The consumption of ice-derived resources is associated with higher mercury contamination in an Arctic seabird

Fanny Cusset, Julie Charrier, Guillaume Massé, Mark Mallory, Birgit Braune, Jennifer Provencher, Gaël Guillou, Philippe Massicotte, Jérôme Fort

The consumption of ice-derived resources is associated with higher mercury contamination in an Arctic seabird

Fanny Cusset[1,2,a*], Julie Charrier[2,a*], Guillaume Massé[1,3], Mark Mallory[4], Birgit Braune[5], Jennifer Provencher[5], Gaël Guillou[2], Philippe Massicotte[1] and Jérôme Fort[2]

[1] Takuvik International Research Laboratory (IRL 3376) ULaval-CNRS, Biolgy Department, Laval University, 1045 Avenue de la Médecine, Québec, QC, Canada G1V 0A6

[2] LIENSs, UMR 7266, CNRS-La Rochelle Université, 2 Rue Olympe de Gouges, 17000 La Rochelle, France

[3] LOCEAN, UMR 7159, CNRS, MNHN, IRD, Sorbonne-Université, Station Marine de Concarneau, BP225, 29900, Concarneau, France

[4] Biology Department, Acadia University, 15 University Avenue, Wolfville, NS, B4P 2R6, Canada

[5] Environment and Climate Change Canada, National Wildlife Research Centre, Carleton University, Raven Road, Ottawa, ON, K1A 0H3, Canada

* Co-first author

[a]Corresponding author: Fanny Cusset (cussetfanny@gmail.com), Julie Charrier (julie.charrier@univ-lr.fr)

ABSTRACT

Sea ice plays a fundamental role in Arctic marine environments, by driving primary productivity and sustaining ice-associated ecosystems. Simultaneously, sea ice influences the contamination of Arctic marine organisms, by modifying contaminant cycles or their bioavailability. Changes in sea ice conditions could therefore profoundly impact the functioning of Arctic marine food webs and their contamination. Top predators such as seabirds, which are subject to bioaccumulation and biomagnification of contaminants, are particularly exposed. In this context, the present study aims to investigate the influence of sea ice and of the use of ice-derived resources on the contamination of seabirds by mercury (Hg). To this end, eggs of thick-billed murres (Brünnich's guillemots, *Uria lomvia*; $n = 60$) were collected on Prince Leopold Island (Canadian High Arctic) during four years of varying ice conditions (2010–2013). Trophic tracers (*i.e.,* Highly Branched Isoprenoids, HBIs – an indicator of the use of ice-derived resources; carbon and nitrogen stable isotopes – indicators of foraging habitats and trophic status), as well as total Hg concentrations were quantified. Results showed that feeding on ice-derived resources (as indicated by HBI concentrations) was positively correlated to sea ice cover, and both positively influenced Hg concentrations in murre eggs. However, when testing for the best predictor with model selection, sea ice concentration only drove Hg contamination in murres. This work provides new insights into the role of sea ice and ice-derived resources in the contamination by Hg of Arctic wildlife. Further research is now needed to better understand the relationship between sea ice and Hg contamination in Arctic biota and its underlying mechanisms, but also to identify Hg sources in rapidly changing environmental conditions in the Arctic.

KEYWORDS

Brünnich's guillemot – Hg – Highly Branched Isoprenoids – Sea ice – Sympagic food web – Thick-billed murre

FUNDING SOURCES

Funding for sample collections was provided by Environment and Climate Change Canada, the Northern Contaminants Program of Crown-Indigenous Relations and Northern Affairs Canada and Acadia University. FC benefited from scholarships from Natural Sciences and Engineering Research Council (Discovery grant to GM). JC is funded by the project ARCTIC-STRESSORS (French National Research Agency; ANR- 20- CE34- 0006 to JF). The preparation of this paper was supported by the Sentinel North program of Laval University, made possible, in part thanks to funding from the Canada First Research Excellence Fund.

1. INTRODUCTION

Sea ice plays a fundamental role in shaping Arctic ecosystems, by driving the phenology of marine primary production. During spring, melting sea-ice drives several consecutive blooms of primary producers. In early spring, while ocean surface waters are still covered by ice, a bloom of ice algae represents a major contributor to the total primary production. This can represent up to 57% of the entire primary production in the water column and sea ice in the central Arctic (Gosselin et al., 1997), which constitutes the first levels of sympagic (*i.e.,* ice-associated) food webs. Later, a bloom of pelagic phytoplankton occurs in ice-free water, supporting pelagic food webs (Horner and Schrader, 1982; Tremblay et al., 2012). These two consecutive blooms and their phenology are crucial for secondary production and the functioning of entire marine ecosystems in the Arctic, highlighting the potential impacts of melting sea ice under the influence of global warming (Wassmann and Reigstad, 2011).

In addition, sea ice plays a central role in the biogeochemical cycle of chemical contaminants, in their bioavailability to marine wildlife, and thus in the contamination of Arctic marine ecosystems. This is the case for mercury (Hg), a non-essential and toxic metal that represents a major threat to Arctic wildlife (AMAP, 2021). Mercury is naturally present in the environment, yet anthropogenic activities release considerable amounts of Hg into the environment (Zhang et al., 2014). Once in the atmosphere, this highly volatile contaminant travels *via* the global atmospheric circulation and reaches even the most remote oceanic regions, such as the Arctic Ocean (Dastoor et al., 2022). As Hg is highly soluble, it reacts with snow, rain and is easily absorbed by particles (Chételat et al., 2022). Therefore, it may be deposited in all ecosystems, including sea ice, where it can be stored for long periods of time (Beattie et al., 2014). Melting sea ice thus represents a major route of Hg deposition to the ocean, where it is assimilated by primary producers and magnifies along the food

webs (Campbell et al., 2005). Furthermore, sea ice can influence the Hg cycle by regulating air-sea exchanges of Hg species (DiMento et al., 2019) and controlling the photochemical transformation processes of Hg (Zheng et al., 2021). These processes further modify Hg bioavailability and the contamination of Arctic marine organisms. Sea ice can also drive Hg levels in marine predators through changes in their trophic ecology and diet. In marine mammals, different contaminant patterns were observed in different sea ice conditions, which were related to prey shifts (Gaden and Stern, 2010; McKinney et al., 2009). In seabirds, changing ice conditions in northern Hudson Bay has resulted in prey shifts (Gaston et al., 2003) toward a higher consumption of less contaminated prey (Braune et al., 2014; Gaston et al., 2012b). Further, weather patterns over multiple years influence exposure of Arctic seabirds to Hg (Foster et al. 2019), presumably due both to deposition but also release or access to Hg already stored in Arctic archives (e.g., glaciers and permafrost). Understanding how Arctic predators rely on sympagic versus pelagic food webs is crucial, not only to foresee future impacts of climate change and associated sea ice melt on ecosystem structure and dynamics, but also because it could largely alter, directly or indirectly, their exposure to contaminants. This is particularly important for species that are used in long-term contaminant monitoring programs, so that changes in contaminant burdens can be interpreted within the context of climate change (Braune et al., 2014).

Multi-tracer approaches enable to track and distinguish the fingerprint of primary producers (*i.e.*, ice algae and pelagic phytoplankton) throughout the entire food web, revealing the dependency of Arctic consumers to sympagic and pelagic resources (Amiraux et al., 2021; Kohlbach et al., 2016; Yurkowski et al., 2020). Highly branched isoprenoids (HBIs) are lipid biomarkers which are specifically synthesized by a restricted number of diatom species (Belt and Müller, 2013). In the Arctic, a restricted number of ice-associated diatoms belonging to genera *Haslea* and *Navicula*

synthesize IP_{25} and diene (a mono- and a di-unsaturated HBI, respectively), which allow the tracking of a sympagic fingerprint through a food web. In contrast, triene (a tri-unsaturated HBI) is mainly synthesized by open-water diatom species, allowing for the tracking of a pelagic fingerprint through a food web (Belt et al., 2013; Brown et al., 2011). In the last decade, these biomarkers have provided key insights into the association with sympagic and pelagic food webs in various consumers (e.g. Amiraux et al., 2023; Brown et al., 2013; Brown and Belt, 2012). Recently, HBIs were combined with carbon ($\delta^{13}C$) and nitrogen stable isotopes (δ^{15} N) – which are commonly used as proxies of predator trophic ecology: the feeding habitat and the diet, respectively (Kelly, 2000) – to establish an index of sea ice use (i.e., Ice Use Index, Cusset et al., 2019).

In this context, the present study investigated the relationship between sea ice, the use of sympagic resources and Hg contamination of an Arctic seabird: the thick-billed murre (or Brünnich's guillemot, *Uria lomvia*, hereafter «murres») breeding in the Canadian high Arctic. Canadian murres, like other marine top predators, show high levels of Hg (Braune et al., 2015, Bond et al., 2015; Chastel et al., 2022; Mallory et al., 2015), and rely on sea ice and ice-derived resources when ice conditions are favorable (Cusset et al., 2019). More specifically, we (i) investigated differences in Hg concentrations measured in bird eggs (an indicator of female short-term contamination, Bond and Diamond, 2010) during four years of contrasting ice conditions (between 2010 and 2013), and (ii) evaluated the drivers of egg Hg contamination (*i.e.*, ice association and trophic ecology of birds). According to previous studies, we expected an increase of egg Hg concentrations during icier years and hence a positive association between Hg contamination and the reliance on sympagic resources.

