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Mercury compound distribution and stable isotope composition in the different compartments of seabird eggs: The case of three species breeding in East Greenland

Julie Charrier^{a,*}, Jérôme Fort^a, Emmanuel Tessier^b, Océane Asensio^b, Gaël Guillou^a, David Grémillet^{c,d}, Valère Marsaudon^c, Sophie Gentès^a, David Amouroux^b

^a Littoral Environnement et Sociétés (LIENSs), UMR 7266 CNRS - La Rochelle Université, 2 rue Olympe de Gouges, 17000, La Rochelle, France

^b Université de Pau et des Pays de l'Adour, E2S UPPA - CNRS, Institut des Sciences Analytiques et de Physico-Chimie pour l'Environnement et la Matériaux (IPREM), 2 avenue P. Angot, 64053, Pau, France

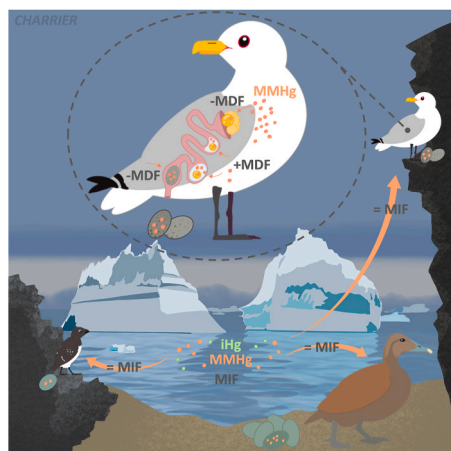
^c Centre d'Ecologie Fonctionnelle & Evolutive (CEFE), UMR 5175 CNRS - Université de Montpellier, 1919 route de Mende, 34293, Montpellier 5, France

^d FitzPatrick Institute of African Ornithology, Department of Biological Sciences, University of Cape Town, University Ave N, Rondebosch, 7701, Cap Town, South Africa

HIGHLIGHTS

- Protein-rich egg compartments exhibited highest Hg contamination, mostly as MMHg.
- MDF values likely reflected Hg organotrophism for protein-rich compartments.
- Odd-isotopes MIF indicated a single origin of MMHg contamination among seabirds.
- Egg membranes/shells are non-invasive indicators of the source of Hg contamination.

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury (Hg) is a toxic contaminant of global concern and the impact on Arctic ecosystems, particularly in seabirds, is critical due to large-scale Hg transport towards polar regions and its biomagnification in marine trophic systems. While the adverse effects of Hg on reproductive processes in seabirds are established, the understanding of Hg maternal transfer pathways and their control on Hg reproductive toxicity is limited. The combination of Hg compounds speciation (inorganic mercury and monomethylmercury MMHg) and Hg stable isotope composition in the different egg compartments (yolk, albumen, membrane, and shell) before embryo development was investigated to provide information on (i) Hg maternal transfer mechanisms, (ii) influence of

* Corresponding author.

E-mail address: julie.charrier@univ-lr.fr (J. Charrier).

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egg biochemical composition on Hg organotropism and (iii) proxies of inputs of Hg contamination. Eggs of three seabird species (the common eider, the black-legged kittiwake and the little auk) collected within the same breeding period (summer 2020) in East Greenland were investigated. For all seabirds, albumen and membrane, the most protein-rich compartments, were the most contaminated (from 1.2 to 2.7 $\mu\text{g g}^{-1}$ for albumen and from 0.3 to 0.7 $\mu\text{g g}^{-1}$ for membrane). In these two compartments, more than 82% of the total Hg amount was in the form of MMHg. Additionally, mass-dependent fractionation values ($\delta^{202}\text{Hg}$) were higher in albumen and membrane in the three species. This result was mainly due the organotropism of MMHg as influenced by the biochemical properties and chemical binding affinity of these proteinous compartments. Among the different egg compartments, individuals and species, mass-independent fractionation values were comparable (mean \pm sd were $0.99 \pm 0.11\text{‰}$, $0.78 \pm 0.11\text{‰}$, $0.03 \pm 0.05\text{‰}$, $0.04 \pm 0.10\text{‰}$ for $\Delta^{199}\text{Hg}$, $\Delta^{201}\text{Hg}$, $\Delta^{200}\text{Hg}$ and $\Delta^{204}\text{Hg}$, respectively). We conclude that initial MMHg accumulated in the three species originated from Arctic environmental reservoirs exhibiting similar and low photodemethylation extent. This result suggests a unique major source of MMHg in those ecosystems, potentially influenced by sea ice cover.

1. Introduction

Mercury (Hg) is a highly toxic contaminant of global concern. Despite international regulations and the adoption of the Minamata Convention on Hg aiming to control and regulate global emissions, between 6000 and 9000 tons of Hg are still emitted annually into the atmosphere, through anthropogenic activities, biomass burning, geogenic degassing, and reemissions from land and ocean (Dastoor et al., 2022; Zhou et al., 2021). Through different processes, these atmospheric emissions are then deposited on the pedosphere, hydrosphere and cryosphere and ultimately contaminate the associated ecosystems. Once released into marine systems, Hg can be methylated via biological pathways and eventually abiotic mechanisms (Tsui et al., 2020), into one of the most toxic form, the monomethylmercury (MMHg). This form bioaccumulates and biomagnifies through marine food webs (Wiener et al., 2002), resulting in highest levels of contamination in long-lived top predators (Dietz et al., 1996; Jæger et al., 2009), such as aquatic birds and marine mammals (Ackerman et al., 2016b; Campbell et al., 2005; Mills et al., 2022). This contamination is responsible for various ecophysiological damages, affecting for instance individual behavior, immune system or reproduction (Chan et al., 2003; Ibañez et al., 2024; Wolfe et al., 1998). In aquatic birds, reprotoxic effects and Hg maternal transfer have been well studied (Ackerman et al., 2020). *In ovo* effects were identified even at low levels of Hg exposure on embryo deformation (Heinz et al., 2009; Heinz and Hoffman, 1998), embryo malposition (Herring et al., 2010) and/or reduced egg hatchability (Braune et al., 2012; Kenow et al., 2011; Varian-Ramos et al., 2014). Post-hatching effects at different development stages are also well documented and include impacts on chick growth (Kenow et al., 2003), chick physiological function, such as their immune system (Kenow et al., 2007) and/or chick survival (Ackerman et al., 2008). Nevertheless, very little is known about the biochemical mechanisms impacting Hg during maternal transfer and egg development. This is unfortunate as these mechanisms could condition Hg toxicity and thus its aforementioned effects. Investigating the dynamics of Hg in the different egg compartments (yolk, albumen, membrane, and shell) prior to embryo development in wild organisms is a complex task. The combined analysis of Hg compounds, including monomethylated Hg (MMHg) and inorganic Hg (iHg), along with the isotope composition of Hg could provide conclusive insights into the biochemical mechanisms underlying maternal transfer during the development of the different egg compartments. These isotopic signatures serve as powerful multi-tracers, as the seven stable isotopes of Hg (^{196}Hg , ^{198}Hg , ^{199}Hg , ^{200}Hg , ^{201}Hg , ^{202}Hg and ^{204}Hg) are subject to various mass-dependent (MDF) and mass-independent (MIF) isotope fractionation processes (see Obrist et al., 2018). MDF is directly proportional to the mass difference between isotopes following kinetic processes or thermodynamic equilibrium in abiotic systems (Kwon et al., 2020). MDF also results from biological processes as those induced by biomagnification (Bouchet et al., 2023; Perrot et al., 2012) and/or metabolic reactions such as accumulation of Hg compounds in specific organs, inter-tissue Hg

