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Céline Albert

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La Rochelle University

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Prepared at the Institute Littoral, Environnements et Sociétés, UMR 7266

Presented by Céline ALBERT

Defended on September 8th 2020

Submitted for the degree of doctor of La Rochelle University

Specialty: Environment and Population Biology, Ecology

**EXPOSURE OF ARCTIC SEABIRDS TO POLLUTANTS AND THE
ROLE PLAYED BY INDIVIDUAL MIGRATORY MOVEMENTS AND
NON-BREEDING DISTRIBUTION.**

PhD supervisor: **Jérôme Fort**

Jury composition:

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THESE DE DOCTORAT DE LA ROCHELLE UNIVERSITE

École doctorale EUCLIDE
Préparée à l'Institut Littoral, Environnements et Sociétés, UMR 7266

Présentée par **Céline ALBERT**

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Pour l'obtention du grade de docteur de La Rochelle Université
Spécialité : Biologie de l'Environnement, des Populations, Écologie

**EXPOSITION AUX POLLUANTS DES OISEAUX MARINS
ARCTIQUES : SOURCE ET RÔLE DES DÉPLACEMENTS
MIGRATOIRES.**

Directeur de thèse : **Jérôme Fort**

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Table of contents

Preface	v
1. Acknowledgements	v
2. Professional acknowledgements	vii
3. Abstract	ix
4. Résumé	x
5. Publications	xi
5.1. Publications related to the doctoral thesis	xi
5.2. Publications not related to the doctoral thesis, published during the thesis:	xii
6. Other scientific activities	xii
6.1. Conference and workshop participation	xii
6.2. Teaching and supervising	xii
6.3. Grants	xiii
6.4. Fieldwork and laboratory work - contributions	xiii
6.5. Public communication	xiii
Chapter 1	1
Introduction	1
1. The Arctic, the remote area that got polluted	2
2. Emissions and transportation of Hg	3
2.1. Hg cycle	3
2.2. Hg transportation and introduction in the Arctic	5
2.3. Hg incorporation to the food web	6
3. Hg toxicity	8
4. Arctic seabirds and large-scale Hg contamination	9
4.1. Arctic seabirds' ecologies and Hg contamination	9
4.2. Hg contamination in Arctic seabirds: what do we know.	9
5. Spatial ecotoxicology	10
5.1. Spatial ecotoxicology through the use of non-lethal tissues	10
5.2. Hg contamination, migratory movements and wintertime	11
6. Thesis objectives and outline	12
Chapter 2	14
General methodology: biological sampling, laboratory and spatial analyses	14
1. ARCTOX	15
1.1. Sampling protocols	15
1.1.1. Blood sampling	17
1.1.1.1. Blood as a tool to monitor Hg contamination during the breeding season.	17
1.1.1.2. Blood sampling	17
1.1.2. Feather sampling	18
1.1.2.1. Hg excretion into feathers	18
1.1.2.2. Types of feathers sampled	19
1.2. Mercury (Hg) laboratory analysis	20
1.2.1. Blood and feathers samples preparation	20
1.2.2. Hg analysis	20
2. SEATRACK	22
3. ARCTOX and SEATRACK collaborations	24
Chapter 3	25
Are Hg contaminations seasonally constant?	25

1.	Hg contamination during the breeding period	26
1.1.	Interspecific variations	27
1.1.1.	Blood	27
1.1.2.	Body feathers	29
1.2.	Spatial variations	32
1.2.1.	Brünnich's guillemots	32
1.2.1.1.	Blood	32
1.2.1.2.	Body feathers:	33
1.2.2.	Black-legged kittiwakes	33
1.2.2.1.	Blood	33
1.2.2.2.	Body feathers	34
1.3.	Toxicity and risks	37
1.3.1.	Interspecific variations	37
1.3.2.	Brünnich's guillemots	38
1.3.3.	Black-legged kittiwakes	38
2.	Large-scale Hg contamination in the Arctic: seabirds as bioindicators of the marine ecosystem	39
2.1.	Hg concentrations in blood adjusted by $\delta^{15}\text{N}$.	39
2.2.	Results and discussion	40
2.3.	Conclusion	42
3.	Large-scale Hg contamination during the non-breeding period and Hg seasonality: study case of the Alcid family	43
3.1.	Introduction, hypotheses and predictions	43
3.2.	Results and discussion.	44
3.2.1.	Hg concentrations during the non-breeding season.	44
3.2.2.	Hg seasonality	45
3.2.3.	Pacific vs Atlantic	48
3.3.	Conclusion	48
4.	Summary of Chapter 3	50
Chapter 4		51
<i>Does seabird migration play a role on Hg exposure at the individual level?</i>		51
1.	Introduction, hypotheses and predictions	52
2.	Results and discussion	53
2.1.	How does faithfulness to wintering grounds impact mercury contamination?	53
2.2.	How does inter-annual variation in Hg contamination relates to spatial variations in migratory grounds?	56
3.	Conclusion	58
4.	Summary of Chapter 4	59
Chapter 5		60
<i>Spatial ecotoxicology: determining the spatial origin of Hg contamination</i>		60
1.	Are there hotspots of Hg?	61
1.1.	Introduction, hypotheses and predictions	61
1.2.	Results and interpretations	62
1.2.1.	Intraspecific analysis	62
1.2.2.	Multi-species analysis	67
1.3.	Comparison with abiotic monitoring	69
1.4.	Conclusion	72
2.	What is the origin of Hg and its spatial variability in the North-Atlantic Arctic?	73
2.1.	Introduction, hypotheses and predictions	73
2.1.1.	Hg isotopes: what, why, how?	73
2.1.2.	Hypotheses and predictions	74
2.2.	Tracking the source of Hg contamination: results and discussion	75

2.2.1.	Hg variations	75
2.2.2.	MDF ($\delta^{202}\text{Hg}$)	75
2.2.3.	Odd-MIF ($\Delta^{199}\text{Hg}$)	76
2.2.4.	Even-MIF ($\Delta^{200}\text{Hg}$)	77
2.2.5.	Carbon ($\delta^{13}\text{C}$) and Hg stable isotopes: terrestrial vs marine Hg inputs	78
3.	Conclusion	79
4.	Summary of chapter 5	81
Chapter 6		82
General discussion, limitations and perspectives		82
1.	Highlights of the doctoral work	83
2.	Seasonality in Hg contaminations	83
3.	Migratory movements of Arctic seabirds and their role in Hg exposure	84
4.	Limitations	86
5.	What about other pollutants?	88
6.	What's next?	89
7.	ARCTOX, what's next?	90
References		91
ANNEX		99
Annex 1		100
<i>species descriptions</i>		100
Annex 2		127
Annex 3		150
Papers		157
Paper 1		158
Paper 2		187
Paper 3		188
Paper 4		213
Paper 5		239
Paper 6		254

Preface

1. Acknowledgements

Twelve years ago, I got this crazy idea to quite my permanent job in a bank office to... go to the university and study ecology. I grew up in an environment where I could do anything I wanted...but where your social origin determines who you are and what you will do. So I thought for a very long time that I could absolutely never do scientific studies, that I was not smart enough for doing this and that even if I decided to try, I would have end up only at a “girl job”. The infamous “silent sexism”. But also probably because it was all we knew. So, thank you to my family for trying to do their best in a very complicated life.