2. MATERIAL AND METHODS

2.1. Study area, species and egg collection

The present study was carried out on Prince Leopold Island (Lancaster Sound, hereafter "PLI"; 74°N, 90°W; Figure 1). This site represents one of the largest seabird colonies in the Canadian Arctic, supporting approximately 100,000 pairs of thick-billed murres during the breeding season (Gaston et al., 2012a). On PLI, the breeding season runs as follows: colony attendance in mid-June, egg formation (one single egg per nest) in the vicinity of the colony (*i.e.*, for 15 days on average; Gaston and Nettleship, 1981) until laying in early July, hatching in end-July and chick fledging from late August to September.

A total of 60 eggs (15 eggs per year) were collected at the beginning of the breeding season (late June – early July) each year between 2010 and 2013, under appropriate permits as part of a long-term monitoring program supported by Environment and Climate Change Canada and the Northern Contaminants Program (e.g., Braune et al., 2016). Eggs were collected from nests located on the eastern and southern cliffs of PLI. Eggs were kept cool in the field and shipped to the National Wildlife Research Centre (NWRC, Ottawa) for processing. Egg contents were homogenized for 10-20 s using either a stainless steel Sorvall Omni Mixer or a Brinkmann Polytron homogenizer. Egg homogenates (1-2 g aliquots) were then stored frozen at −40 °C in acid-rinsed polyethylene vials for chemical analyses.

In seabirds, Hg acquired through feeding accumulates over time in the whole body (including liver, muscle, kidney). In female seabirds, Hg is excreted *via* two pathways: feather moult and egg formation/laying. Egg Hg could thus result from Hg acquired locally during egg formation in the Arctic, but also more globally, during the previous wintering in subarctic regions where Arctic

seabirds, including murres, can accumulate large concentrations of Hg (Albert et al., 2021). However, murres are income breeders (*i.e.,* allocation of local nutrient to egg formation; Bond and Diamond, 2010; Gaston and Hipfner, 2020). Thus, murre eggs from PLI were assumed to reflect mainly local diet and Hg contamination of females during egg formation (~two weeks).

2.2. Ice conditions

As a high Arctic colony, PLI is surrounded by a dense sea-ice cover in winter that progressively breaks up from west to east in spring and summer. Across years, accessibility to open waters for birds to forage is highly variable, as the ice edge position can range from the colony as far east as the Lancaster Sound Polynya in northern Baffin Bay (> 250 km) (Gaston et al., 2005). Sea-ice conditions were considered during four sampling years (from 2010 to 2013). Since weather patterns over multiple years influence Hg exposure of Arctic birds (Foster et al., 2019), ice conditions during the previous years were also considered to investigate potential time-lags. Because murres exhibit a local feeding range (0−150 km; Gaston et al., 2013), Sea Ice Concentrations (SIC) were calculated daily for the overall breeding season (between May 1 and August 31), over a 9-year period (2006−2013), in a 100 km radius around PLI. SIC were expressed as proportion of ice versus open water pixels, using satellite data from the National Oceanic and Atmospheric Administration dataset (ftp://sidads.colorado.edu/DATASETS/NOAA/G02186/shapefiles/4km/); 4 km resolution raster images). Since egg formation was the critical phase investigated in this study, SIC were compared for June 15 over the years. The 4-year target period exhibited an appreciable sea ice gradient during

murre egg formation, from highest SIC in 2012 (latest ice break-up) to the lowest SIC in 2011 (earliest ice break-up) as follows: 2012 > 2010 > 2013 > 2011 (Table 1).

2.3. Chemical analyses

2.3.1. Trophic tracers

Highly Branched Isoprenoid (HBI) analyses were carried out at Laval University (Québec), using established techniques (Belt et al., 2012), and were published in Cusset et al. (2019). Briefly, egg aliquots were freeze-dried for 48 h prior to lipid extraction. Internal standards (7-hexylnonadecane and 9-octylheptadec-8-ene, 10 µL, 9.68 µg/ml each) were added to egg homogenates to quantify IP_{25} and blanks were analysed with each sample set (n=14) to guarantee analytical accuracy. Total lipids were extracted four consecutive times with dichloromethane:methanol mixture (2:1, 3 min vortex, 15 min sonication). Once dried under a gentle stream of N_2, total lipid extracts were saponified (4 mL, 5% MeOH/H_2O, 90/10, 90°C, 1h). Non-saponifiable lipids (NSL) were then extracted from the saponification mixture by liquid-liquid extraction (hexane, 3 x 2 mL), evaporated (nitrogen stream < 35°C) and purified using open column chromatography (SiO_2 50g/g NSL, hexane), to yield an apolar lipid fraction containing HBIs. HBIs were then analyzed by gas chromatography-mass spectrometry (GC-MS) and quantified by measuring mass spectral intensities for each HBI (IP_{25}: m/z 350.3, diene: m/z 348.3, triene:m/z 346.3). Finally, HBI relative concentrations were combined into the H-Print index (Brown et al., 2014) using Eq. 1:

$$\text{H–Print}\ (\%) = [\text{triene}/(IP_{25} + \text{diene} + \text{triene})] \times 100 \qquad \text{(Eq. 1)}$$

where H–Print indicates a range of association to sympagic (low H-Print), mixed (intermediate) and pelagic (high H-Print) resources.

Stable isotope analyses were conducted at the Littoral, Environment and Societies (LIENSs) Joint Research Unit and its stable isotope facility (La Rochelle) on delipidized samples loaded into tin cups (0.2 to 0.8 mg dry weight). Results were previously published in Cusset et al. (2019). Briefly, stable isotope abundances were determined using an elemental analyzer (Flash EA 1112, Thermo Scientific, Milan, Italy) coupled in continuous flow mode to an isotope ratio mass spectrometer (Delta V Plus with a Conflo IV interface, Thermo Scientific, Bremen, Germany). Results are expressed in the δ notation as deviations from standards (Vienna Pee Dee Belemnite for $\delta^{13}C$ and N_2 in air for $\delta^{15}N$) following the formula: $\delta^{13}C$ or $\delta^{15}N = [(R_{sample}/R_{standard}) - 1] \times 10^3$, where R is $^{13}C/^{12}C$ or $^{15}N/^{14}N$, respectively. Replicate measurements of laboratory standards (USGS-61 and USGS-62) indicated that the measurement uncertainty was < 0.15 ‰ for both $\delta^{15}N$ and $\delta^{13}C$ values. Based on the distinct $\delta^{13}C$ signatures of ice algae and phytoplankton in the Arctic (Hobson et al. 1995), $\delta^{13}C$ isotopic values and H-Print were combined to create an index of sea-ice use, the Ice Use Index (see Cusset et al., 2019 for detailed methodology).

2.3.2. Mercury analyses

Analyses of total Hg (thereafter 'Hg') in egg homogenates were carried out at the NWRC (Ottawa) and were published in Braune et al. (2016). Briefly, samples were homogenized, freeze-dried, homogenized again and weighed into nickel combustion boats. They were then analysed for Hg by direct combustion of the solid sample in an oxygen-rich atmosphere (Salvato and Pirola, 1996), using an Advanced Mercury Analyser (AMA-254, Altec) equipped with an autosampler. Moisture content was determined by weight loss during freeze-drying. One or two blank samples were

206 analysed with each sample set to determine analytical accuracy, as well as standard reference
207 materials (SRMSs). Practical detection limits were 0.006 and 0.004 µg/g dry weight for the 2010-
208 2012 samples and the 2013 samples, respectively. All Hg concentrations are expressed in µg/g dry
209 weight (dw). For further analytical details, including reference materials and recoveries, the reader
210 is referred to Braune et al. (2016).

211 **2.4. Statistical analyses**

212 All statistical analyses and figures were computed using R version 4.0.3 (R Core Team, 2020).
213 First, inter-annual differences in egg Hg concentrations were tested using one-way ANOVA. Data
214 homogeneity and residuals normality were checked prior interpretation, using Breush-Pagan
215 (package «lmtest»; Zeileis and Hothorn, 2002) and Shapiro tests, respectively. In addition,
216 differences in ice regimes (extensive- *versus* reduced-ice years) were tested using Welch two-
217 sample t-tests.

218 Then, drivers of egg Hg concentrations were investigated following two steps. First, simple linear
219 regressions were used to investigate independently the relationships between egg Hg
220 concentrations and (i) ice concentrations, (ii) ice association (Ice Use Index) and (iii) δ^{15}N values.
221 Second, we used general linear models (GLMs) to investigate the role of these explanatory
222 variables and their combination on Hg concentrations in murre eggs («nlme» package; Pinheiro et
223 al., 2020). Correlations between variables were checked prior to model construction («cor»
224 function). Explanatory variables were standardized (mean = 0, SD = 1) to facilitate comparison of
225 effect sizes. Model assumptions were checked visually with diagnostic functions («plot» function).
226 All potential combinations of variables (Table 2) were subjected to model selection using the
227 «AICcmodavg» package (Mazerolle, 2020). Models were compared using Akaike's Information

228 Criterion corrected for small sample sizes (AIC_c). The model with the lowest AIC_c value was
229 selected as the best model (Burnham and Anderson, 2002). Model support was assessed using
230 Akaike weights (w_i), following Johnson and Omland (2004) and model fit was checked by residual
231 analysis.