transport or Hg detoxification (Masbou et al., 2018; Perrot et al., 2016; Renedo et al., 2021). The MIF of Hg isotopes, reflects a deviation of some isotopic ratio from the theoretical MDF line (Bergquist and Blum, 2007). Odd isotopes MIF (odd-MIF), reported as $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$, is caused by both magnetic isotope and nuclear volume effects. Largest positive odd-MIF signatures result mostly from magnetic isotope effect associated to photochemical demethylation of MMHg in aquatic systems before reaching the food web (Bergquist and Blum, 2007). In addition, $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ ratios can be used to constrain the photochemical extent and pathway Hg undergoes before entering the food web (Bergquist and Blum, 2007; Blum et al., 2014). Even isotopes MIF (even-MIF), reported as $\Delta^{200}\text{Hg}$ and $\Delta^{204}\text{Hg}$, is possibly caused by elementary Hg (Hg^0) oxidation by halogen radicals in the upper atmosphere (Sun et al., 2016). Variations of Even-MIF in surface environments are mainly related to both reactive iHg in precipitation (Demers et al., 2013; Enrico et al., 2016) and the complementary pool of atmospheric Hg^0 (Enrico et al., 2016; Obrist et al., 2017). However, as the magnitude of even-MIF is lowest in biological samples once incorporated into the food web (Lepak et al., 2015; Meng et al., 2020; Motta et al., 2019), this ecological tracer should be interpreted in conjunction with odd-MIF for identifying the source of Hg contamination. Thus, the combination of Hg speciation and stable isotopic composition can provide complementary information to investigate both biological and physico-chemical processes of Hg in the environment and animal tissues, including in seabird eggs (e.g. Point et al., 2011; Day et al., 2012).

Arctic seabirds have been extensively studied in the context of toxicity and conservation (e.g. Fort et al., 2016; Chastel et al., 2022; Cusset et al., 2023), since this region is considered as a sink of Hg (Dastoor et al., 2022). Yet, nitrogen and carbon stable isotopes are proxies for bird trophic status/diet and feeding habitat, respectively (Kelly, 2000). The use of nitrogen, carbon and Hg stable isotopes to describe Arctic seabird source of contamination and biological processes in which this contaminant is involved is only very recent (Day et al., 2012; Point et al., 2011; Renedo et al., 2020). For instance, Hg MDF and odd-MIF values in Alaskan seabird eggs have demonstrated the effectiveness of Hg stable isotopes in distinguishing terrestrial Hg sources from oceanic reservoirs (Day et al., 2012). Another study, using eggs from the same species and regions, reported higher Hg odd-MIF signatures in southern latitudes ($1.13 \pm 0.16\text{‰}$) compared to northern latitudes ($0.53 \pm 0.15\text{‰}$), which are characterized by a greater sea ice coverage, suggesting that the presence of sea ice inhibited Hg marine photochemistry (Point et al., 2011). However, this hypothesis alone may not account for all factors driving MeHg photodemethylation, as demonstrated by the opposing latitudinal trend in odd-MIF values observed in the feathers of little auk compared to the Alaskan seabird eggs (Renedo et al., 2020). Therefore, the recent utilization of stable isotopes has significantly enhanced our global comprehension of the complex relationships between Hg accumulation pathways through food web and environmental parameters shaping these systems. The accumulation of data and the implementation of new studies employing Hg stable isotopes are imperative for a comprehensive understanding of Hg

dynamics within this ecosystem. In the present study, we investigated the speciation and isotopic composition (MDF and MIF) of Hg in egg compartments (yolk, albumen, membrane and shell) in three Arctic seabird species: the common eider (*Somateria mollissima*), the black-legged kittiwake (*Rissa tridactyla*) and the little auk (or dovekie, *Alle alle*). These three seabird species breed in the same area in East Greenland but have distinct ecologies. The common eider primarily feeds on coastal benthic mollusks (Merkel et al., 2006), the black-legged kittiwake on pelagic invertebrates and small fish (Lewis et al., 2001), and the little auk exclusively on zooplankton (Harding et al., 2009). The black-legged kittiwake and little auk, both income breeders, use energy acquired during the breeding season to produce their eggs (Moe et al., 2009), whereas the common eider, a capital breeder, relies on energy reserves accumulated before the breeding season (Sénéchal et al., 2011). The metabolic processes involved in egg production could vary from one species to another, but a general trend in the transfer of Hg from the female to the egg seems to be emerging among species (Ackerman et al., 2016a). We explored the Hg contamination and MDF patterns among egg compartments to elucidate the preferential transfer pathways of Hg to specific compartments, guided by their biogeochemical composition. Furthermore, we compared Hg MIF among egg compartments, individuals, and species which exhibit distinct ecological and reproductive strategies, to try to identify the source of MMHg contamination assimilated by birds via their diet/habitat and transferred from the maternal organism to the developing egg.

2. Material and methods

2.1. Sample collection

Eggs of common eider (n = 6) and black-legged kittiwake (n = 4) were randomly collected at the colonies of Dunholm in East Greenland (69°55'N 22°39'W) from late June to early July 2020, and eggs of little auk (n = 2) were collected at the colony of Ukaleqarteq (Kap Höegh, 70°43'N 21°34'W) in July 2020. Eggs were stored frozen within 24 h after collection and until the four egg compartments (albumen, yolk, membrane, and shell) were split and stored again at -20 °C. Prior to analyses, yolks and albumens were freeze-dried for 72 h and homogenized. Shell and membrane samples were washed in a bath of 2:1 chloroform:methanol solution in an ultrasound bath for 3 min to remove dirt or external contamination. Samples were then rinsed twice in methanol baths, and dried at 45 °C for 48 h, before being homogenized. We declare that the eggs collection was conducted with the permit from the relevant authorities (departmental management for population protection) for the import from Greenland (Denmark) into France of research and diagnostic samples of animal origin.