14 years ago, I started a job that soon enough I started to hate. It made me realize I needed to do something meaningful for me, more into my believes. This was also a time during which I met the right people. From there, I decided it was time to be happy in my job, even if the people supposed to support me and believed in me did not.

Along the journey, I heard many of my closest friends telling me that I did all of this on my own. I am starting to understand what they mean, but they made this adventure much easier. And I want to thank you all. So be prepared, because it might be the longest acknowledgement section you have ever read.

So that you can follow my adventures, I will write my acknowledgments chronologically: from Paris to Vienna, Norway, and La Rochelle.

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During my bachelor, I ended up having one free semester. So I decided to take my chance abroad to (finally) learn (almost) proper English (I was so terrible before that...). Thanks to **Clotilde** who nicely recommended me few teams, I ended up in Austria, at the Lorentz Konrad Institute, in Vienna. I first want to thank **Herbert** and **Matteo** to welcoming me in their team. I learned a lot during my stay and had my very first scientific publication because of you. Thank you. Of course, I have a special thought for you, **Matteo**. Life has been incredibly unfair. But as **Herbert** said, you now live through the students you kindly supervised during your career. I also met one person who was particularly important during this internship: **Valeria**. Thanks for all the time spend together, the long talk, and the trips. Learning English with an Italian was a challenge, but I actually think you helped me learning much faster ☺ Hopefully, we will meet again soon. Thanks for everything my friend.

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3. Abstract

The Arctic, even far from intensive human activities, is contaminated by pollutants emitted at Northern mid-latitudes. Because of their physico-chemical characteristics, pollutants are transported over large distances through atmospheric or oceanic currents. Among them is mercury (Hg), a naturally occurring and non-essential trace element whose emissions increased since the 19th century because of human activities. This neurotoxic negatively impacts animals' health and induces behavioral changes, reproduction issues and in the most extreme case, death. The marine environment is particularly sensitive to Hg, which incorporates the food chain (under its toxic and methylated form – MeHg) in which its concentration increases from one trophic level to the other (e.g. biomagnification process) and accumulates within organisms (e.g. bioaccumulation process). Hence, long-lived top predators like seabirds, found at the end of the food chain usually show some of the highest contamination to Hg. They are commonly and efficiently used as bio-indicators of the health of their environment. Most of the current knowledge about Hg contamination in Arctic seabirds focused on the breeding period during which Hg was found to spatially vary, with usually higher Hg concentrations in the Canadian Arctic. During this period, which represents a part of the year only, seabirds aggregate in colonies for reproduction where they are more easily accessible. However, at the end of this period, seabirds migrate to overwinter mostly in open seas, outside of the Arctic. A previous study on a little auk (*Alle alle*) population breeding in East Greenland found that Hg concentrations were higher during the non-breeding period than during the breeding period, with carryover effects on the following reproduction. In the present doctoral work, based on a multi-species and multi-colony approach, we studied winter Hg exposure and the role of seabird migration in their contamination to Hg at large spatial scale. We found a seasonality in Hg concentrations allowing us to extend the results found in little auks to several species and at a larger spatial scale. We also found that this seasonality was spatially different with some of the highest variations for seabirds breeding in the West Atlantic (West Greenland and Canadian Arctic). We therefore proposed that such variations were due to seabird's migration and the areas they overwintered at. To test such hypotheses, we used seabirds as bio-indicators of winter Hg contamination through the North-Atlantic Arctic. More specifically, we combined Hg measurements with geolocators devices to track the spatial origin of winter Hg contamination. We found an east-west increase in Hg concentrations allowing us to extend the results found during the breeding period to the winter period, through the entire North-Atlantic Arctic marine region. Results of the present doctoral work allow us to conclude that beyond migration, seabird distribution during the breeding and non-breeding periods drive their contamination to Hg.

Key words: Arctic, seabirds, migration, contaminants, spatial ecotoxicology, mercury, marine ecology.

4. Résumé

L'Arctique, bien que région éloignée de toutes activités anthropiques intensives, est contaminé par les polluants émis à de plus basses latitudes. En effet, les caractéristiques physico-chimiques de certains polluants leur permettent d'être transportés sur de longues distances, via les courants atmosphériques ou océaniques. Parmi eux se trouve notamment le mercure (Hg), élément trace non-essentiel naturellement émis dans l'environnement mais dont les émissions d'origine anthropiques ont fortement augmenté depuis le 19^{ème} siècle. Ce neurotoxique engendre chez les espèces animales des troubles comportementaux, mais aussi des problèmes de reproduction et dans les cas extrêmes, la mort. Le milieu marin est particulièrement sensible à la contamination au Hg. En effet, une fois dans l'environnement marin, le Hg intègre la chaîne alimentaire (sous sa forme toxique méthylée - MeHg), dans laquelle sa concentration augmente d'un niveau trophique à l'autre (processus de bioamplification) et/ou accumule au sein des organismes (processus de bioaccumulation). Ainsi, les prédateurs supérieurs longévifs tels que les oiseaux marins, en fin de chaîne alimentaire, présentent les concentrations de Hg parmi les plus élevées. Ils sont largement et efficacement utilisés comme bio-indicateurs de la contamination au Hg de leur environnement. La majeure partie des connaissances actuelles sur la contamination au Hg des oiseaux marins Arctique concerne la saison de reproduction, durant laquelle les concentrations varient spatialement, avec notamment les concentrations les plus élevées trouvées en Arctique Canadien. Durant cette période, qui ne représente qu'une partie de l'année, les oiseaux se reproduisent à terre et sont donc plus facilement accessibles. Mais une fois la saison de reproduction terminée, la majorité des oiseaux marins migrent en haute mer, en dehors de l'Arctique. Une étude précédente a montré qu'une population de mergules nains (*Alle alle*) se reproduisant à l'Est du Groenland a des concentrations de Hg plus élevées en période de non-reproduction qu'en période de reproduction, avec des effets délétères sur la reproduction suivante. Au cours de ce travail de thèse, et grâce à une approche multi espèces et multi colonies, nous nous sommes intéressés au rôle de la migration des oiseaux marins sur leur contamination au Hg et ce à l'échelle de l'Arctique. Nos résultats montrent une saisonnalité dans la contamination au Hg, nous permettant d'étendre les résultats mis en avant pour les mergules nains à de nombreuses autres espèces d'oiseaux marins. Nous avons également trouvé que cette saisonnalité varie spatialement avec des variations saisonnières plus importantes pour les populations se reproduisant en Atlantique Ouest (Ouest du Groenland et Est du Canada). Ces résultats nous ont amenés à faire l'hypothèse que cette variation était une conséquence de la migration des oiseaux marins et de la distribution de leur zone d'hivernage. Pour tester cette hypothèse, nous avons utilisé les oiseaux comme bio-indicateurs de leur environnement. Pour cela, nous avons combiné des analyses de Hg avec du biologging afin de retracer l'origine spatiale de la contamination hivernale au Hg. Nos résultats montrent une augmentation des concentrations de Hg suivant un gradient est-ouest nous permettant ainsi d'étendre les résultats des précédentes études concernant la période de reproduction à l'hiver et à l'ensemble des régions marines à l'échelle de l'Atlantique Nord - Arctique. Ainsi, les résultats du présent travail de thèse nous permettent de conclure qu'au-delà de leur migration, ce sont les zones de reproduction et d'hivernages des oiseaux marins arctiques qui conditionnent leur contamination au Hg.