232 During data exploration, time-lags were investigated following previous findings (Foster et al.,
233 2019). Available ice data for the present dataset only dated back to 2006, implying a maximum
234 time-lag of four years for sea ice (instead of seven years as showed by Foster et al., 2019). Given
235 the lack of statistical improvement when added in the models, time-lags were excluded from final
236 analyses.

237

238 **3. RESULTS**

239 **3.1. Hg concentrations in changing ice conditions**

240 Analyses of Hg concentrations in eggs varied significantly among years (p<0.0001, $F_{3,56}$ = 7.4,
241 ANOVA) : 2012 (1.2 ± 0.2 µg/g; heavy dense ice) differed from 2011 (0.9 ± 0.2 µg/g; ice-free)
242 and 2013 (1.0 ± 0.2 µg/g; low ice), and 2010 (1.1 ± 0.2 µg/g; heavy patchy ice) differed from 2011
243 (Figure S1). Hg concentrations were lower during reduced-ice years (2011 and 2013; 0.92 ± 0.19
244 µg/g, n=30) than during extensive-ice years (2010 and 2012; 1.16 ± 0.23 µg/g, n=30) (p< 0.0001,
245 $t_{56.7}$=4.6, Welch two-sample t-test; Figure S2). Hence, Hg concentrations increased by 30%
246 between the minimum and maximum ice years (Table 1).

247 **3.2. Drivers of egg Hg contamination in changing ice conditions**

Unifactorial analyses showed that Hg concentrations in murre eggs increased linearly with increasing ice concentrations (R^2=0.25, p<0.0001, y=0.0037x + 0.78), and with increasing Ice Use Index (R^2=0.19, p=0.001, y=0.082 x + 1.0; Figure 2), but not with δ^{15} N values (p=0.08) (Figure 3). Multifactorial analyses revealed that the best model only included ice concentrations as an explanatory variable (Table 2; Figure 4). Effect size for this model was −0.14 ± 0.03 (−0.2, −0.08; Gamma inverse).

4. DISCUSSION

Climate change has a strong impact on sea ice in the Arctic, for instance its distribution, extent, thickness and phenology (Garcia-Soto et al., 2021). Yet, melting sea ice could in turn play a significant role in the contamination by Hg of Arctic marine biota and human populations, through changes in Hg biogeochemistry or modifications of the food web structure (AMAP, 2022; Chételat et al., 2022). By using a new approach combining multiple trophic tracers, Hg analyses and sea ice data, we highlighted the influence of sea ice and sympagic food webs on Hg contamination in murres during the early breeding season.

In years with more extensive sea ice, murre eggs contained higher concentrations of Hg (Figure 4). When considering two distinct ice regimes with relatively "warm" (in 2011 and 2013) and "cold" (in 2010 and 2012) years characterized by reduced and extensive ice cover, respectively, Hg concentrations were higher during cold years compared to warm ones (Figure S2). These results are in agreement with previous investigations using different ice metrics (Mallory and Braune, 2018). Contrasting Hg levels could result from different foraging ecologies and consumed prey according to ice conditions. The main route of Hg uptake in predators is through their diet

(Atwell et al., 1998; Campbell et al., 2005; Power et al., 2002; Thompson et al., 1998). As pursuit-divers, murres at PLI feed extensively on fish, such as Arctic cod (*Boreogadus saida*), capelin (*Mallotus villosus*) and sandlance (*Ammodytes sp.*), as well as on macrozooplankton, including amphipods (Hyperiidae and Gammaridae) and copepods (*Calanus spp.*; Gaston and Nettleship, 1981). Higher Hg contamination during extensive ice years could therefore result from predation on (i) larger prey species which have bioaccumulated more Hg in their tissues and/or (ii) prey with higher δ^{15}N values resulting in higher Hg contamination (biomagnification; Dietz et al., 1996). Following this hypothesis, murres' δ^{15}N values should directly influence their Hg contamination. However, there was a weak relationship between egg Hg concentrations and δ^{15}N isotopic values (Figure 3), suggesting that temporal changes in trophic ecology alone could not explain higher Hg contamination during icier years, a result consistent with previous findings (Braune et al. 2016, Moody et al. 2012, Provencher et al. 2012).

Alternatively, higher Hg concentrations in murres during icy years could be the consequence of a higher contamination of sympagic food webs, compared to the more pelagic ones. Indeed, our results showed that egg Hg contamination was positively related to the Ice Use Index (Figure 2), which reflects bird association with sympagic resources. Hence, murres relying more heavily on ice-derived resources during the early breeding season exhibited higher Hg concentrations. Similar findings were observed in polar bears (*Ursus maritimus)*, which exhibited higher Hg concentrations when feeding on food webs dominated by sea-ice associated diatoms (Routti et al. 2012). Higher Hg concentrations were also documented in Arctic cod, an emblematic Arctic fish species that is closely tied to sea ice (Kohlbach et al. 2017). Arctic cod, the main prey of murres in the High Arctic, especially at PLI (Provencher et al., 2012), use under-ice habitats for part of their life cycle (David et al. 2016, Gradinger & Bluhm 2004), for instance as a refuge to escape

visual predators (LeBlanc et al., 2019). Marginal ice zones (MIZ) thus represent an important foraging ground for murres (Bradstreet and Cross, 1982). Yet, high Hg concentrations were observed in MIZ, below the productive surface layer (Heimbürger et al. 2015), enhancing Hg uptake and bioavailability in ice-associated food webs up to top predators and seabirds such as murres. Consequently, foraging in these productive habitats may also expose murres to higher Hg in their diet.

Sea ice is composed of different semi-solid matrices, such as brines, gases, particles, and biota (e.g., bacteria, viruses, algae). These matrices are subject to physical, biological, and chemical processes which could influence Hg contamination in Arctic marine ecosystems during years of extensive ice (Steffen et al., 2008; Thomas and Dieckmann, 2009). Sea-ice permeability, shaped by temperatures and the creation of brine pockets and drainage channels, considerably influences Hg processes through changing redox conditions and the presence of sea-ice biota (Thomas and Dieckmann, 2009). Indeed, Hg methylation has been documented in sea ice (Beattie et al., 2014) and Hg concentrations were the highest in brines (Chaulk et al., 2011), which represent the primary habitat for microbial communities that sustain sympagic food webs (Thomas and Dieckmann, 2002). Brines provide more Hg-enriched environments for primary producers (Schartup et al. 2020), including ice algae, potentially increasing food web contamination (Stern et al. 2012). Sea ice could act as an important entry point for Hg at the base of sympagic food webs, additionally enhancing murre Hg contamination when relying on sympagic resources. Thus, sources of Hg found in murre eggs could either originate from melting sea ice and snow where Hg can be stored (Zdanowicz et al., 2013) or from air-ocean exchanges of Hg species (Lalonde et al., 2002; Poulain et al., 2007).

315 Despite the existing relationship between Hg contamination in murre eggs and their use of
316 sympagic resources, model selection revealed that the best predictor for egg Hg concentrations
317 was sea ice concentration only, and did not include the Ice Use Index nor the δ^{15} N. This suggests
318 that the processes involved are more complex than the reliance on sympagic resources alone. Hg
319 contamination in murres can result from long-term processes with a timescale of years, decades
320 and even centuries (UNEP, 2018). For instance, environmental factors such as the North Atlantic
321 Oscillation (NAO), snow cover or air temperatures were previously identified as drivers of Hg
322 contamination in murre eggs, with a lag time of several years (Foster et al., 2019). Mercury can
323 indeed remain up to one year in the atmosphere (Streets et al., 2019), before deposition and its
324 resulting methylation, assimilation and final transfer along the food chain. Mercury dynamics are
325 driven by a large number of interacting post-depositional processes, which control the transport,
326 methylation and biological uptake of Hg stored in the Arctic Ocean (Dastoor et al., 2022; Outridge
327 et al., 2008). Thus, reduced sea ice cover or earlier ice break-up may expose murres to lower Hg
328 contamination if this was the only change underway. However, air and ocean warming actually
329 increase local export of Hg to Arctic marine areas from environmental archives found largely on
330 land (Chételat et al., 2022; Dastoor et al., 2022). For this reason, it is difficult to disentangle all the
331 processes involved here and one might expect a period of flux and unpredictability, at least until
332 Arctic Hg archives decrease their contribution to marine environments.

333 Considering its long half-life, Hg is highly persistent in the body of living organisms. Although
334 eggs mainly reflect local diet and Hg contamination in income-breeders like murres (i.e., in the
335 Arctic, during egg formation), they could also integrate, to a low extent, Hg accumulated outside
336 the Arctic (i.e., during the non-breeding season) and remobilized from storing tissues during egg
337 synthesis. Previous studies have reported carry-over effects of adult winter contamination on the

subsequent summer Hg concentrations in blood of little auks (Carravieri et al., 2023), as well as higher Hg contamination during the non-breeding season relative to the breeding season in many alcids, including thick-billed murres (Albert et al., 2021). Future research should now further investigate how the reliance of Arctic seabirds to ice-derived resources during the non-breeding season can expose them to Hg and other chemical contaminants.