2.2. Carbon and nitrogen stable isotope analyses

Prior to stable isotope analyses, lipids were extracted from homogenized yolk samples. A mass of approximately 13 g of sample was weighed into a glass container. 4 mL of 2:1 chloroform:methanol solution was added to the sample, then shaken for 10 min. After shaking, the sample was centrifuged for 5 min, and the supernatant was discarded, leaving only the pellet. These steps were run a total of three times. Following the three cycles, the samples were left overnight in an oven at 45 °C. Aliquots of ~0.6–1.0 mg dry weight (dw) of lipid-extracted yolk, albumen, membrane and shell were placed in tin capsules (8 mm × 5 mm, Elemental Microanalysis Ltd, Okehampton, United Kingdom) and analyzed for carbon and nitrogen stable isotope compositions, using a continuous-flow system consisting of an elemental analyzer (Flash 2000, Thermo Scientific, Milan, Italy) equipped with the smart EA option and an autosampler (Zero Blank, Costech, Valencia, CA, United States). The elemental analyzer was connected via a Conflo IV peripheral to a Delta V Plus isotope ratio mass spectrometer (Thermo Scientific, Bremen, Germany). Results are reported in per mil (‰) using the δ notation as

deviation from standards Vienna Pee Dee Belemnite for carbon and atmospheric N₂ for nitrogen, following the formula: $\delta^{13}\text{C}$ or $\delta^{15}\text{N} = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 10^3$, where R is $^{13}\text{C}/^{12}\text{C}$ or $^{15}\text{N}/^{14}\text{N}$, respectively. Normalization was done using USGS61 and USGS63 standard (US Geological Survey, Reston, VA, USA). Each reference material was measured in duplicate at the beginning and the end of each daily group of analysis. The uncertainty of the reported isotope-delta values was evaluated as the standard deviation of the mean of repeated (n = 4) measurements of each reference material within a single group of analyses. Uncertainty did not exceed 0.10‰ for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values. As shell samples contained carbonates known to affect carbon isotopic composition (Jacob et al., 2005), $\delta^{13}\text{C}$ values measured in shells were not reported or considered in the following analyses.

2.3. Total Hg concentration analyses

Total Hg (THg) measurements were performed on subsamples (~0.7 mg for albumen, ~1–2 mg for membrane, and ~15–20 mg for yolk and shell) of homogenized dried eggs compartments, using a direct Hg analyzer by online pyrolysis, gold amalgamation and atomic absorption spectrophotometry (Altec AMA 254). The analysis of each sample was repeated two to three times until the relative standard deviation for two aliquots was <10%. Mean concentration for those two measurements, in $\mu\text{g g}^{-1}$ dw, was then used in statistical analyses. Prior to THg analyses, blanks, and a certified reference material (CRM) were run. Then a CRM was run every 15 samples. The CRM was a lobster hepatopancreas (TORT3 from the National Research Council of Canada) (reference value was $0.29 \pm 0.02 \mu\text{g g}^{-1}$, mean measured value was $0.29 \pm 0.01 \mu\text{g g}^{-1}$).

2.4. Hg speciation analyses

To analyze Hg compounds, membrane (~50 mg dw) and albumen (~50 mg dw) samples were extracted by alkaline digestion using 4 mL tetramethylammonium hydroxide (25% TMAH in H₂O, Sigma Aldrich), while the yolks (~100–250 mg dw) were digested with 3 mL of this reagent. Shells (~250 mg dw) were extracted by acid digestion with 3 mL of nitric acid (HNO₃ 6 N, INSTRA-ANALYZED, J.T.Baker), following the standard operating protocols described elsewhere (Clémens et al., 2011; Renedo et al., 2017). Further information on the protocol are detailed in the supplementary material. For data validation, the same protocol was used for Hg speciation analyses on 3 blanks digested in 4 mL of 25% TMAH, and on two CRMs in 5 mL of 25% TMAH: dogfish liver reference material (DOLT-5, National Research Council of Canada) and tuna fish (ERM-CE-464, from the Institute for Reference Materials and Measurements) (Table S1). Speciation recoveries were validated comparing AMA and GC-ICPMS measurement of THg concentration and the sum of methylmercury and iHg concentrations of egg compartments of common eider, black-legged kittiwake, and little auk (Table S2). From both THg and speciation analyses, the total amount of Hg compounds in the fresh egg compartments was estimated using the total dry mass of each egg compartment sample. Methylmercury was measured during speciation analyses. Nonetheless, methylmercury in the biota is exclusively in different forms of MMHg (Harris et al., 2003) so MMHg will be considered in the following analyses.

2.5. Hg isotopic composition analyses

For Hg stable isotopes analyses, aliquots of ~100 mg dw of albumen, ~500 mg dw of yolk, ~100–200 mg dw of membrane, and ~500–1000 mg dw of shell samples were predigested overnight at room temperature in 3 mL of nitric acid (HNO₃ 69%, INSTRA-ANALYZED, J.T.Baker). Two different extraction protocols were used depending on the type of egg compartment analyzed. Shell and yolk samples were extracted by Hot-block during 12 h at 85 °C. After the addition of 1 mL of hydrogen peroxide (H₂O₂ 30%, OPTIMA, Fisher Chemical), shell and yolk samples were leaved overnight at room temperature before a second round of

extraction by Hotblock during 12 h at 85 °C. Albumen and membrane samples were digested using a high-pressure microwave system (Ultra-WAVE, Milestone, Italy) with internal temperature and pressure control. The general digestion method for food sample was applied (Step 1: 20 min, T1 = 220 °C, T2 = 60 °C, P = 110 bars, Power = 1500 W; Step 2: 10 min, T1 = 220 °C, T2 = 60 °C, P = 110 bars, Power = 1500 W). After the addition of 1 mL of hydrogen peroxide (H₂O₂ 30%, OPTIMA, Fisher Chemical), albumen and membrane samples were left overnight at room temperature before an ultimate step of digestion processed by Hotblock during 12 h at 85 °C. All samples were finally stored at 4 °C until final dilution step and analysis.

For Hg isotopic composition determination, the samples were then diluted at a concentration of 0.5 ppb in an acidic matrix (10% HNO₃ and 2% HCl), following the protocol described elsewhere (Perrot et al., 2012; Renedo et al., 2021). Six stable Hg isotope values (¹⁹⁸Hg, ¹⁹⁹Hg, ²⁰⁰Hg, ²⁰¹Hg, ²⁰²Hg and ²⁰⁴Hg) were reported as a delta notation, calculated relative to the bracketing standard NIST SRM-3133 (see the supplementary material), using a cold-vapor generator multiple-collector inductively coupled plasma mass spectrometer (CVG-MC-ICPMS, Nu Instruments, Wrexham, UK). NIST SRM-997 thallium standard solution was used for the instrumental mass-bias correction using the exponential law. For data validation of Hg isotopic composition analyses, three CRMs were also used: ERM-CE-464 tuna fish (from the IRMM); TORT-3 lobster hepatopancreas and DOLT-5 dogfish liver (from NRCC). A secondary standard NIST RM-8610 (former UM-Almadén) was used for validation of the analytical session. For each egg compartment, between three and four samples were measured as duplicate or triplicate to check the reproducibility. Uncertainty for δ values were calculated using 2SD typical errors from the intern standard NIST RM-8610 and the three CRMs analyzed and mean \pm 2SD values were then compared with published literature (Table S3). As a convention, we have used $\delta^{202}\text{Hg}$ values to describe the MDF (Bergquist and Blum, 2007).

