gilles Le Guillou:** Writing – review & editing, Investigation. **Paco Bustamante:** Writing – review & editing, Validation, Supervision, Project administration, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgements

The authors are grateful to the GIP Seine Aval for funding the project ECOTONES “Effets de la COnTamination sur les OrgaNismes de l'Estuaire de la Seine” and B. Xuereb as the PI of this project for his support. The permit for the collection of samples was number SRE-UEP-2015/363-051-001. The authors thank C. Ribout from CEBC for processing the molecular sexing, the Plateforme Analyses Élémentaires of LIENSs laboratory for access to their analytical facilities, G. Guillou from the Plateforme Analyses Isotopiques of LIENSs laboratory for running isotope analyses, the CPER (Contrat de Projet Etat-Région) and the FEDER (European Regional Development Fund) for funding the IRMS and ICP devices. L.J. Binkowski's stays at LIENSs were financed by the French Government Scholarship programmes in 2018 and 2021 and by the IDUB Excellent Mobility Programme 2023 at the Pedagogical University of Krakow, Poland. P. Bustamante is an honorary member of the IUF (Institut Universitaire de France). This work benefited from the French GDR “Aquatic Ecotoxicology” framework which aims at fostering stimulating scientific discussions and collaborations for more integrative approaches.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.175891.

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