Mots clés : Arctique, oiseaux marins, migration, écotoxicologie spatiale, mercure, écologie marine.

5. Publications

5.1. Publications related to the doctoral thesis

Paper 1: Albert C., Renedo M., Bustamante P., Fort J., 2019. Using blood and feathers to investigate large-scale Hg contamination in Arctic seabirds: a review. *Published in Environmental research*, 177-108588 - <https://doi.org/10.1016/j.envres.2019.108588>

Paper 2: Fort J., Albert C., Strøm H., Helgason H.H., Brault-Favrou M., Amélineau F., Anker-
Nilssen T., Bustnes J.O., Cherenkov A., Danielsen J., Daunt F., Descamps S., Dietz R., Elliott K.,
Erikstad K.E., Ezhov A., Fauchald P., Gavrilov M., Gilchrist G., Gilg O., Hansen E.S, Hanssen S.A.,
Hatch S., Helberg M., Huffeldt N.P., Jakubas D., Jónsson J.E., Kitaysky A., Krasnov Y., Langseth M.,
Leclaire S., Thórarinnsson T.L., Lorentsen S.H., Lorentzen E., Mallory M., Merkel F., Moe B.,
Montevecchi W.A., Mosbech A., Olsen B., Orben R., Pratte I., Provencher J., Ragnarsdóttir S.B.,
Robertson G.J., Sagerup K., Semashko V., Systad G.H., Takahashi A., Tertitski G., Thompson P.,
Hallgrímsson G.T., Tolmacheva E., Will A., Wojczulanis-Jakubas K., Bustamante P., Grémillet D.
Tracking mercury in Arctic seabirds and marine food webs.

In preparation. Draft not included in the annex. Journal not determined yet.

Paper 3: 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.,
Gavrilov M., Golubova E., Grémillet D., Hatch S., Jakubas D., Kitaysky S., Kolbeinnsson Y., Krasnov Y.,
Lorentsen S-H, Lorentzen E., Mallory M.L., Merkel B., Merkel F.R., Montevecchi W., Mosbech A.,
Olsen B., Patterson A., Provencher J., Huffeldt N.P., Plumejeaud C., Pratte I., Reiertsen T.K., Renner H.,
Rojek N., Romano M., Strøm H., Systad G.H., Thórarinnsson T.L., Will A.W., Wojczulanis-Jakubas K.,
Bustamante P., Fort J. **Seasonal variation of mercury contamination in Arctic seabirds: a pan-arctic
assessment.** *Submitted in The Science of the Total Environment.*

Paper 4: Albert C., Moe B., Bråthen V.S., Anker-
Nilssen T., Cherenkov A., Christensen-
Dalsgaard S., Danielsen J., Descamps S., Erikstad K.E., Gavrilov M., Hanssen S.A., Jónsson J.E.,
Kolbeinnsson Y., Krasnov Y., Helgason H.H., Langset M., Lorentzen E., Olsen B., Reiertsen T.K., Strøm
H., Systad G.H., Tertitski G., Thompson P. M., Thórarinnsson T.L., Bustamante P., Fort J. **Interannual
variations in migratory movements impact individual seabird exposure to contaminants.**
Submitted in Marine Ecology Progress Series

Paper 5: Albert C., Moe B., Bråthen V.S, Helgason H.H., Åstrom J., Amélineau F., Angelier
F., Anker-
Nilssen T., Chastel O., Christensen-Dalsgaard S., Danielsen J., Descamps S., Elliott K.,
Erikstad K.E., Ezhov A., Gavrilov M., Grémillet D., Krasnov Y., Langset M., Lorentsen S.-H., Merkel

B., Olsen B., Patterson A., Reiertsen T.K., Systad G.H., Thompson P., Thórarinnsson T.L., Brault Favrou M., Bustamante P., Strøm H., Fort J. **Seabirds as bioindicators of winter mercury distribution through the North-Atlantic Arctic.**

First draft formatted for journal requiring 2000-4000 words. Journal not determined yet.

Papier 6: Renedo M., Amouroux D., **Albert C.**, Bérail S., Bråthen V.S., Gavriilo M., Grémillet D., Helgason H.H., Jakubas D., Mosbech A., Strøm H., Tessier E., Wojczulanis-Jakubas K., Bustamante P., Fort J. **Contrasted spatial and seasonal trends of methylmercury exposure pathways of Arctic seabirds: combination of large-scale tracking and stable isotopic approaches**

Submitted in Environmental Science and Technology.

5.2. Publications not related to the doctoral thesis, published during the thesis:

Courchamp F., Jarić I. **Albert C.**, Meinard Y., Ripple W. J., Chapron G., 2018. The paradoxical extinction of the most charismatic animals. 2018. *Published in PLoS Biology* 16(‘): e2003997. DOI: 10.1371/journal.pbio.2003997

Albert C., Luque G. M., Courchamp F., 2018. The twenty most charismatic species. *Published in PLoS ONE* 13(7):e0199149. DOI:1371/journal.pone.0199149

6. Other scientific activities

6.1. Conference and workshop participation

- SEATRACK workshop, 19-21 March 2018, Trondheim, Norway. Oral communication
- Future directions in French polar pollution research, 14 & 15 October 2019, Toulouse, France. Oral communication

6.2. Teaching and supervising

- Teaching ecology to bachelor students:
 - fall 2018: 26 hours,
 - fall 2019: 13 hours.
- **Co-Supervision during spring 2019:** Thibault Le Verge (Master 2 student). La Rochelle University. "Migratory seabirds, Hg vectors at a large spatial scale". Co-Supervision: Jérôme Fort, Céline Albert, Pierrick Bocher.

6.3. Grants

- Mobility grant from La Rochelle University (1200€) for 4 months stay at the Norwegian Institute for Nature Research, Trondheim, Norway
- Arctic Field Grant, 2019, "Exposure of arctic seabirds to pollutants: what is the role played by individual migratory movements and non- breeding distribution? (ExpoBird) », October 2018 ($\approx 8000\text{€}$ (79000 NOK)).