2.6. Statistical analyses

Statistical tests were performed using R version 4.3.2 (R Core Team 2023). Because of low sample sizes, non-parametrical tests were performed. We used a pairwise Wilcoxon rank sum tests corrected by the method of "BH", (see Benjamini and Hochberg, 1995) to compare overall egg compartment $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, $\Delta^{199}\text{Hg}$, $\Delta^{200}\text{Hg}$, $\Delta^{201}\text{Hg}$, $\Delta^{204}\text{Hg}$, $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ ratio mean values within the same species and between species.

3. Results

3.1. Carbon and nitrogen stable isotopes

$\delta^{13}\text{C}$ mean values measured in the different egg compartments ranged from $-21.9 \pm 0.49\text{‰}$ to $-20.6 \pm 0.69\text{‰}$ for common eider, from

$-22.0 \pm 0.15\text{‰}$ to $-20.7 \pm 0.20\text{‰}$ for black-legged kittiwake and from $-23.2 \pm 0.46\text{‰}$ to $-20.4 \pm 0.13\text{‰}$ for little auk. $\delta^{13}\text{C}$ mean values were very similar from one egg compartments to another ($-21.3 \pm 0.44\text{‰}$, $-21.9 \pm 0.49\text{‰}$, $-20.6 \pm 0.69\text{‰}$ for the lipid-extracted yolk, albumen, membrane of common eider's eggs respectively, $-21.4 \pm 0.28\text{‰}$, $-22.0 \pm 0.15\text{‰}$, $-20.7 \pm 0.20\text{‰}$ in black-legged kittiwake and $-21.3 \pm 0.04\text{‰}$, $-23.2 \pm 0.46\text{‰}$, $-20.4 \pm 0.13\text{‰}$ in little auk). The $\delta^{13}\text{C}$ values were not significantly different between species and egg compartments (all p-value >0.05). $\delta^{15}\text{N}$ values were similar between all egg compartments of each species (all p-value >0.05; Table 1). Among lipid-extracted yolk, albumen, membrane and shell samples of all species, $\delta^{15}\text{N}$ values ranged from $10.3 \pm 0.94\text{‰}$ to $11.1 \pm 1.15\text{‰}$ for common eider, from $14.0 \pm 0.18\text{‰}$ to $14.4 \pm 1.69\text{‰}$ for black-legged kittiwake and from $10.7 \pm 0.03\text{‰}$ to $11.5 \pm 0.29\text{‰}$ for little auk. $\delta^{15}\text{N}$ values varied significantly between species, with black-legged kittiwake exhibiting higher values (Table 1).

3.2. Total Hg concentrations and Hg speciation

The highest THg concentrations were measured in the albumen and in the same range for the three bird species ($2.10 \pm 0.44 \mu\text{g g}^{-1}$ dw, $2.04 \pm 0.50 \mu\text{g g}^{-1}$ dw and $1.41 \pm 0.30 \mu\text{g g}^{-1}$ dw in common eider, black-legged kittiwake and little auk, respectively), followed by the egg membrane ($0.50 \pm 0.12 \mu\text{g g}^{-1}$ dw, $0.56 \pm 0.10 \mu\text{g g}^{-1}$ dw and $0.42 \pm 0.05 \mu\text{g g}^{-1}$ dw respectively; Table 2). More than 70% of THg in the total eggs was found in the albumen for all species ($9.36 \pm 2.98 \mu\text{g}$, $5.65 \pm 1.38 \mu\text{g}$, $2.41 \pm 0.80 \mu\text{g}$ for common eider, black-legged kittiwake and little auk, respectively; see Fig. 1). For all the three species, a dominant fraction of Hg was in the form of MMHg in the four egg compartments (~ 90 – 94% in the albumen, ~ 68 – 82% in the yolk, ~ 82 – 89% in the membrane, and ~ 85 – 97% in the shell). As expected, MMHg concentrations were the highest in egg albumen and membrane of common eider ($1.83 \pm 0.45 \mu\text{g g}^{-1}$ dw and $0.43 \pm 0.11 \mu\text{g g}^{-1}$ dw), black-legged kittiwake ($1.74 \pm 0.43 \mu\text{g g}^{-1}$ dw and $0.48 \pm 0.06 \mu\text{g g}^{-1}$ dw), and little auk ($1.08 \pm 0.25 \mu\text{g g}^{-1}$ dw and $0.39 \pm 0.03 \mu\text{g g}^{-1}$ dw, respectively). MMHg concentrations were the lowest in yolk and shell of common eider ($0.05 \pm 0.02 \mu\text{g g}^{-1}$ dw and $0.020 \pm 0.005 \mu\text{g g}^{-1}$ dw), black-legged kittiwake ($0.20 \pm 0.09 \mu\text{g g}^{-1}$ dw and $0.04 \pm 0.01 \mu\text{g g}^{-1}$ dw), and little auk ($0.06 \pm 0.01 \mu\text{g g}^{-1}$ dw and $0.03 \pm 0.01 \mu\text{g g}^{-1}$ dw respectively).

3.3. Hg isotopic composition

3.3.1. Mass-dependent fractionation (MDF)

Statistical differences between egg compartments were found only in common eider, with albumen and membrane samples exhibiting significantly higher MDF values than yolks and shells (all p-values <0.05). $\delta^{202}\text{Hg}$ values were ranging from $0.08 \pm 0.25\text{‰}$ to $0.78 \pm 0.07\text{‰}$ for common eider, from $0.39 \pm 0.19\text{‰}$ to $0.98 \pm 0.05\text{‰}$ for

Table 1

Carbon ($\delta^{13}\text{C}$, proxy of foraging habitat) and nitrogen ($\delta^{15}\text{N}$, proxy of trophic status) isotopic values (Mean \pm SD, ‰) of egg compartments of common eider, black-legged kittiwake, and little auk.