5. CONCLUSION

Sea ice is a key component of Arctic marine environments, where it drives primary productivity and shape the phenology of ice algae and pelagic phytoplankton. At the base of Arctic food webs, these primary producers provide the vital energy pulses for the entire marine food web to thrive. Simultaneously, they assimilate contaminants, such as Hg, which are then transferred to all consumers and predators along the food web. In this study, association with sea ice, and more particularly the reliance on sympagic resources, was positively related to Hg concentrations in eggs of thick-billed murres from the Canadian High Arctic. This work provides new insights into the role of sea ice and ice-derived resources in Hg contamination for Arctic wildlife and human populations that heavily depend on marine resources for subsistence. Further research is now required to fully understand the role of ice-derived resources and sea-ice melt in future Hg contamination of Arctic biota under climate change, as well as the ecological and biogeochemical interactions involved.

ACKNOWLEDGMENTS

The authors are thankful to the numerous people involved in field work over the years. Logistical support for the fieldwork out of Resolute Bay was provided by the Polar Continental Shelf Program of Natural Resources Canada. Special thanks are due to Caroline Guilmette for her support during HBI analyses, and to Céline Albert and Alice Carravieri for statistical guidance during data analyses. We acknowledge the use of the NOAA satellite dataset. This study also represents a contribution to the French Polar Institute ADACLIM research program (IPEV Pgr 388).

ADDITIONAL INFORMATION

Competing of interest. Authors have no conflict of interest to declare.

Contributions.

Conception and design: FC, JF, GM, MM

Laboratory work and data acquisition: FC (trophic tracers, ice parameters), BB (mercury), GG (stable isotopes) and PM (ice analyses)

Data analysis: FC

Interpretation: FC, JC, JF, GM, MM, JP

First draft: FC, JC

Supervision: GM, JF

Revision and final approval: FC, JC, GM, MM, BB, JP, GG, PM, JF

REFERENCES

Albert, C., Helgason, H.H., Brault-Favrou, M., Robertson, G.J., Descamps, S., Amélineau, F., Danielsen, J., Dietz, R., Elliott, K., Erikstad, K.E., Eulaers, I., Ezhov, A., Fitzsimmons, M.G., Gavrilo, M., Golubova, E., Grémillet, D., Hatch, S., Huffeldt, N.P., Jakubas, D., Kitaysky, A., Kolbeinsson, Y., Krasnov, Y., Lorentsen, S.-H., Lorentzen, E., Mallory, M.L., Merkel, B., Merkel, F.R., Montevecchi, W., Mosbech, A., Olsen, B., Orben, R.A., Patterson, A., Provencher, J., Plumejeaud, C., Pratte, I., Reiertsen, T.K., Renner, H., Rojek, N., Romano, M., Strøm, H., Systad, G.H., Takahashi, A., Thiebot, J.-B., Thórarinsson, T.L., Will, A.P., Wojczulanis-Jakubas, K., Bustamante, P., Fort, J., 2021. Seasonal variation of mercury contamination in Arctic seabirds: A pan-Arctic assessment. Sci. Total Environ. 750, 142201. https://doi.org/10.1016/j.scitotenv.2020.142201

AMAP, 2021. AMAP Assessment 2021: Mercury in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Tromsø, Norway. viii + 324pp

Amiraux, R., Archambault, P., Moriceau, B., Lemire, M., Babin, M., Memery, L., Massé, G., Tremblay, J.-E., 2021. Efficiency of sympagic-benthic coupling revealed by analyses of n-3 fatty acids, IP_{25} and other highly branched isoprenoids in two filter-feeding Arctic benthic molluscs: *Mya truncata* and *Serripes groenlandicus*. Org. Geochem. 151, 104160. https://doi.org/10.1016/j.orggeochem.2020.104160

Amiraux, R., Mundy, C.J., Pierrejean, M., Niemi, A., Hedges, K.J., Brown, T.A., Ehn, J.K., Elliott, K.H., Ferguson, S.H., Fisk, A.T., Gilchrist, G., Harris, L.N., Iken, K., Jacobs, K.B., Johnson, K.F., Kuzyk, Z.A., Limoges, A., Loewen, T.N., Love, O.P., Matthews, C.J.D., Ogloff, W.R., Rosenberg, B., Søreide, J.E., Watt, C.A., Yurkowski, D.J., 2023. Tracing carbon flow and trophic structure of a coastal Arctic marine food web using highly branched isoprenoids and carbon, nitrogen and sulfur stable isotopes. Ecol. Indic. 147, 109938. https://doi.org/10.1016/j.ecolind.2023.109938

Atwell, L., Hobson, K.A., Welch, H.E., 1998. Biomagnification and bioaccumulation of mercury in an Arctic marine food web: Insights from stable nitrogen isotope analysis 55, 8.

Beattie, S.A., Armstrong, D., Chaulk, A., Comte, J., Gosselin, M., Wang, F., 2014. Total and methylated mercury in Arctic multiyear sea ice. Environ. Sci. Technol. 48, 5575–5582. https://doi.org/10.1021/es5008033

Belt, S.T., Brown, T.A., Ringrose, A.E., Cabedo-Sanz, P., Mundy, C.J., Gosselin, M., Poulin, M., 2013. Quantitative measurement of the sea ice diatom biomarker IP_{25} and sterols in Arctic sea ice and underlying sediments: Further considerations for palaeo sea ice reconstruction. Org. Geochem. 62, 33–45. https://doi.org/10.1016/j.orggeochem.2013.07.002

Belt, S.T., Brown, T.A., Rodriguez, A.N., Sanz, P.C., Tonkin, A., Ingle, R., 2012. A reproducible method for the extraction, identification and quantification of the Arctic sea ice proxy IP_{25} from marine sediments. Anal. Methods 4, 705–713. https://doi.org/10.1039/C2AY05728J

Belt, S.T., Müller, J., 2013. The Arctic sea ice biomarker IP_{25}: A review of current understanding, recommendations for future research and applications in palaeo sea ice reconstructions. Quat. Sci. Rev. 79, 9–25. https://doi.org/10.1016/j.quascirev.2012.12.001

Bond, A.L., Diamond, A.W., 2010. Nutrient allocation for egg production in six Atlantic seabirds. Can. J. Zool. 88, 1095–1102. https://doi.org/10.1139/Z10-082

Bond, A.L., Hobson, K.A., Branfireun, B.A., 2015. Rapidly increasing methyl mercury in endangered ivory gull (*Pagophila eburnea*) feathers over a 130 year record. Proc. R. Soc. B. 282:20150032. http://doi.org/10.1098/rspb.2015.0032

Bradstreet, M.S.W., Cross, W.E., 1982. Trophic relationships at High Arctic ice edges. Arctic 35, 1–12.

Braune, B., Chételat, J., Amyot, M., Brown, T., Clayden, M., Evans, M., Fisk, A., Gaden, A., Girard, C., Hare, A., Kirk, J., Lehnherr, I., Letcher, R., Loseto, L., Macdonald, R., Mann, E., McMeans, B., Muir, D., O'Driscoll, N., Poulain, A., Reimer, K., Stern, G., 2015. Mercury in the marine environment of the Canadian Arctic: Review of recent findings. Sci. Total Environ., Special Issue: Mercury in Canada's North 509–510, 67–90. https://doi.org/10.1016/j.scitotenv.2014.05.133

Braune, B.M., Gaston, A.J., Elliott, K.H., Provencher, J.F., Woo, K.J., Chambellant, M., Ferguson, S.H., Letcher, R.J., 2014. Organohalogen contaminants and total mercury in forage fish preyed upon by thick-billed murres in northern Hudson Bay. Mar. Pollut. Bull. 78, 258–266. https://doi.org/10.1016/j.marpolbul.2013.11.003

Braune, B.M., Gaston, A.J., Mallory, M.L., 2016. Temporal trends of mercury in eggs of five sympatrically breeding seabird species in the Canadian Arctic. Environ. Pollut. 214, 124–131. https://doi.org/10.1016/j.envpol.2016.04.006

Brown, T.A., Belt, S.T., 2012. Identification of the sea ice diatom biomarker IP_{25} in Arctic benthic macrofauna: Direct evidence for a sea ice diatom diet in Arctic heterotrophs. Polar Biol. 35, 131–137. https://doi.org/10.1007/s00300-011-1045-7

Brown, T.A., Belt, S.T., Philippe, B., Mundy, C.J., Massé, G., Poulin, M., Gosselin, M., 2011. Temporal and vertical variations of lipid biomarkers during a bottom ice diatom bloom in the Canadian Beaufort Sea: Further evidence for the use of the IP_{25} biomarker as a proxy for spring Arctic sea ice. Polar Biol. 34, 1857–1868. https://doi.org/10.1007/s00300-010-0942-5

Brown, T.A., Bicknell, A.W.J., Votier, S.C., Belt, S.T., 2013. Novel molecular fingerprinting of marine avian diet provides a tool for gaining insights into feeding ecology. Environ. Chem. Lett. 11, 283–288. https://doi.org/10.1007/s10311-013-0402-x

Burnham, K.P., Anderson, D.R., 2002. Model selection and multimodel inference: A practical information 616 - Theoretic approach, 2nd ed. Springer US.