6.4. Fieldwork and laboratory work - contributions

- Fieldwork
 - Little auks field work in Kap Hoegh, Greenland (8 July to 21 August 2018)
 - Common eiders and skuas (great, Arctic and long-tailed) in Ny-Ålesund, Svalbard (13 June to 18 July 2019), in collaboration with the Norwegian Institute for Nature Research, Trondheim (Norway).
- Laboratory work
 - Hg and $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ isotopes analyses: All samples (feathers and blood) were analyzed for Hg, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ isotopes. Even if not highlighted in the present doctoral work, all feathers were analyzed for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ during the doctoral work by Maud Brault-Favrou, Jérémy Lemaire (during the first year of my thesis) and myself. All Hg, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ analysis in blood and highlighted in **Chapter 3** were performed by Maud Brault-Favrou. Some analyses were performed before the doctoral work by several students.
 - Hg isotopes analyses: preparation of samples was realized by both Marina Renedo and myself. All the Hg isotopes analyses were performed by Marina Renedo during her post-doctoral contract (lead author of **Paper 6**).

6.5. Public communication

- TPs Sandwichs: public communication about pollutants and Arctic seabirds (21 August 2018)
- Fête de la Science : public communication about pollutants (Hg and microplastics) " La contamination des oiseaux marins en Arctique", co-presented with F. Amélineau, at [La Médiathèque](#), La Rochelle. 18 October 2018
- Picture and scientific exhibition: " **Polar Ecology, from the Arctic to the Antarctic**" at the Museum National d'Histoire Naturelle, La Rochelle, from June to December 2019. Scientific coordination and co-management of the project. More information: <https://ecologie-polaire.univ-lr.fr/>

Chapter 1

Introduction



1. The Arctic, the remote area that got polluted

The Arctic, the northernmost area of the Earth, is a polar region characterized by extreme climate and life conditions. At such latitudes, the sun conditions are such that the Arctic experiences long periods of dark (i.e. Polar night) and light (i.e. midnight sun), resulting in harsh climate, short summer and extensive snow and ice cover. Still, even with such particular conditions, numerous species, including human beings, have adapted to survive in this environment. However, human activities have induced fast environmental changes in the Arctic, rapidly altering species' habitats and therefore making them extremely sensitive to environmental modifications.

Since the industrial revolution in the middle of the 20th century, human activities have released greenhouse gases (GHG), resulting in an increase in the global temperature (ACIA 2004, AMAP 2011a). In the Arctic, the temperature has risen at almost twice the rate of the rest of the world, causing a decrease in sea ice cover. For example, over the past 30 years, the average extent of the Arctic summer sea ice has decreased by 15-20% (ACIA 2004, AMAP 2011a), negatively impacting both wildlife and local communities relying on this habitat. The most infamous example concerns polar bears *Ursus maritimus* whose survival highly depends on sea ice. But, with the decrease of the sea ice extent, their favorite preys (e.g. seals) are no longer available inducing a drastic population decrease (Stern & Laidre 2016). Beyond the impacts on local life, the decrease of sea ice has created new shipping roads resulting in an increase of the traffic, the access to natural resources and a subsequent increase of associated pollutions.

Human activities have also released pollutants (e.g. Persistent Organic Pollutants - POPs, heavy metals) in the environment. In comparison to global emissions, pollutants have been emitted at low concentrations in the Arctic (AMAP 2004). For example, PCB (Polychlorinated Biphenyls) is only emitted at a low level in Scandinavia and Russia, while there are no emissions of DDT (Dichlorodiphenyltrichloroethane - AMAP, 2004) or mercury (Hg - AMAP, 2012). Hence, as the Arctic is located far away from major human activities, the high concentrations of pollutants measured in Arctic wildlife mainly result from long-range transportation activities by atmospheric, oceanic and riverine currents (AMAP 2004, 2012). The ability of Hg to be transported over large distances, to incorporate food web and to subsequently cause deleterious effects on living organisms has led mercury (Hg) to become a pollutant of high interest and the focus of the present doctoral work.

2. Emissions and transportation of Hg

2.1. Hg cycle

Mercury (Hg) is a non-essential element naturally emitted in the environment by volcanoes, forest fires or weathering of rock. However, around 31% of the actual Hg released in the atmosphere comes from anthropogenic emissions with the major sources coming from the combustion of fossil-fuel (coal) in power plant (Pacyna et al. 2006, Wilson et al. 2006, Pirrone et al. 2010), followed by artisanal gold mining (Pirrone et al. 2010). Anthropogenic Hg is mostly emitted in mid-northern latitudes, but its volatility and solubility allow it to be transported through atmospheric or oceanic currents (Fig. 1.1). In the atmosphere, mercury is mostly found under its inorganic and gaseous form Hg (0) (~98%), which can be transformed into the inorganic form Hg (II). This reactive Hg (II) can then either be deposited and/or transformed into its methylated and highly toxic form (methylmercury - MeHg). The aquatic environment is the most sensitive to MeHg as it contains organisms responsible for Hg methylation (see below). It is then under its methylated form that mercury is the most bioavailable to enter the food chain (AMAP, 2011b).

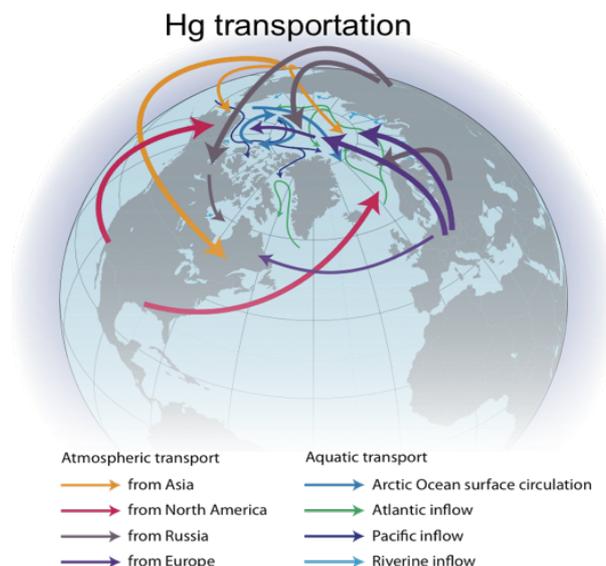


Figure 1.1: extracted from AMAP 2012 – p. 8

Mercury (Hg): a little bit of history

In the industry, Hg was early discovered and used for instance as a fungicide or a catalyzer. Then, it was disseminated in crops for years, poisoning both wildlife (through seed consumption) and humans (through bread consumption). But it took many years and one particular disaster before the link between Hg and its toxicity was made. In 1907 in Minamata (Japan), a petrochemical industry used Hg as a catalyzer, and started in 1932 to release it in the Bay of Minamata. The local population started to show signs of an unknown neurological disease, and the first report about it was published in 1956. It was only in 1968 that the Japanese government acknowledged that Hg was responsible for the so-called Minamata disease. Thus, even if Hg toxicity was first described in 1865 (Edwards 1865), it was only in the late 20th century that Hg was recognized as a neurotoxic with irreversible effects on wildlife and human beings (Grandjean et al. 2010). In 2009, the UN Environment (United Nation Environmental Program) initiated a global project for Hg assessment. Since then, the Minamata Convention, aimed at decreasing and mitigating global Hg emissions, was set up in 2013 in order to protect the health of the environment and populations (UNEP 2013). To this date, this convention has been signed by 128 countries, ratified by 113 of them since then (Fig. 1.2) and entered into force in 2017.