Species	Compartment	n	$\delta^{13}\text{C}$ (‰)	%C	$\delta^{15}\text{N}$ (‰)	%N
Common eider	Albumen	6	-21.94 ± 0.49^a	46.52 ± 0.88^a	10.50 ± 1.11^b	12.77 ± 0.16^b
	Yolk		-21.29 ± 0.44^a	43.95 ± 0.47^c	11.35 ± 0.93^b	13.45 ± 0.13^d
	Membrane		-20.62 ± 0.69^a	44.31 ± 0.66^c	11.08 ± 1.15^b	13.85 ± 0.38^d
	Shell		–	–	10.83 ± 1.22^b	0.63 ± 0.15^c
Black-legged kittiwake	Albumen	4	-22.00 ± 0.15^a	45.49 ± 1.98^{abc}	14.33 ± 0.07^a	12.10 ± 0.33^a
	Yolk		-21.40 ± 0.28^a	42.88 ± 0.98^{bc}	15.08 ± 0.15^a	13.03 ± 0.36^{ab}
	Membrane		-20.66 ± 0.20^a	42.96 ± 1.07^{bc}	14.44 ± 1.69^a	12.86 ± 0.23^{ab}
	Shell		–	–	14.01 ± 0.18^a	0.98 ± 0.03^a
Little auk	Albumen	2	-23.21 ± 0.46^a	48.02 ± 0.38^{abcd}	11.06 ± 0.26^{ab}	9.84 ± 0.69^{abcd}
	Yolk		-21.31 ± 0.04^a	40.88 ± 1.57^{abcd}	12.12 ± 0.07^{ab}	12.49 ± 0.51^{abcd}
	Membrane		-20.43 ± 0.13^a	41.56 ± 0.09^{abcd}	11.54 ± 0.29^{ab}	12.51 ± 0.18^{abcd}
	Shell		–	–	10.81 ± 0.36^{ab}	1.21 ± 0.00^{abcd}

n is the number of samples. Values not sharing the same superscript letter within each group are statistically different.

Table 2

Total Hg concentrations (Mean ± SD, $\mu\text{g}\cdot\text{g}^{-1}$ dw) and Hg stable isotopes ratios (mean ± sd, %) of egg compartments of common eider, black-legged kittiwake, and little auk.

Species	Compartment	n	THg ($\mu\text{g}\cdot\text{g}^{-1}$)	$\delta^{202}\text{Hg}$ (‰)	$\Delta^{199}\text{Hg}$ (‰)	$\Delta^{200}\text{Hg}$ (‰)	$\Delta^{201}\text{Hg}$ (‰)	$\Delta^{204}\text{Hg}$ (‰)	$\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ ratio
Common eider	Albumen	6	2.10 ± 0.44 ^b	0.78 ± 0.07 ^c	1.03 ± 0.11 ^a	0.02 ± 0.05 ^a	0.82 ± 0.11 ^a	0.03 ± 0.09 ^a	1.26 ± 0.07 ^a
	Yolk		0.08 ± 0.02 ^d	0.08 ± 0.25 ^d	1.01 ± 0.10 ^a	0.02 ± 0.05 ^a	0.78 ± 0.07 ^a	0.10 ± 0.10 ^a	1.30 ± 0.04 ^a
	Membrane		0.50 ± 0.12 ^e	0.68 ± 0.09 ^{bc}	1.07 ± 0.13 ^a	0.04 ± 0.02 ^a	0.80 ± 0.09 ^a	-0.06 ± 0.06 ^a	1.34 ± 0.08 ^a
	Shell		0.02 ± 0.01 ^c	0.12 ± 0.18 ^d	1.02 ± 0.09 ^a	0.03 ± 0.05 ^a	0.79 ± 0.08 ^a	0.07 ± 0.11 ^a	1.30 ± 0.11 ^a
Black-legged kittiwake	Albumen	4	2.04 ± 0.50 ^{ab}	0.98 ± 0.30 ^{abc}	0.95 ± 0.10 ^a	0.07 ± 0.09 ^a	0.74 ± 0.09 ^a	0.00 ± 0.10 ^a	1.30 ± 0.03 ^a
	Yolk		0.28 ± 0.07 ^{ae}	0.39 ± 0.19 ^{abc}	0.90 ± 0.09 ^a	-0.01 ± 0.03 ^a	0.72 ± 0.04 ^a	0.02 ± 0.02 ^a	1.25 ± 0.07 ^a
	Membrane		0.56 ± 0.10 ^{ae}	0.98 ± 0.05 ^a	0.89 ± 0.05 ^a	0.06 ± 0.02 ^a	0.72 ± 0.08 ^a	-0.03 ± 0.08 ^a	1.26 ± 0.10 ^a
	Shell		0.04 ± 0.02 ^{acd}	0.74 ± 0.18 ^{abc}	0.90 ± 0.12 ^a	-0.01 ± 0.04 ^a	0.66 ± 0.07 ^a	0.15 ± 0.12 ^a	1.37 ± 0.04 ^a
Little auk	Albumen	2	1.41 ± 0.30 ^{abcde}	0.07 ± 0.28 ^{abcd}	1.10 ± 0.11 ^a	-0.01 ± 0.02 ^a	0.80 ± 0.04 ^a	0.07 ± 0.11 ^a	1.37 ± 0.11 ^a
	Yolk		0.09 ± 0.01 ^{abcde}	0.03 ± 0.13 ^{abcd}	1.01 ± 0.05 ^a	0.03 ± 0.02 ^a	0.81 ± 0.04 ^a	0.10 ± 0.08 ^a	1.25 ± 0.01 ^a
	Membrane		0.42 ± 0.05 ^{abcde}	0.29 ± 0.09 ^{abcd}	1.00 ± 0.06 ^a	0.05 ± 0.00 ^a	0.82 ± 0.01 ^a	-0.02 ± 0.06 ^a	1.23 ± 0.09 ^a
	Shell		0.04 ± 0.01 ^{abcde}	0.02 ± 0.14 ^{abcd}	0.95 ± 0.01 ^a	0.02 ± 0.00 ^a	1.12 ± 0.08 ^a	0.01 ± 0.04 ^a	0.86 ± 0.06 ^a

n is the number of samples. Values not sharing the same superscript letter within each group are statistically different.

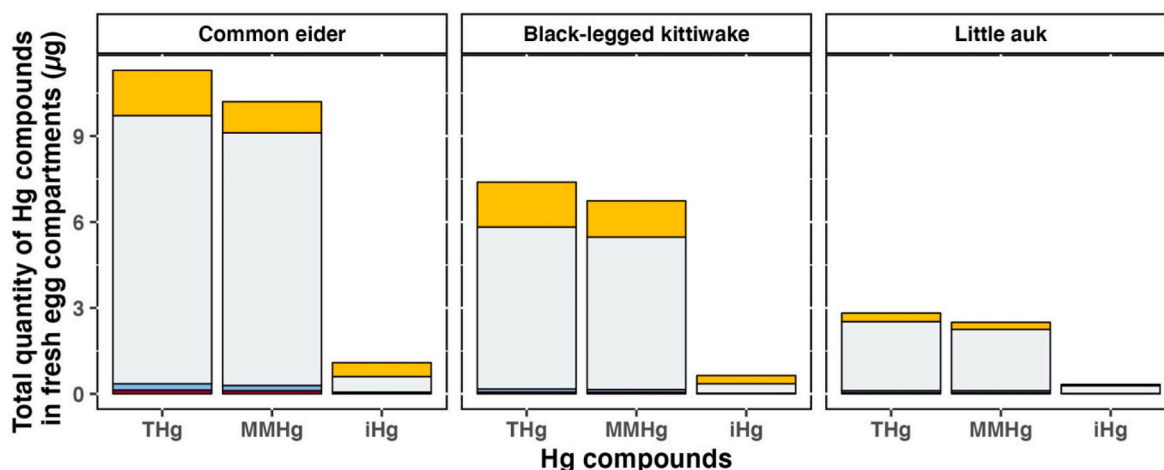


Fig. 1. Mean quantity in μg of total Hg (THg) monomethylated Hg (MMHg) and inorganic Hg (iHg) in fresh total egg compartments (white: albumen, orange: yolk, blue: membrane, and red: shell) of common eider, black-legged kittiwake, and little auk.

black-legged kittiwake and from $0.02 \pm 0.14\%$ to $0.29 \pm 0.09\%$ for little auk; Table 2). Egg compartments with the heaviest $\delta^{202}\text{Hg}$ isotopic composition (i.e. albumen and membrane) were also those with higher concentrations of THg (Fig. S1), and ultimately of MMHg (Table 2), a similar pattern between individuals of the same species, but slightly different from one species to another (Fig. 2).