Campbell, L.M., Norstrom, R.J., Hobson, K.A., Muir, D.C.G., Backus, S., Fisk, A.T., 2005a. Mercury and other trace elements in a pelagic Arctic marine food web (Northwater Polynya, Baffin Bay). Sci. Total Environ., 351–352, 247–263. https://doi.org/10.1016/j.scitotenv.2005.02.043

Carravieri, A., Lorioux, S., Angelier, F., Chastel, O., Albert, C., Bråthen, V.S., Brisson-Curadeau, É., Clairbaux, M., Delord, K., Giraudeau, M., Perret, S., Poupart, T., Ribout, C., Viricel-Pante, A., Grémillet, D., Bustamante, P., Fort, J. 2023. Carryover effects of winter mercury contamination on summer concentrations and reproductive performance in little auks, Environ. Pollut., 318, 120774, ISSN 0269-7491. https://doi.org/10.1016/j.envpol.2022.120774.

Chastel, O., Fort, J., Ackerman, J.T., Albert, C., Angelier, F., Basu, N., Blévin, P., Brault-Favrou, M., Bustnes, J.O., Bustamante, P., Danielsen, J., Descamps, S., Dietz, R., Erikstad, K.E., Eulaers, I., Ezhov, A., Fleishman, A.B., Gabrielsen, G.W., Gavrilo, M., Gilchrist, G., Gilg, O., Gíslason, S., Golubova, E., Goutte, A., Grémillet, D., Hallgrimsson, G.T., Hansen, E.S., Hanssen, S.A., Hatch, S., Huffeldt, N.P., Jakubas, D., Jónsson, J.E., Kitaysky, A.S., Kolbeinsson, Y., Krasnov, Y., Letcher, R.J., Linnebjerg, J.F., Mallory, M., Merkel, F.R., Moe, B., Montevecchi, W.J., Mosbech, A., Olsen, B., Orben, R.A., Provencher, J.F., Ragnarsdottir, S.B., Reiertsen, T.K., Rojek, N., Romano, M., Søndergaard, J., Strøm, H., Takahashi, A., Tartu, S., Thórarinsson, T.L., Thiebot, J.-B., Will, A.P., Wilson, S., Wojczulanis-Jakubas, K., Yannic, G., 2022. Mercury contamination and potential health risks to Arctic seabirds and shorebirds. Sci. Total Environ. 156944. https://doi.org/10.1016/j.scitotenv.2022.156944

Chaulk, A., Stern, G.A., Armstrong, D., Barber, D.G., Wang, F., 2011. Mercury distribution and transport across the ocean−sea-ice−atmosphere interface in the Arctic Ocean. Environ. Sci. Technol. 45, 1866–1872. https://doi.org/10.1021/es103434c

Chételat, J., McKinney, M.A., Amyot, M., Dastoor, A., Douglas, T.A., Heimbürger-Boavida, L.-E., Kirk, J., Kahilainen, K.K., Outridge, P.M., Pelletier, N., Skov, H., St. Pierre, K., Vuorenmaa, J., Wang, F., 2022. Climate change and mercury in the Arctic: Abiotic interactions. Sci. Total Environ. 824, 153715. https://doi.org/10.1016/j.scitotenv.2022.153715

Cusset, F., Fort, J., Mallory, M., Braune, B., Massicotte, P., Massé, G., 2019. Arctic seabirds and shrinking sea-ice: Egg analyses reveal the importance of ice-derived resources. Sci. Rep. 9, 1–15. https://doi.org/doi.org/10.1038/s41598-019-51788-4

Dastoor, A., Angot, H., Bieser, J., Christensen, J.H., Douglas, T.A., Heimbürger-Boavida, L.-E., Jiskra, M., Mason, R.P., McLagan, D.S., Obrist, D., Outridge, P.M., Petrova, M.V., Ryjkov, A., St. Pierre, K.A., Schartup, A.T., Soerensen, A.L., Toyota, K., Travnikov, O., Wilson, S.J., Zdanowicz, C., 2022a. Arctic mercury cycling. Nat. Rev. Earth Environ. 3, 270–286. https://doi.org/10.1038/s43017-022-00269-w

Dastoor, A., Angot, H., Bieser, J., Christensen, J.H., Douglas, T.A., Heimbürger-Boavida, L.-E., Jiskra, M., Mason, R.P., McLagan, D.S., Obrist, D., Outridge, P.M., Petrova, M.V.,

Ryjkov, A., St. Pierre, K.A., Schartup, A.T., Soerensen, A.L., Toyota, K., Travnikov, O., Wilson, S.J., Zdanowicz, C., 2022b. Arctic mercury cycling. Nat. Rev. Earth Environ. 3, 270–286. https://doi.org/10.1038/s43017-022-00269-w

David, C., Lange, B., Krumpen, T., Schaafsma, F., van Franeker, J.A., Flores, H., 2016. Under-ice distribution of polar cod *Boreogadus saida* in the central Arctic Ocean and their association with sea-ice habitat properties. Polar Biol. 39, 981–994. https://doi.org/10.1007/s00300-015-1774-0

Dietz, R., Letcher, R.J., Aars, J., Andersen, M., Boltunov, A., Born, E.W., Ciesielski, T.M., Das, K., Dastnai, S., Derocher, A.E., Desforges, J.-P., Eulaers, I., Ferguson, S., Hallanger, I.G., Heide-Jørgensen, M.P., Heimbürger-Boavida, L.-E., Hoekstra, P.F., Jenssen, B.M., Kohler, S.G., Larsen, M.M., Lindstrøm, U., Lippold, A., Morris, A., Nabe-Nielsen, J., Nielsen, N.H., Peacock, E., Pinzone, M., Rigét, F.F., Rosing-Asvid, A., Routti, H., Siebert, U., Stenson, G., Stern, G., Strand, J., Søndergaard, J., Treu, G., Víkingsson, G.A., Wang, F., Welker, J.M., Wiig, Ø., Wilson, S.J., Sonne, C., 2022. A risk assessment review of mercury exposure in Arctic marine and terrestrial mammals. Sci. Total Environ. 829, 154445. https://doi.org/10.1016/j.scitotenv.2022.154445

Dietz, R., Riget, F., Johansen, P., 1996. Lead, cadmium, mercury and selenium in Greenland marine animals. Sci. Total Environ. 186, 67–93. https://doi.org/10.1016/0048-9697(96)05086-3

DiMento, B.P., Mason, R.P., Brooks, S., Moore, C., 2019. The impact of sea ice on the air-sea exchange of mercury in the Arctic Ocean. Deep Sea Res. Part Oceanogr. Res. Pap. 144, 28–38. https://doi.org/10.1016/j.dsr.2018.12.001

Foster, K.L., Braune, B.M., Gaston, A.J., Mallory, M.L., 2019. Climate influence on mercury in Arctic seabirds. Sci. Total Environ. 693, 133569. https://doi.org/10.1016/j.scitotenv.2019.07.375

Garcia-Soto, C., Cheng, L., Caesar, L., Schmidtko, S., Jewett, E.B., Cheripka, A., Rigor, I., Caballero, A., Chiba, S., Báez, J.C., Zielinski, T., Abraham, J.P., 2021. An overview of ocean climate change indicators: Sea surface temperature, ocean heat content, ocean pH, dissolved oxygen concentration, Arctic sea ice extent, thickness and volume, sea level and strength of the AMOC (Atlantic Meridional Overturning Circulation). Front. Mar. Sci. 8, 642372. https://doi.org/10.3389/fmars.2021.642372

Gaston, A.J., Elliott, K.H., Ropert-Coudert, Y., Kato, A., Macdonald, C.A., Mallory, M.L., Gilchrist, H.G., 2013. Modeling foraging range for breeding colonies of thick-billed murres *Uria lomvia* in the Eastern Canadian Arctic and potential overlap with industrial development. Biol. Conserv. 168, 134–143. https://doi.org/10.1016/j.biocon.2013.09.018

Gaston, A.J., Gilchrist, H.G., Mallory, M.L., 2005. Variation in ice conditions has strong effects on the breeding of marine birds at Prince Leopold Island, Nunavut. Ecography 28, 331–344. https://doi.org/10.1111/j.0906-7590.2005.04179.x

Gaston, A. J., & Hipfner, J. M. (2020). Thick-billed Murre (*Uria lomvia*), version 1.0. In Birds of the World (S. M. Billerman, Editor). Cornell Lab of Ornithology, Ithaca, NY, USA. https://doi-org/10.2173/bow.thbmur.01

Gaston, A.J., Mallory, M.L., Gilchrist, H.G., 2012a. Populations and trends of Canadian Arctic seabirds. Polar Biol. 35, 1221–1232. https://doi.org/10.1007/s00300-012-1168-5

Gaston, A.J., Nettleship, D.N., 1981. The thick-billed murre of Prince Leopold Island, Monograph Series. Environment Canada - Canadian Wildlife Service, Ottawa.