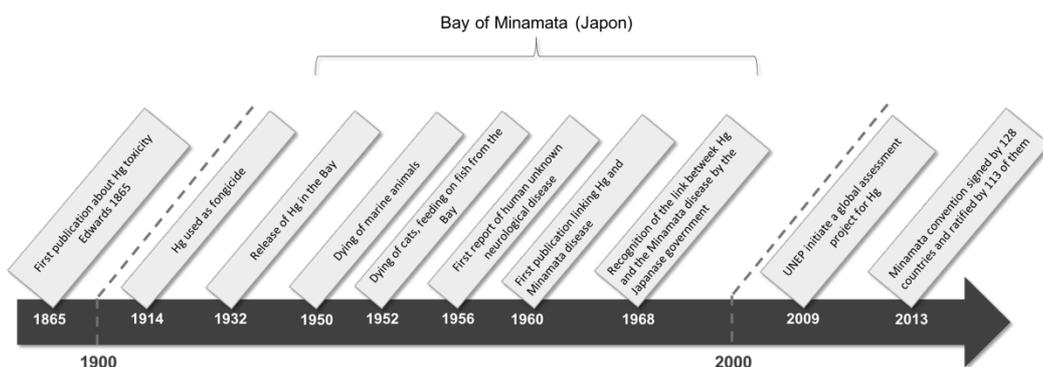


Figure 1.2: chronology of the discovery of Hg toxicity, based on Grandjean et al. 2010 © Céline Albert

2.2. Hg transportation and introduction in the Arctic

Once Hg enters atmospheric currents, it can then be deposited in the Arctic. Such deposition is enhanced by Atmospheric Mercury Depletion Events (AMDEs – Schroeder et al., 1998) during Spring. When deposited in the Arctic ocean, Hg becomes available for reemission into the atmosphere (under its Hg(0) form) or involved into the biochemistry of the Arctic Ocean (AMAP, 2011) (Fig. 1.4). There, it is either stored into the sediments, or is bioavailable for transformation into its toxic methylated form (MeHg). Beyond those deposits, Hg can also be transported to the Arctic through the Pacific and Atlantic Oceans currents from lower latitudes, at a slower rate. Indeed, while the residence time of Hg in the atmosphere varies from 0.7 to 1.4 years, the residence time in the ocean last around 27 years (AMAP, 2011). Hence, even with a decrease of Hg emissions in the ocean, past Hg emissions can still be transported to the Arctic.

When deposited on terrestrial environments, Hg is assimilated by the vegetation and transferred to both tundra and boreal peat soils where it accumulates. During spring and the subsequent snowmelt, peat soil Hg is remobilized and transferred to the Arctic Ocean through riverine currents (Sonke et al., 2018) (Fig. 1.3).

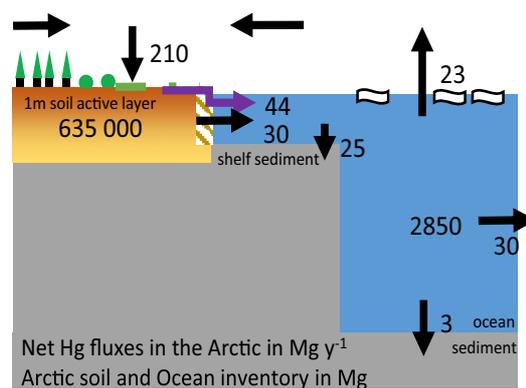


Figure 1.3: extracted from Sonke et al., 2018. Net fluxes (Mg g⁻¹) in Arctic Hg cycle with exchanges between the different terrestrial, marine, atmospheric and riverine (purple arrow) reservoirs. The hatched area represents the coastal erosion Hg flux.

2.3. Hg incorporation to the food web

Once Hg enters the aquatic environment, it can either be assimilated by phytoplankton or by consumers of bacteria responsible for Hg methylation. From then on, it enters the food chain and its concentration increases from one trophic level to the other (biomagnification process - Fig. 1.4). Most of species have a Hg excretion path (urea, feathers, hair, faeces, see **Paper 1** for a review), but the excretion rate can be lower than the intake rate. In addition, while some internal organs or tissues can remobilize Hg during excretion (e.g. liver, blood), some others cannot and therefore store pollutants (e.g. kidney, brain - bioaccumulation process - Kim et al., 1996). Therefore, by being long-lived species, in addition to be at the top of the food chains, marine mammals or seabirds present some of the highest wildlife Hg concentrations (Fig. 1.4), making them particularly sensitive to the toxicity of this pollutant (Dietz et al. 2013).