3.3.2. Mass-independent fractionation (MIF)

The Odd-MIF values ($\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$) were similar from one egg compartment to another within the same species and between species (all p-values >0.05). All samples combined, the mean $\Delta^{199}\text{Hg}$ and

$\Delta^{201}\text{Hg}$ values were $0.99 \pm 0.11\%$ and $0.78 \pm 0.11\%$, respectively. The $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ ratios were similar from one compartment to another within the same species and between species (all p-values >0.05; Table 2; Fig. 3). The Even-MIF signatures ($\Delta^{200}\text{Hg}$ and $\Delta^{204}\text{Hg}$) were also similar among all the samples (all p-values >0.05; Fig. S2). All samples combined, the mean $\Delta^{200}\text{Hg}$ and $\Delta^{204}\text{Hg}$ values were $0.03 \pm 0.05\%$ and $0.04 \pm 0.10\%$, respectively (Table 2).

4. Discussion

The Hg contamination of ecosystems is a persistent global

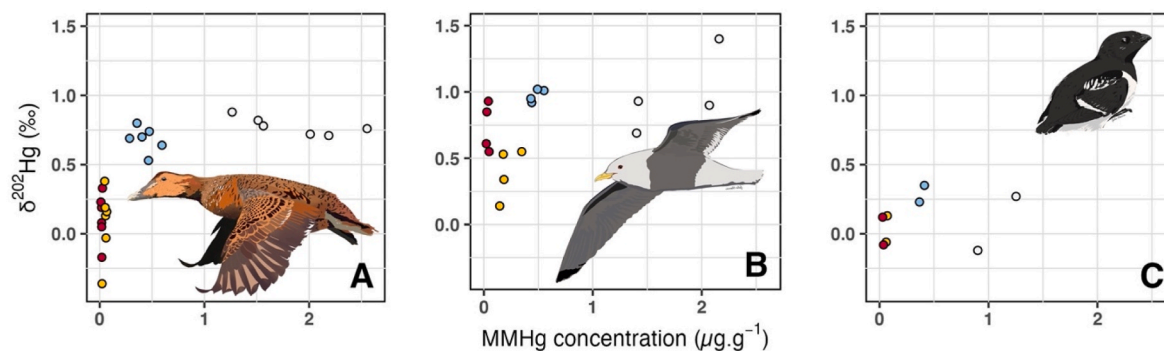


Fig. 2. Hg MDF versus MMHg concentration values ($\delta^{202}\text{Hg}$ in ‰ versus MMHg in $\mu\text{g}\cdot\text{g}^{-1}$ dw) of egg compartments (white: albumen, orange: yolk, blue: membrane, and red: shell) of common eider (A), black-legged kittiwake (B) and little auk (C).

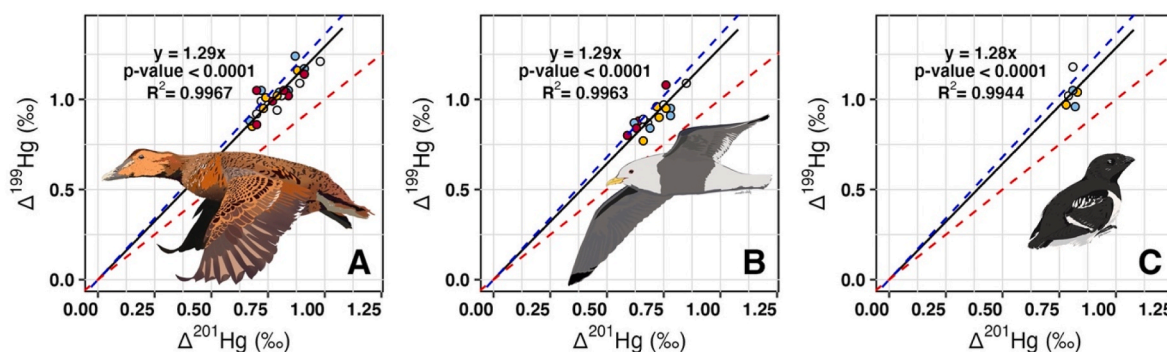


Fig. 3. $\Delta^{199}\text{Hg}$ versus $\Delta^{201}\text{Hg}$ relationship observed in all egg compartments (white: albumen, orange: yolk, blue: membrane, and red: shell) from the three species. Overall slopes are 1.30 ± 0.08 ($R^2 = 0.997$, $p\text{-value} < 0.001$), 1.29 ± 0.08 ($R^2 = 0.963$, $p\text{-value} < 0.001$) and 1.28 ± 0.09 ($R^2 = 0.994$, $p\text{-value} < 0.001$) for common eider (A), black-legged kittiwake (B), and little auk (C) (shell removed), respectively. The blue dashed line represents the theoretical $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ slope of 1.36 ± 0.04 for MMHg photodemethylation in the water column and the red dashed line represents the theoretical $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ slope of 1.00 ± 0.01 expected for photoreduction of inorganic Hg in the water column (Bergquist and Blum, 2007).

environmental problem. Despite efforts to control emissions, substantial quantities of Hg are released in marine ecosystems and biomagnify along the food chains, making marine top predators the most exposed species. This contamination has well-documented ecophysiological effects, particularly in long-lived predators such as seabirds (Wiener et al., 2002). Although the effects on reproduction have been relatively well studied in seabirds (Heinz et al., 2009; Ibañez et al., 2024), there is a significant gap in our understanding of the biochemical mechanisms involved in maternal transfer of Hg during egg development. To the best of our knowledge, this study is the first to investigate the stable isotopes composition and speciation of Hg in the four egg compartments of seabirds breeding in the same area. The objective was to enhance our understanding of the biological and physicochemical processes undergone by Hg during maternal transfer and photochemical reactions before reaching the food web.