Gaston, A.J., Smith, P.A., Provencher, J.F., 2012b. Discontinuous change in ice cover in Hudson Bay in the 1990s and some consequences for marine birds and their prey. ICES J. Mar. Sci. 69, 1218–1225. https://doi.org/10.1093/icesjms/fss040

Gaston, A.J., Woo, K., Hipfner, J.M., 2003. Trends in forage fish populations in Northern Hudson Bay since 1981, as determined from the diet of nestling thick-billed murres, *Uria lomvia*. ARCTIC 56, 227–233. https://doi.org/10.14430/arctic618

Gosselin, M., Levasseur, M., Wheeler, P.A., Horner, R.A., Booth, B.C., 1997. New measurements of phytoplankton and ice algal production in the Arctic Ocean. Deep Sea Res. Part II Top. Stud. Oceanogr. 44, 1623–1644. https://doi.org/10.1016/S0967-0645(97)00054-4

Gradinger, R.R., Bluhm, B.A., 2004. In-situ observations on the distribution and behavior of amphipods and Arctic cod (Boreogadus saida) under the sea ice of the High Arctic Canada Basin. Polar Biol. 27, 595–603. https://doi.org/10.1007/s00300-004-0630-4

Heimbürger, L.-E., Sonke, J.E., Cossa, D., Point, D., Lagane, C., Laffont, L., Galfond, B.T., Nicolaus, M., Rabe, B., van der Loeff, M.R., 2015. Shallow methylmercury production in the marginal sea ice zone of the central Arctic Ocean. Sci. Rep. 5, 10318. https://doi.org/10.1038/srep10318

Horner, R., Schrader, G.C., 1982. Relative contributions of ice algae, phytoplankton, and benthic microalgae to primary production in nearshore regions of the Beaufort Sea. Arctic 35, 485–503.

Johnson, J.B., Omland, K.S., 2004. Model selection in ecology and evolution. Trends Ecol. Evol. 19, 101744 108. https://doi.org/10.1016/j.tree.2003.10.013

Kelly, J.F., 2000. Stable isotopes of carbon and nitrogen in the study of avian and mammalian trophic ecology. Can. J. Zool. 78, 1–27. https://doi.org/10.1139/z99-165

Kohlbach, D., Graeve, M., Lange, B.A., David, C., Peeken, I., Flores, H., 2016. The importance of ice algae-produced carbon in the central Arctic Ocean ecosystem: Food web relationships revealed by lipid and stable isotope analyses. Limnol. Oceanogr. 61, 2027–2044. https://doi.org/10.1002/lno.10351

Kohlbach, D., Schaafsma, F.L., Graeve, M., Lebreton, B., Lange, B.A., David, C., Vortkamp, M., Flores, H., 2017. Strong linkage of polar cod (*Boreogadus saida*) to sea ice algae-produced

carbon: Evidence from stomach content, fatty acid and stable isotope analyses. Prog. Oceanogr. 152, 62–74. https://doi.org/10.1016/j.pocean.2017.02.003

Lalonde, J.D., Poulain, A.J., Amyot, M., 2002. The role of mercury redox reactions in snow on snow-to-air mercury transfer. Environ. Sci. Technol. 36, 174–178. https://doi.org/10.1021/es010786g

LeBlanc, M., Gauthier, S., Garbus, S.E., Mosbech, A., Fortier, L., 2019. The co-distribution of Arctic cod and its seabird predators across the marginal ice zone in Baffin Bay. Elem Sci Anth 7, 1. https://doi.org/10.1525/elementa.339

Mallory, M.L., Braune, B.M., 2018. Do concentrations in eggs and liver tissue tell the same story of temporal trends of mercury in high Arctic seabirds? J. Environ. Sci. 68, 65–72. https://doi.org/10.1016/j.jes.2017.10.017

Mallory, M.L., Braune, B.M., Provencher, J.F., Callaghan, D.B., Gilchrist, H.G., Edmonds, S.T., Allard, K., O'Driscoll, N.J., 2015. Mercury concentrations in feathers of marine birds in Arctic Canada. Mar. Pollut. Bull. 98, 308–313. https://doi.org/10.1016/j.marpolbul.2015.06.043

Mazerolle, M.J., 2020. AICcmodavg: Model selection and multimodel inference based on (Q)AIC(c). R package version 2.3-1. https://cran.r-project.org/package=AICcmodavg.

McKinney, M.A., Peacock, E., Letcher, R.J., 2009. Sea ice-associated diet change increases the levels of chlorinated and brominated contaminants in polar bears. Environ. Sci. Technol. 43, 4334–4339. https://doi.org/10.1021/es900471g

Outridge, P.M., Macdonald, R.W., Wang, F., Stern, G.A., Dastoor, A.P., 2008. A mass balance inventory of mercury in the Arctic Ocean. Environ. Chem. 5, 89. https://doi.org/10.1071/EN08002

Perrette, M., Yool, A., Quartly, G.D., Popova, E.E., 2011. Near-ubiquity of ice-edge blooms in the Arctic. Biogeosciences 8, 515–524. https://doi.org/10.5194/bg-8-515-2011

Pinheiro, J., Bates, D., DebRoy, S., Sarkar, D., R Core Team, 2020. nlme: Linear and nonlinear mixed effects models. R package version 3.1-149, https://CRAN.R-project.org/package=nlme

Poulain, A.J., Garcia, E., Amyot, M., Campbell, P.G.C., Raofie, F., Ariya, P.A., 2007. Biological and chemical redox transformations of mercury in fresh and salt waters of the High Arctic during spring and summer. Environ. Sci. Technol. 41, 1883–1888. https://doi.org/10.1021/es061980b

Power, M., Klein, G.M., Guiguer, K.R.R.A., Kwan, M.K.H., 2002. Mercury accumulation in the fish community of a sub-Arctic lake in relation to trophic position and carbon sources: Hg in a sub-Arctic lacustrine fish community. J. Appl. Ecol. 39, 819–830. https://doi.org/10.1046/j.1365-2664.2002.00758.x

602 Provencher, J.F., Gaston, A.J., O'Hara, P.D., Gilchrist, H.G., 2012. Seabird diet indicates changing
603 Arctic marine communities in eastern Canada. Mar. Ecol. Prog.Ser. 454, 171–182.
604 https://doi.org/10.3354/meps09299

605 R Core Team, 2020. R: A language an environment for statistical computing. R Foundation for
606 statistical computing, Vienna, Austria. URL: https://www.R-project.org/

607 Routti, H., Letcher, R.J., Born, E.W., Branigan, M., Dietz, R., Evans, T.J., McKinney, M.A.,
608 Peacock, E., Sonne, C., 2012. Influence of carbon and lipid sources on variation of mercury
609 and other trace elements in polar bears (*Ursus maritimus*). Environ. Toxicol. Chem. 31,
610 2739–2747. https://doi.org/10.1002/etc.2005

611 Salvato, N., Pirola, C., 1996. Analysis of mercury traces by means of solid sample atomic
612 absorption spectrometry. Microchim. Acta 123, 63–71.
613 https://doi.org/10.1007/BF01244379

614 Schartup, A.T., Soerensen, A.L., Heimbürger-Boavida, L.-E., 2020. Influence of the Arctic sea-
615 ice regime shift on sea-ice methylated mercury trends. Environ. Sci. Technol. Lett. 7, 708–
616 713. https://doi.org/10.1021/acs.estlett.0c00465

617 Steffen, A., Douglas, T., Amyot, M., Ariya, P., Aspmo, K., Berg, T., Bottenheim, J., Brooks, S.,
618 Cobbett, F., Dastoor, A., Dommergue, A., Ebinghaus, R., Ferrari, C., Gardfeldt, K.,
619 Goodsite, M.E., Lean, D., Poulain, A.J., Scherz, C., Skov, H., Sommar, J., Temme, C.,
620 2008. A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and
621 snow. Atmospheric Chem. Phys. 8, 1445–1482. https://doi.org/10.5194/acp-8-1445-2008

622 Stern, G.A., Macdonald, R.W., Outridge, P.M., Wilson, S., Chételat, J., Cole, A., Hintelmann, H.,
623 Loseto, L.L., Steffen, A., Wang, F., Zdanowicz, C., 2012. How does climate change
624 influence Arctic mercury? Sci. Total Environ. 414, 22–42.
625 https://doi.org/10.1016/j.scitotenv.2011.10.039

626 Streets, D.G., Horowitz, H.M., Lu, Z., Levin, L., Thackray, C.P., Sunderland, E.M., 2019. Five
627 hundred years of anthropogenic mercury: spatial and temporal release profiles*. Environ
628 Res Lett 14:084004. https://doi.org/10.1088/1748-9326/ab281f

629 Thomas, D.N., Dieckmann, G.S., 2009. Sea ice. John Wiley & Sons.

630 Thomas, D.N., Dieckmann, G.S., 2002. Antarctic sea ice: A Habitat for extremophiles. Science
631 295, 641–644. https://doi.org/10.1126/science.1063391

632 Thompson, D.R., Furness, R.W., Monteiro, L.R., 1998. Seabirds as biomonitors of mercury inputs
633 to epipelagic and mesopelagic marine food chains. Sci. Total Environ. 213, 299–305.
634 https://doi.org/10.1016/S0048-9697(98)00103-X