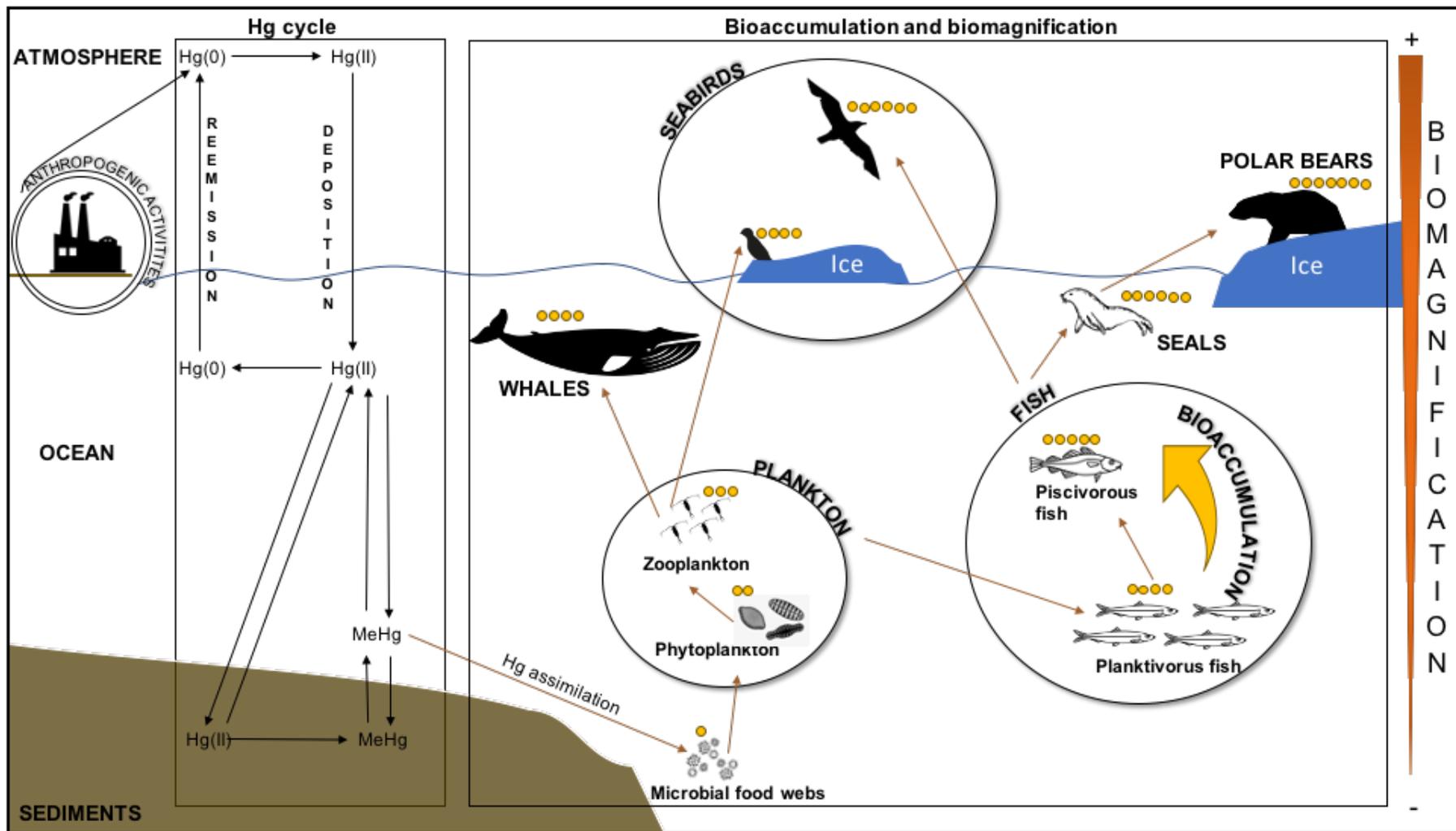


Figure 1.4: Schematic representation of the Hg cycle in Arctic aquatic environment, biomagnification (i.e. Hg concentration increases from one trophic level to the other) and bioaccumulation (i.e. [Hg] increases within individuals over time represented by the gold dots). © Céline ALBERT

3. Hg toxicity

Hg induces deleterious effects on numerous marine species like mammals or birds (Wolfe et al. 1998, Scheuhammer et al. 2007, Crump & Trudeau 2009, Grandjean et al. 2010, Ackerman et al. 2016). As a neurotoxic, it affects the nervous system, the behavior but also the reproduction. For example, experimental studies on wildlife mammals have demonstrated visual and sensory deficits (Wolfe et al., 1998). In birds, Hg contamination has been linked to changes of behavior, leading to a reduction of food intake, subsequently impacting survival (Wolfe et al., 1998). Hg exposure has also been linked to a diminution of the reproduction effort with fewer laid eggs in higher contaminated individuals (Heinz 1979, Barr 1986). In addition, high Hg concentration in eggs, in which Hg is excreted during the reproduction process, has been linked to a low breeding success (Heinz 1974, Wolfe et al. 1998, Scheuhammer et al. 2007) or the alteration of chick behavior (Heinz 1979). In extreme cases, Hg contamination induces death (Fig. 1.5 – Mergler & Lebel 2001).

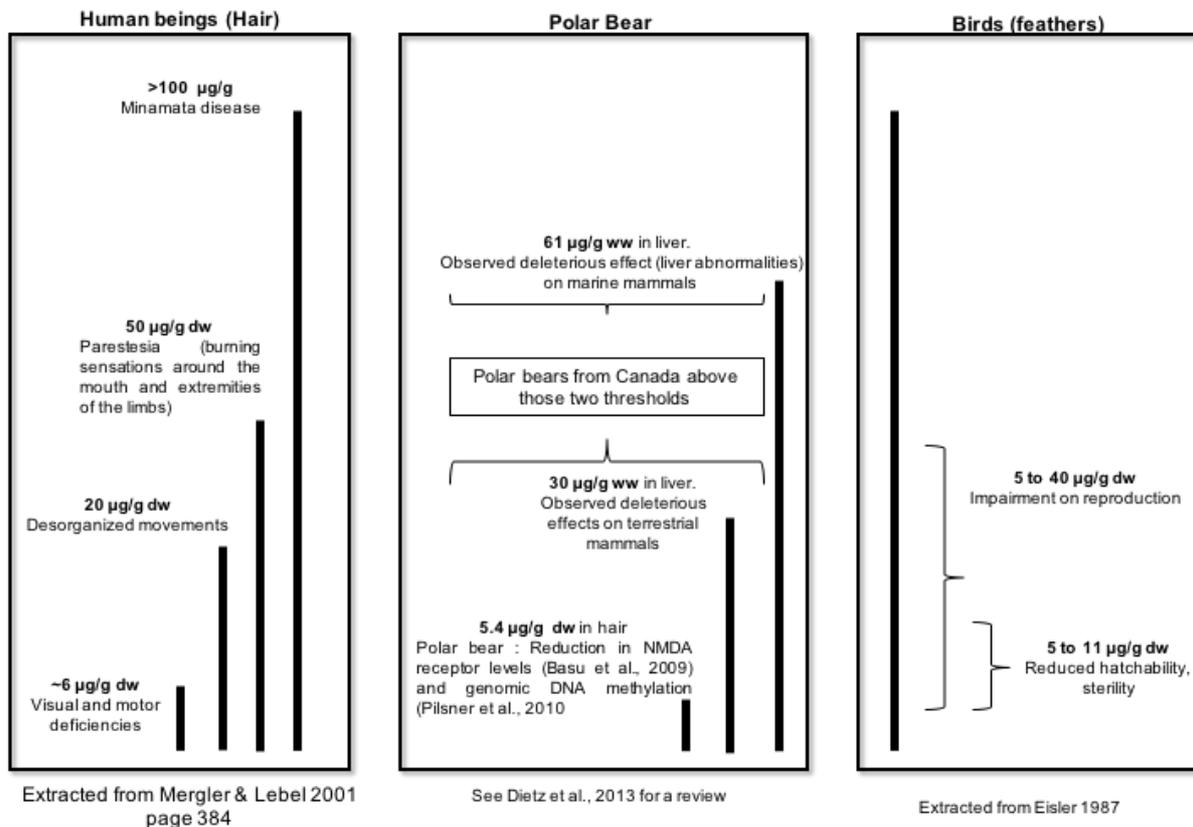


Figure 1.5: Thresholds of deleterious effects of Hg. Human beings, polar bears and seabirds have been represented as examples