4.1. Inter-species comparison of THg contamination in egg compartments: insights into Hg maternal transfer mechanisms and maternal diet influence

Eggs of little auk, common eider and black-legged kittiwake showed different $\delta^{15}\text{N}$ values, reflecting the trophic position of females. Little auk feed almost exclusively on zooplankton (Harding et al., 2009), and common eider mainly on coastal mollusks (Merkel et al., 2006), and so had lower $\delta^{15}\text{N}$ values than black-legged kittiwake feeding at higher trophic level on a variable combination of invertebrates and small energy-rich schooling fish (Lewis et al., 2001). However, Hg concentrations in eggs, supposed to be related to the species trophic status following biomagnification processes (Akearok et al., 2010) were of the same order of magnitude between species. Levels of Hg in eggs is therefore not solely related to the trophic status of the female. This result is consistent with previous studies that suggested differences between species in maternal Hg intakes (prey items less contaminated in Hg for black-legged kittiwake despite a higher trophic position) and/or in depuration rates and/or in metabolism with increased *in vivo* demethylation capabilities leading to Hg excretion (Provencher et al., 2014). These differences subsequently influence the proportion of MMHg transferred from the female to the eggs (Ackerman et al., 2020). Despite similar Hg concentrations, common eider eggs were those containing the highest amount of Hg, and eggs of black-legged kittiwake had three times more Hg than those of little auk. The different compartments of an egg are synthesized successively in the female genital tract in the order from yolk, albumen, membrane and shell (Nys and Guyot, 2011). They could therefore incorporate different amounts of MMHg depending on their biogeochemical composition. In the three studied species, the albumen was by far the most contaminated compartment, concentrating most of the measured THg, mainly in the form of MMHg, which is

consistent with the results of previous studies (Ackerman et al., 2013; Kenamer et al., 2005). Similarly to total Hg contamination measured in eggs, this prevalent MMHg contamination in the albumen is only partially correlated with the trophic level of the associated species. Maternal transfer of MMHg into eggs therefore appears to be dependent on metabolic processes as mentioned above, involved in energy allocation strategy of female, which can vary significantly from one species to another.

4.2. Hg MDF in egg compartments as influenced by both egg biochemical composition and seabird physiology

The biochemical composition of the four egg compartments could slightly vary in each species, subsequently influencing Hg transfer. Nevertheless, the trend was similar between common eider, black-legged kittiwake, and little auk, as the general egg structure is the same for all bird species. Proteins represent about 90% of the dry albumen, mainly composed of glycoproteins, and the egg membrane is a full protein compartment (Nys and Guyot, 2011). Most of these proteins contain amino acids with sulfhydryl groups (-SH) (Guha et al., 2018), such as cysteine residues, for which MMHg has a very high affinity forming a stable covalent bond (R-S-Hg-CH₃) (Ajsuvakova et al., 2020). As a consequence, and as previously mentioned, the albumen and membrane were the compartments with the highest concentration of THg, mainly in the form of MMHg. The MDF pattern (measured as $\delta^{202}\text{Hg}$) between the different egg compartments was similar between eggs of each species, with higher values measured in albumen and membrane samples. This MDF could therefore result from the high chemical affinity of MMHg to thiol groups present in protein-rich egg compartments. Their specific biochemical composition can lead to the bioaccumulation of MMHg via stable bonds resulting in MDF of Hg stable isotopes and heavier isotopic composition values in albumen and membrane (higher $\delta^{202}\text{Hg}$) (Bergquist and Blum, 2007; Jackson et al., 2008; Perrot et al., 2010), compared with the yolk and the shell. Such MMHg bioaccumulation is closely linked to the process of Hg maternal transfer from the female to its offspring. Notably, the transfer of proteins and lipids, synthesized in the female liver and essential for the egg development, plays a key role (Sturkie, 1986). During this transfer, they are transported by the blood in the circulatory system from the liver to the genital tract, to finally becoming incorporated into the egg during its formation. Therefore, higher MDF values could be induced by the binding of MMHg to cysteine groups (i.e. thiol/thiolate) from these proteins during the bloodstream transport from one tissue to another, as already described to explain the depuration of isotopically heavier MMHg enriched in $\delta^{202}\text{Hg}$ in seabird feathers (Renedo et al., 2021).

Among the three different species, the higher MDF values in albumen

of common eider and black-legged kittiwake eggs could be related to both trophic levels and Hg detoxification mechanisms of each species. Given that MMHg detoxification mechanisms are suggested to preferentially involve the lightest Hg stable isotopes, the resulting pool of non-detoxified MMHg excreted in eggs is likely to exhibit higher MDF values. Alternatively, seabirds at lower trophic levels, such as the little auk, are exposed to lower levels of MMHg intake. This lower THg exposure could lead to a decreased need for detoxification mechanisms, shrinking the residual pool of non-detoxified MMHg excreted in little auk eggs, therefore inducing lower MDF values. In the case of the black-legged kittiwake, the level of egg THg concentrations did not reflect bird's trophic status since they were comparable to eiders contamination which feed at lower trophic levels. However, MDF values in protein-rich egg compartments were higher in black-legged kittiwake eggs than in common eider. The MDF values in proteinic compartments could therefore be affected by residual MMHg excretion from the female to the egg enriched in heavier isotopes, after *in vivo* hepatic demethylation (Bond and Diamond, 2009). This result suggests that Hg MDF in eggs could be used as an indicator of the efficiency of MMHg demethylation in female seabirds as previously suggested (Perrot et al., 2016).

But another hypothesis is that the energy allocation strategy for egg production also influenced MDF, with egg MDF reflecting MMHg stored in internal tissues prior to egg formation (capital strategy) or reflecting the MMHg recently acquired during the egg formation (income strategy). The three species studied here show distinct strategies, ranging from the common eider (more capital breeder; Sénéchal et al., 2011) to the little auk (more income breeder; Moe et al., 2009). These energy allocation strategies could elucidate some of the observed differences between species, particularly in relation to detoxification mechanisms. Specifically, species that rely more on capital breeding may transmit a residual pool of non-detoxified MMHg to their eggs, resulting in higher MDF values compared to species that primarily rely on income breeding. This suggests that in income-breeding species, maternal transfer processes are more direct, allowing less time for detoxification mechanisms to take effect. We therefore propose that the processes leading to MDF of Hg isotopes in the different compartments of seabird eggs are directly linked to Hg maternal transfer pathways, such as (1) the organotropism of MMHg for the biochemical proteinous properties and chemical binding affinity of the albumen and membrane, (2) the dietary MMHg intake and demethylation extent of MMHg in female organs and (3) the seabird species-dependent energy allocation strategy for reproduction.