635 Tremblay, J.-É., Robert, D., Varela, D.E., Lovejoy, C., Darnis, G., Nelson, R.J., Sastri, A.R., 2012.
636 Current state and trends in Canadian Arctic marine ecosystems: I. Primary production.
637 Clim. Change 115, 161–178. https://doi.org/10.1007/s10584-012-0496-3

638 Wassmann, P., Reigstad, M., 2011. Future Arctic Ocean seasonal ice zones and implications for
639 pelagic-benthic coupling. Oceanography 24, 220–231.
640 https://doi.org/10.5670/oceanog.2011.74

641 Yurkowski, D.J., Brown, T.A., Blanchfield, P.J., Ferguson, S.H., 2020. Atlantic walrus signal
642 latitudinal differences in the long-term decline of sea ice-derived carbon to benthic fauna
643 in the Canadian Arctic. Proc. R. Soc. B Biol. Sci. 287, 20202126.
644 https://doi.org/10.1098/rspb.2020.2126

645 Zeileis, A., Hothorn, T., 2002. Diagnostic checking in regression relationships. R News 2(3), 7-
646 10. https://CRAN.R-project.org/doc/Rnews

647 Zdanowicz, C., Krümmel, E.M., Lean, D., Poulain, A.J., Yumvihoze, E., Chen, J., Hintelmann,
648 H., 2013. Accumulation, storage and release of atmospheric mercury in a glaciated Arctic
649 catchment, Baffin Island, Canada. Geochim. Cosmochim. Acta 107, 316–335.
650 https://doi.org/10.1016/j.gca.2012.11.028

651 Zhang, Y., Jaeglé, L., Thompson, L., Streets, D.G., 2014. Six centuries of changing oceanic
652 mercury: Anthropogenic mercury in ocean. Glob. Biogeochem. Cycles 28, 1251–1261.
653 https://doi.org/10.1002/2014GB004939

654 Zheng, W., Chandan, P., Steffen, A., Stupple, G., De Vera, J., Mitchell, C.P.J., Wania, F.,
655 Bergquist, B.A., 2021. Mercury stable isotopes reveal the sources and transformations of
656 atmospheric Hg in the high Arctic. Appl. Geochem. 131, 105002.
657 https://doi.org/10.1016/j.apgeochem.2021.105002
658

FIGURES

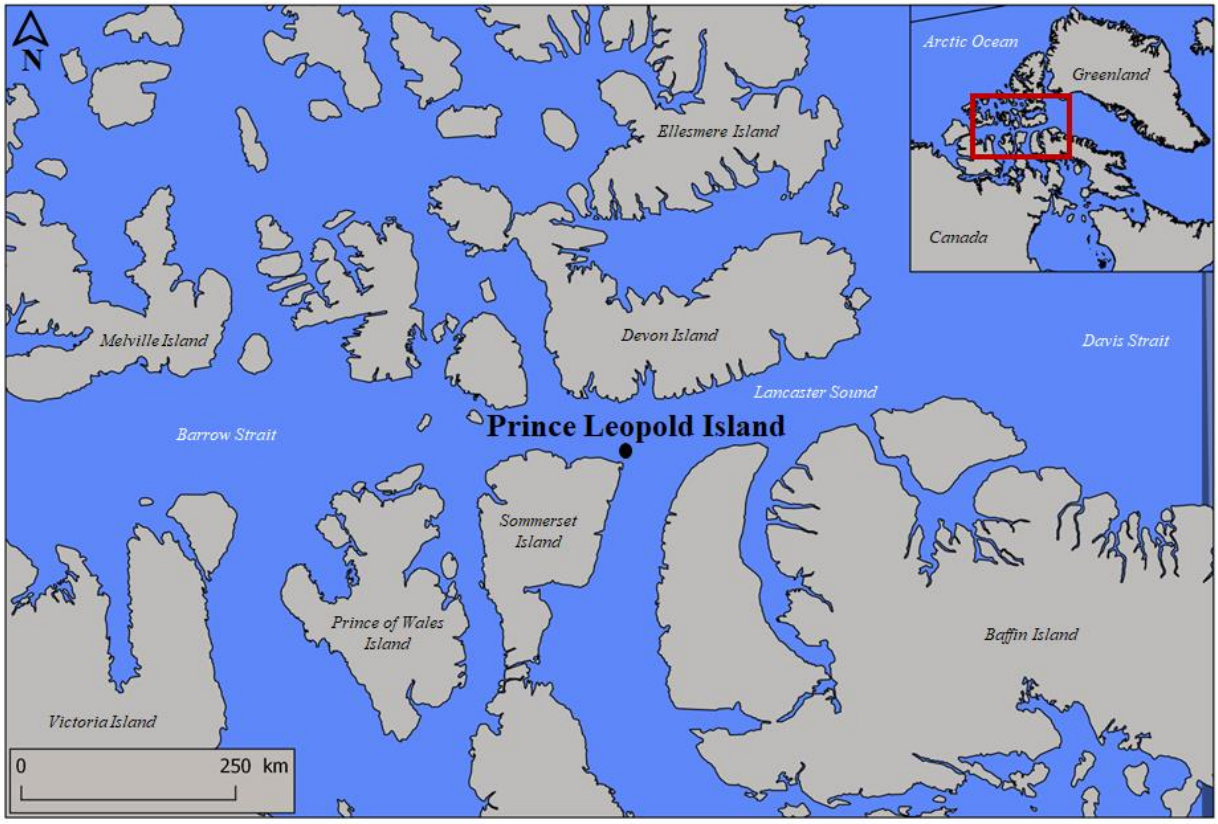
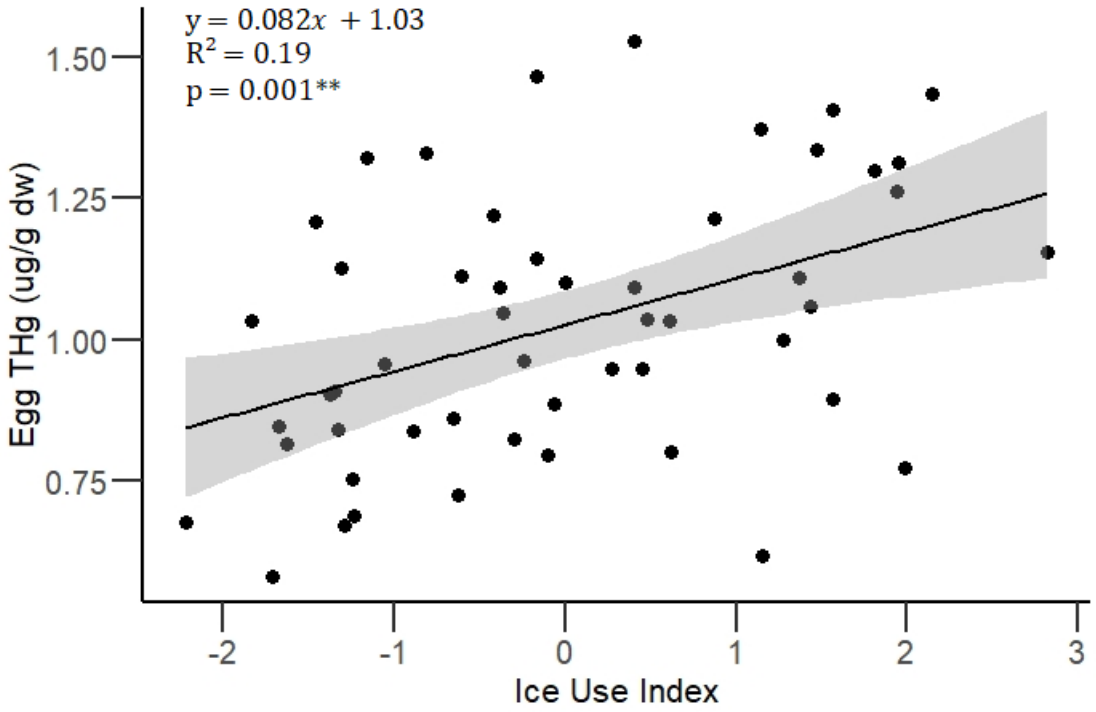


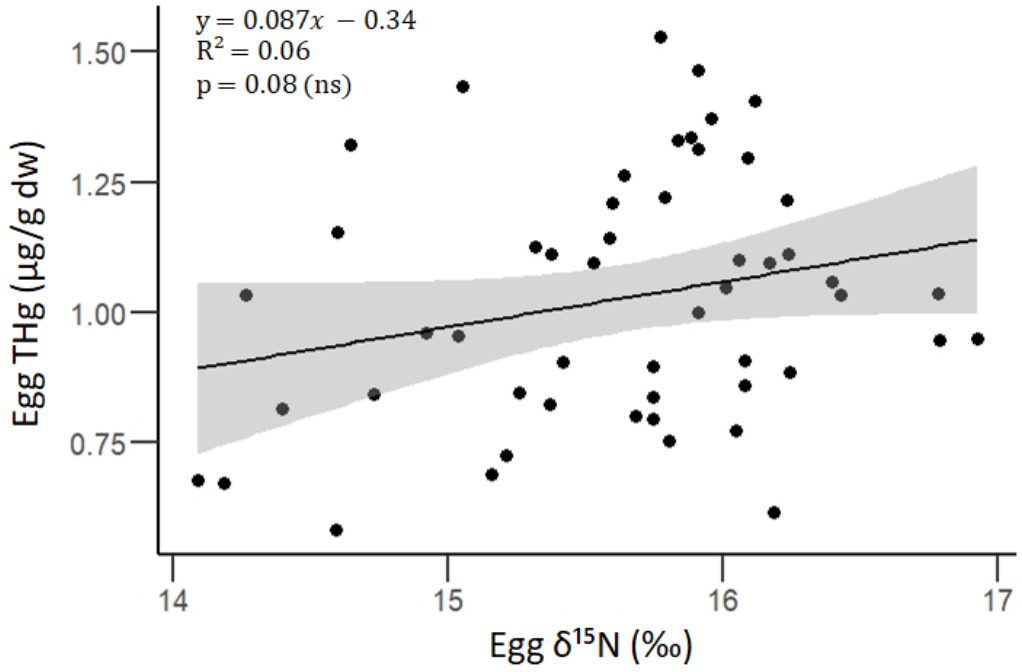
Figure 1. Location of Prince Leopold Island in the Canadian High Arctic