4. Arctic seabirds and large-scale Hg contamination

4.1. Arctic seabirds' ecologies and Hg contamination

Arctic seabirds, defined as top predators breeding in the Arctic, are particularly exposed to environmental changes and Hg toxicity (Dietz et al. 2013, Ackerman et al. 2016). Among the most numerous top predators in the Arctic, we mostly find them breeding in large colonies (i.e. most of the auks, some gulls and anatids). Some species are endemic from the Pacific region (i.e. red legged kittiwakes *Rissa brevirostris*, murrelets, auklets), the Atlantic region (i.e. razorbill *Alca torda*) or distributed in both Arctic areas (i.e. Brünnich's guillemots *Uria lomvia*, common guillemots *Uria aalge*, black-legged kittiwake *Rissa tridactyla*). Beyond those different distributions, Arctic seabirds rely on different diets and/or habitats. For example, gulls are mostly piscivorous, while anatids mostly feed on crustacea. But dietary differences are also found within families. While large gulls are piscivorous or omnivorous (Gabrielson & Lincoln 1959), small gulls like black-legged kittiwakes preferentially feed on small fish or zooplankton (Hobson et al. 2002). Within the auk family (hereafter called Alcids), large species like Brünnich's and common guillemots mostly rely on fish, while small species like little auks *Alle alle* mostly rely on zooplankton (Gaston & Jones 1998) (see **Annex 1** for details on the species studied in this thesis). Finally, Arctic seabirds cover numerous marine environments. For example, some species preferentially feed in coastal areas and on benthic prey (e.g. common eiders) (Hobson et al. 2002, Waltho & Coulson 2015), while others preferentially feed on pelagic areas, feeding at the surface (e.g. black-legged kittiwakes) (Kotzerka et al., 2010) or by diving (i.e. Brünnich's guillemots, common guillemots) (Gaston and Jones, 1998). Thus, Arctic seabirds cover many different ecologies and will therefore face different exposure to Hg. Therefore, using Arctic seabirds as bio-indicators of Hg contamination with a multi-species approach will allow to cover a wide range of environments and exposure.

4.2. Hg contamination in Arctic seabirds: what do we know.

Hg contamination in Arctic seabirds has been extensively studied, with a focus on specific species or tissues. Out of the 42 species for which Hg contamination has been studied in the Arctic (**Paper 1**), three families (and species) received most of the attention: the larids (mostly the black-legged

kittiwake), the alcids (mostly the Brünnich's guillemot) and the anatids (mostly the common eider *Somateria Mollissima*), followed by the glaucous gulls *Larus hyperboreus* and the northern fulmars *Fulmarus glacialis* (extracted from **Paper 1**). Historically, those species started to be monitored early in the Canadian Arctic (Mallory & Braune 2012). Indeed, after witnessing the decrease of seabird populations, Canadian researchers started to collect egg samples on Prince Leopold Island (High Canadian Arctic) in 1975, to study potential links between the decrease of those species and contaminants (Braune et al. 2010, Mallory & Braune 2012). Eggs have been favored because they are easily collected and transported, represent a limited loss for the breeding populations and give essential information about the relatively local Hg contamination of females (Mallory & Braune, 2012 - see **Paper 1** for details). Internal tissues have also been extensively studied. Indeed, as seabirds are consumed by local communities, there was an early need to assess Hg risks coming from local food (Mallory et al. 2018).

Thanks to these studies, Hg contamination has been particularly abundant in the Canadian Arctic, mostly through the study of the internal organs and eggs (**Paper 1**). Beyond the crucial information they provide, those tissues have limitations (e.g. sampling endangered species) and raise ethical questions for large-scale studies. Therefore, it appears important to rely on non-lethal tissues for large-scale Hg studies such as blood and feathers, which provide interesting and complementary information (e.g. local or seasonal Hg contamination) (see Fort et al. 2014, **Paper 1**). In addition, large-scale Hg studies require homogeneous methods, in order to have comparable tissues and to limit statistical and biological biases.

5. Spatial ecotoxicology

5.1. Spatial ecotoxicology through the use of non-lethal tissues

The study of different animals (e.g. seabirds, polar bears) or seabird tissues (e.g. egg and liver) has demonstrated spatial differences with higher Hg concentrations in East Canada than in East Greenland and European Arctic (Borgå et al. 2006, Provencher et al. 2014, AMAP 2018 2018). To confirm the use of blood and feathers as reliable tissues in large-scale Hg monitoring and assessment in Arctic seabirds, we carried out a review of the literature (**Paper 1**). At the pan-Arctic scale, blood and

feathers provided additional information about the Pacific region. The Canadian Arctic and the Pacific region showed higher Hg concentrations compared to Greenland and the European Arctic, confirming and completing the existing knowledge. In the Atlantic region, Hg concentrations increased with an east-west gradient, supporting the existing knowledge provided by other seabird tissues (internal organs, Borgå et al., 2006; eggs, Provencher et al., 2014), or by Arctic mammals (AMAP 2018)(AMAP 2018)(AMAP, 2018)(AMAP, 2018a). Thus, blood and feathers showed their effectiveness in spatial ecotoxicology studies by confirming the existing knowledge and providing new inputs (e.g. new species and areas). However, some areas (blood: Russia, West Greenland, Aleutian Islands; feathers: Russia, Canadian Arctic) and species (blood; e.g. alcids) are still underrepresented at both spatial and temporal (i.e. seasonality) scales and those gaps need to be filled in order to fully comprehend the risks of Hg contaminations in the Arctic.

5.2. Hg contamination, migratory movements and wintertime

Most of the knowledge about Hg contamination in Arctic seabirds concerns the breeding period. Indeed, as seabirds aggregate in land for reproduction, we can access the colonies and collect data to monitor Hg exposure and potential impacted traits (e.g. survival, breeding success). However, the knowledge about contamination during the non-breeding period is scarce as seabirds migrate towards areas where they are less accessible.

Arctic seabirds have a high inter- and intraspecies variability in winter migratory movements and distribution. For example, in the Barents Sea, the common guillemots winter close to their breeding colonies while Brünnich's guillemots extend their winter migration to the North Atlantic (Steen et al. 2013)(Steen et al. 2013). Anatids also show different migratory strategies within a same species or population. For example, common eiders breeding in the High Arctic (e.g. Kongsfjorden) migrate south during winter (e.g. Iceland or Norway; Hanssen et al. 2016)(Hanssen et al. 2016), while individuals from the Low Arctic (e.g. Tromsø) are resident throughout the year (Bustnes and Erikstad, 1993). In addition, some highly migratory species from different populations winter in the same areas. For example, the Arctic terns *Sterna paradisaea* breeding in Northeast Greenland and Iceland overwinter in Antarctica, while the long-tailed skuas *Stercorarius longicaudus* breeding in Northeast Greenland and Svalbard

overwinter in the Southern Hemisphere (Gilg et al. 2013). Moreover, intrapopulation variations were observed. For instance, great skuas *Stercorarius skua* breeding in Bjørnøya were observed wintering off Europe, Canada or North Africa (Leat et al. 2013). Overall, Arctic seabirds rely on many different winter environments during which they might be exposed to diverse levels of contaminants (Fort et al. 2014, see Fleishman et al. 2019). A previous study of a Greenlandic population of little auks found that Hg concentrations during the non-breeding period were ~3.5 higher than during the breeding period (Fort et al., 2014). They also highlighted carryover effects and found a negative relationship between wintertime Hg concentrations and eggs size during the following breeding season. Thus, it is important to determine the role of winter and winter distribution on seabird exposure to Hg to better understand the origin of this contamination in space and time.