4.3. Hg MIF indicates a similar source of trophic MMHg contamination for the three seabird species

Common eider, black-legged kittiwake and little auk were breeding in the same geographical area in East Greenland and all presented positive MIF values for the Hg odd isotopes (i.e. $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$). We found no significant differences in odd-MIF and even-MIF values isotopes between egg compartments, individuals and species. After Hg assimilation by primary producers, MIF isotopic signatures are preserved along the food chain (Kwon et al., 2012; Perrot et al., 2012), making stable isotopes a remarkable tool to characterize the sources of MMHg contamination in seabirds. Among all samples, the mean $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ ratios were close to the slope of 1.36 ± 0.02 obtained after MMHg photochemical demethylation in model freshwaters (Bergquist and Blum, 2007). The overall similarity in Hg MIF among seabird species therefore suggests that MMHg origin in the studied system was unexpectedly similar and independent of the foraging strategies, trophic levels and/or habitats. Our results demonstrate that the majority of THg in eggs was residual MMHg with an enrichment in ^{199}Hg and ^{201}Hg isotopes induced by *in situ* photodemethylation reaction. In addition, $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$ extents were rather low ($0.99 \pm 0.11\%$ and $0.78 \pm 0.11\%$, respectively), suggesting limited MMHg photochemical degradation in this Arctic ecosystems (Masbou et al., 2015; Point et al., 2011). These MIF results in Arctic seabird eggs are even

lower than the odd-MIF values observed in deep foraging fish species of the North Pacific Ocean were MMHg photochemical demethylation is also hampered ($1.5 \pm 0.3\%$), compared with fish predating in the photic zone ($3.1 \pm 1.1\%$) (Blum et al., 2013). The main limitations for photochemical demethylation in the water column results from the solar radiation intensity, the attenuation of light depth penetration, low dissolved organic matter content (Bouchet et al., 2023), particle driven turbidity, and especially in Arctic, sea ice cover (Masbou et al., 2015, 2018; Point et al., 2011). Turbidity of the surface waters is low in the central part of the Scoresby Sound (study region) and decreases further as the water mass moves outside of the fjord, towards the studied seabird colonies (Seifert et al., 2019). Given the prevalence of sea ice at the latitudes of this site, we can assume that the sea ice cover usually plays role in preventing light penetration, especially since sea ice is known to impact solar irradiance in the surface water both vertically and horizontally (Frey et al., 2011), probably preventing the UV-visible light penetration. Additionally, a previous study on Alaskan seabird eggs reported higher Hg odd-MIF signatures in southern latitudes ($1.13 \pm 0.16\%$) compared to northern latitudes ($0.53 \pm 0.15\%$), which are characterized by a greater sea ice coverage, suggesting that the presence of sea ice inhibited Hg marine photochemistry (Point et al., 2011). The Alaskan northern latitudes reported by the study mentioned above (68°N) are comparable to the Greenlandic latitudes studied here ($69\text{--}70^\circ\text{N}$), aligning with the hypothesis of sea ice preventing Hg photochemistry. This pattern of $\Delta^{199}\text{Hg}$ values decreasing with latitudes was also documented in Arctic marine mammal livers (Masbou et al., 2018; Point et al., 2011). This raises the question of the role played by sea ice in shaping the Hg contamination of Arctic organisms, through the Hg biogeochemical cycle. However, sea ice is unlikely to be the only factor regulating Hg photochemistry in the Arctic, as an inverse trend has been observed with odd-MIF increasing with latitude in East Greenland on little auk feathers (Renedo et al., 2020). More generally, it is known that the primary Hg inputs into Arctic ocean surface are wet and dry atmospheric Hg deposition (Jiskra et al., 2021; Zhang et al., 2014), and river inputs (Amos et al., 2014). In that respect, zero to low $\Delta^{199}\text{Hg}$ values could additionally indicate terrestrial Hg (Obriest et al., 2017), due to riverine inputs for instance as it has been suggested for low $\Delta^{199}\text{Hg}$ values ($0.60 \pm 0.04\%$) in Arctic seabird eggs (Day et al., 2012). However, odd-MIF and MDF measured in East Greenland seabird eggs (present study) were not as low as found in this specific case, suggesting that egg MMHg was more likely issued from an oceanic reservoir. Similarly to $\Delta^{199}\text{Hg}$ values, $\Delta^{200}\text{Hg}$ signatures are also known to decrease significantly with latitude exhibiting near-zero values in the northern latitude (Masbou et al., 2018; Jiskra et al., 2021), which is also observed in this study ($0.03 \pm 0.05\%$).

We therefore suggest that the source of Hg in eggs of common eider, black-legged kittiwake and little auk was MMHg from oceanic reservoirs, which has initially been prevented from being photochemically fractionated, possibly influenced by sea ice cover. The presence of sea ice could both harbor a microbial source of MMHg to surface water and trigger increased microalgal biomass, promoting increasing MMHg levels for food web uptake (Yue et al., 2023), which could biomagnify up to top-predators through sympagic (i.e. ice-associated) food webs. The consistent MIF between species with very different ecologies, in this study, suggests that Hg contamination in this relatively wide area of East Greenland comes from a same single source. The results of this study also demonstrate that eggs can be collected non-invasively to trace Hg source of contamination by sampling only membrane and shell after chicks hatched. Further studies on the biogeochemical processes of Hg interacting with sea ice are crucial to better understand Hg contamination of marine predators and Arctic ecosystems, while research on Hg stable isotopes in Arctic biota is essential for identifying contamination sources and understanding the broader implications for wildlife and human communities.

5. Conclusion

In this study, we investigated Hg contamination and MDF patterns among different compartments of seabird eggs (yolk, albumen, membrane, and shell) to elucidate preferential Hg maternal transfer pathways. We also compared Hg MIF across egg compartments, individuals, and species with distinct ecological and reproductive strategies to identify the sources of MMHg contamination in an area of East Greenland. Our major findings showed that (1) The MMHg contamination in albumen is only partially correlated with the trophic level of the species, indicating that maternal MMHg transfer is influenced by species-specific metabolic processes and energy allocation strategies; (2) Hg stable isotope MDF in seabird egg compartments is linked to Hg maternal transfer pathways, including MMHg organotropism for albumen and membrane biochemistry, dietary MMHg intake, MMHg demethylation in female organs, and species-dependent energy allocation strategies; and (3) The consistent MIF among species suggests that MMHg contamination in East Greenland seabirds originated from a same oceanic source, potentially influenced by sea ice cover and related microbial activity and microalgal biomass. These findings contribute to our understanding of Hg biogeochemical processes in Arctic ecosystems and have broader implications for wildlife and human communities.

CRedit authorship contribution statement

Julie Charrier: Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Conceptualization. **Jérôme Fort:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Data curation, Conceptualization. **Emmanuel Tessier:** Writing – review & editing, Validation, Methodology. **Océane Asensio:** Writing – review & editing, Validation, Methodology. **Gaël Guillou:** Writing – review & editing, Methodology. **David Grémillet:** Writing – review & editing, Project administration, Funding acquisition. **Valère Marsaudon:** Writing – review & editing, Data curation. **Sophie Gentès:** Writing – review & editing, Data curation. **David Amouroux:** Writing – review & editing, Validation, Supervision, Methodology, Investigation, 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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2024.142857>.

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