662

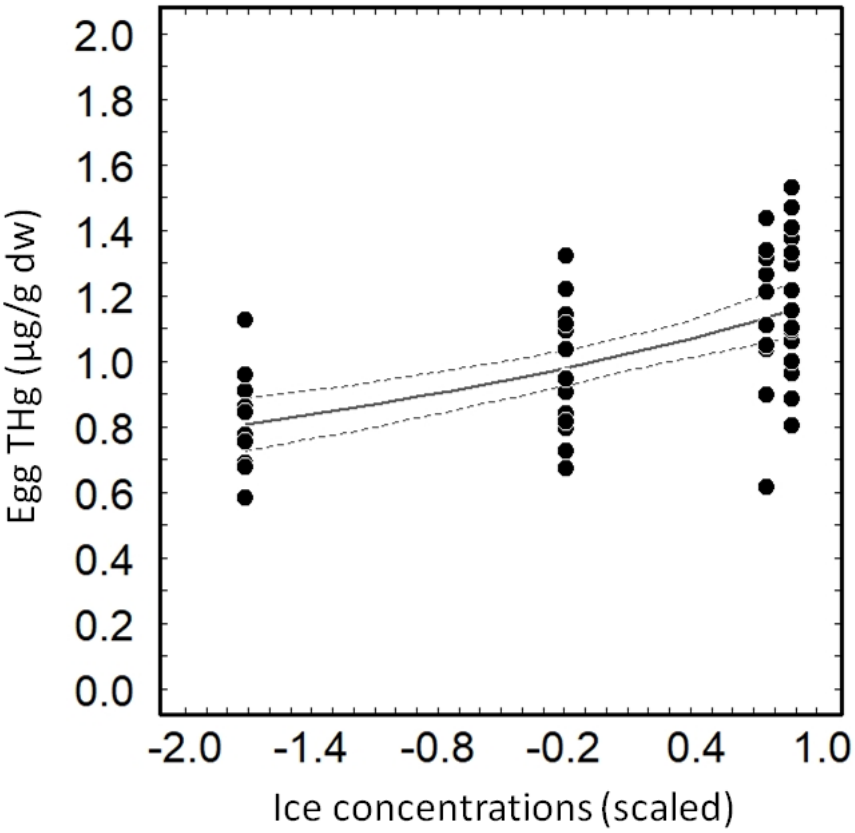
663 **Figure 2.** Relationship between total Hg concentrations measured in eggs of thick-billed murres
664 (*Uria lomvia*) from Prince Leopold Island (Nunavut, Canada) and Ice Use Index. Lower and higher
665 values of Ice Use Index correspond to the most pelagic and sympagic feeding association,
666 respectively.

667



668

669 **Figure 3.** Relationship between total Hg concentrations (µg/g dw) measured in eggs of thick-billed
670 murres (*Uria lomvia*) from Prince Leopold Island (Nunavut, Canada) and δ^{15}N values (proxy for
671 trophic position).



672

673 **Figure 4.** Egg total Hg concentrations of thick-billed murres (*Uria lomvia*) from Prince Leopold
674 Island (Nunavut, Canada) with varying ice concentrations. Ice concentrations were scaled for
675 statistical analyses (raw concentrations range from 17.02 to 97.96 %). Solid and dotted lines
676 represent the best-fitting model detected from construction of Generalized Linear Models (GLMs)
677 and the confidence interval, respectively (see "Material and Methods" for further details).

TABLES

Table 1. Ice conditions and egg parameters from thick-billed murres collected between 2010 and 2013 on Prince Leopold Island (Nunavut, Canada): trophic tracers include the Ice Use Index, carbon- ($\delta^{13}C$) and nitrogen- ($\delta^{15}N$) stable isotopes; total mercury (THg) concentrations (µg/g dw). $\delta^{13}C$ and $\delta^{15}N$ are proxies for bird feeding habitat and trophic position, respectively. Values represent means ± SD (n).

	2010	***2011***	***2012***	***2013***
Ice concentrations[1] (%)	99.0	33.4	99.0	79.8
Ice characteristics[2]	Patchy, large floes, leads with water access	Ice-free	Dense pack-ice, no leads, ice edge > 250 km away	Dense pack ice, ice edge 50 km away
Ice Use Index[3]	1.0 ± 1.1 (15)	-1.1 ± 1.1 (15)	0.7 ± 1.0 (15)	0.7 ± 0.7 (15)
H-Print (%)[4]	28.1 ± 20.8 (15)	82.5 ± 18.8 (15)	56.9 ± 27.3 (15)	63.4 ± 18.1 (15)
$\delta^{13}C$ (‰)	−19.7 ± 0.4 (11)	−20.3 ± 0.5 (11)	−19.3 ± 0.4 (15)	−20.3 ± 0.3 (15)
$\delta^{15}N$ (‰)	16.0 ± 0.5 (11)	15.3 ± 0.7 (11)	15.8 ± 0.5 (15)	15.4 ± 0.8 (15)
THg (µg/g dw)	1.1 ± 0.2 (15)	0.9 ± 0.2 (15)	1.2 ± 0.2 (15)	1.0 ± 0.2 (15)
Moisture (%)	73.3 ± 0.9 (15)	69.1 ± 2.5 (15)	73.7 ± 1.0 (15)	73.6 ± 1.0 (15)

[1]Ice concentrations represent the proportion of sea ice *versus* open water in a 100 km radius around the colony at the time of egg-laying (on June 15). Please refer to the "Material and Methods" for further details.

[2]Despite similar ice concentrations, 2010 and 2012 differed in their ice characteristics. Complete description of ice conditions is available in Cusset et al. (2019).

[3]The Ice Use Index indicates the degree of ice feeding association (values < and > 0 indicates low and high reliance on sympagic resources, respectively).

[4]H-Print (i.e., HBI-fingerprint) indicates the degree of association to ice-derived resources (from 0 to 100% for sympagic and pelagic association, respectively).

Table 2. AIC_c model ranking from statistical analyses of egg THg concentrations from thick-billed murres breeding on Prince Leopold Island (2010-2013). Models are Generalized Linear Models (GLMs) with a Gamma distribution and an inverse link function. Explanatory variables include: ice concentrations within 100 km around the colony («Ice»), the type of association to sea-ice and its derived resources (pelagic *versus* sympagic; «IceUse»), the trophic position («δ^{15} N») and their relevant interactions. Abbreviations: k, the number of parameters; AIC_c, Akaike's Information Criteria adjusted for small sample-size values; w_i, AICc weights (weights across all models sum to 1.00); LL, Log-Likelihood.

Models	**k**	**AIC_c**	**ΔAIC_c**	**w_i**	**LL**
THg ~ Ice*	**3**	**−16.43**	**0.00**	**0.83**	**11.46**
THg ~ δ^{15} N + IceUse + Ice	5	−12.79	3.64	0.13	12.05
THg ~ δ^{15} N + IceUse + δ^{15} N:IceUse + Ice + Ice :IceUse	7	−9.44	6.99	0.03	12.99
THg ~ IceUse	3	−7.04	9.38	0.01	6.77
THg ~ δ^{15} N + IceUse	4	−5.02	11.40	0.00	6.94
THg ~ δ^{15} N + IceUse + δ^{15} N:IceUse	5	−4.13	12.30	0.00	7.72
THg ~ δ^{15} N + IceUse + Ice:IceUse	5	−2.96	13.46	0.00	7.13
THg ~ δ^{15} N	3	0.18	16.60	0.00	3.16
THg ~ NULL	2	1.14	17.56	0.00	1.55

*The best model (in bold) is selected with the lowest AIC_cvalue.

CREDIT AUTHOR STATEMENT

Contributions.

Conception and design: FC, JF, GM, MM

Laboratory work and data acquisition: FC (trophic tracers, ice parameters), BB (mercury), GG (stable isotopes) and PM (ice analyses)

Data analysis: FC

Interpretation: FC, JC, JF, GM, MM, JP

First draft: FC, JC

Supervision: GM, JF

Revision and final approval: FC, JC, GM, MM, BB, JP, GG, PM, JF

Graphical Abstract