The development of new technologies such as geo-tracking devices has allowed the access to crucial information about migratory movements and distribution of Arctic seabirds during the non-breeding period. In addition, information about the non-breeding Hg contamination can be assessed by the use of specific feathers. Indeed, Hg is excreted into the feathers during the molt, where it remains stable between two molts (see **Paper 1** and **Chapter 2** for details). Thus, by using geo-tracking devices and specific feathers, it is possible to link distribution and Hg concentrations during the non-breeding period.

6. Thesis objectives and outline

In this context, it is crucial to improve the existing knowledge about large-scale Hg exposure of Arctic seabirds by taking into account their winter migratory movements and distribution. Hence, based on a multi-species and multi-colony approach, the main goal of my PhD thesis is to answer the following question:

What is the role played by migratory movements of Arctic seabirds on their exposure to mercury at the pan-Arctic scale?

To answer this question, the doctoral work will be divided into five chapters. The main questions of these chapters will be briefly described here, and the hypotheses, predictions, main results and

subsequent conclusions and discussions will be presented along the corresponding chapters. A final chapter will focus on the general conclusion of this doctoral work, limitations and perspectives.

In **Chapter 2**, I will present the general methods used to achieve this doctoral work. **Chapter 3** will be dedicated to the Hg seasonality in Arctic seabirds and the importance of the winter period in seabird Hg contamination. To do so, based on multi-species and multi-colony analyses, I will first describe Hg concentrations during the breeding season through the Arctic (**Paper 2**), followed by a description of Hg contamination during the non-breeding period, and highlight the potential seasonality in Hg concentrations (**Paper 3**). In **Chapter 4**, I will focus on spatio-temporal variation of Hg contamination of Arctic seabirds during the non-breeding period, at the individual scale. Based on a multi-species approach, I will study temporal variations in winter Hg contamination of Arctic seabirds and the potential link with their wintering ground fidelity (**Paper 3**). **Chapter 5** will focus on the role played by the winter migration on Hg contamination of Arctic seabirds at both individual and population scales (**Papers 4, 5, and 6**). Finally, **Chapter 6** will be dedicated to a general discussion and conclusion of the doctoral work, where subsequent perspectives of this work will be presented.

Chapter 2

General methodology: biological sampling, laboratory and spatial analyses



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The general protocols and methods for sampling and laboratory analyses are described in the present chapter. The details related to the specific study cases will be provided in the related chapters and papers.

1. ARCTOX

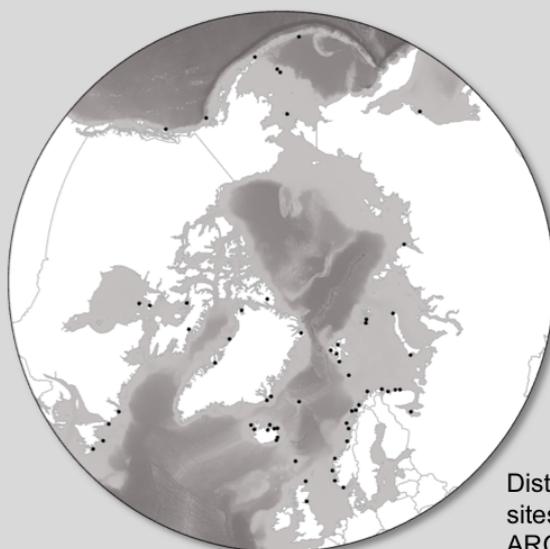
The present doctoral work is part of ARCTOX, a pan-Arctic network aiming at tracking mercury (Hg) contamination across the Arctic marine food webs. The biological samples were collected following a standardized protocol and sent to the Littoral Environnement et Sociétés (LIENSs -La Rochelle University, La Rochelle, France) where they were analyzed with the supports of the laboratory technicians. All financial supports' details are provided in the preface (page i).

1.1. Sampling protocols

This PhD thesis is based on the study of 23 Arctic seabirds' species from five families, with samples collected from 2015 to 2018 on 51 Arctic study sites. The families comprise the Alcids (or auks), the Larids (or gulls), the Procellariids, the Anatids (or ducks), and the Phalacrocoracids (or shags). Details regarding their ecologies are provided in the *Annex 1*. The sampling area covers Norway, Iceland, the Faroe Islands, Svalbard, Bjørnøya, Jan Mayen, Greenland, Canada, Alaska, Aleutian Islands and Russia.

ARCTOX network

The ARCTOX network has been initiated in 2015 and is coordinated by Jérôme Fort, currently a CNRS researcher based at the Littoral Environnement et Sociétés LIENSs (La Rochelle University, La Rochelle, France). It is currently composed of six staff members and collaborators from Canada, Denmark, Estonia, Faroe Islands, France, Iceland, Norway, Poland, Russia, Scotland and U.S.A. More information at www.arctox.cnrs.fr/en/collaborators.



Distribution of the sampling sites (black dot) covered by ARCTOX to date

Alcid

Ancient murrelet (*Synthliboramphus antiquus*)
Atlantic puffin (*Fratercula arctica*)
Brünnich's guillemot (*Uria lomvia*)
Common guillemot (*Uria aalge*)
Crested auklet (*Aethia cristatella*)
Horned puffin (*Fratercula corniculata*)
Least auklet (*Aethia pusilla*)
Little auk (*Alle alle*)
Parakeet auklet (*Aethia psittacula*)
Razorbill (*Alca torda*)
Rhinoceros auklet (*Cerorhinca monocerata*)
Tufted puffin (*Fratercula cirrhata*)



Larids

Black-legged kittiwake (*Rissa tridactyla*)
Common gull (*Larus canus*)
Glaucous gull (*Larus hyperboreus*)
Glaucous-winged gull (*Larus glaucescens*)
Herring gull (*Larus argentatus*)
Lesser black-bakked gull (*Larus fuscus*)
Red-legged kittiwake (*Rissa brevirostris*)



Procellarid

Northern fulmar (*Fulmarus glacialis*)



Anatid

Common eider (*Somateria mollissima*)



Phalacrocoracid

European shag (*Phalacrocorax aristotellus*)
Pelagic Cormorant (*Phalacrocorax pelagicus*)



1.1.1. Blood sampling

1.1.1.1. Blood as a tool to monitor Hg contamination during the breeding season.

Blood is a tissue commonly sampled for Hg studies, which informs about relatively local and short-term Hg contamination (Bearhop et al. 2000, Monteiro & Furness 2001). Depending on the species, blood samplings were realized either during the incubation (e.g. common eiders) or the chick rearing (e.g. auks). Indeed, some species like the common eiders can only be monitored during the incubation as they leave the nest with their ducklings once they have hatched. On the contrary, most of the auks and gulls can be monitored during the chick rearing as chicks are fed by adults until they fledge.

1.1.1.2. Blood sampling

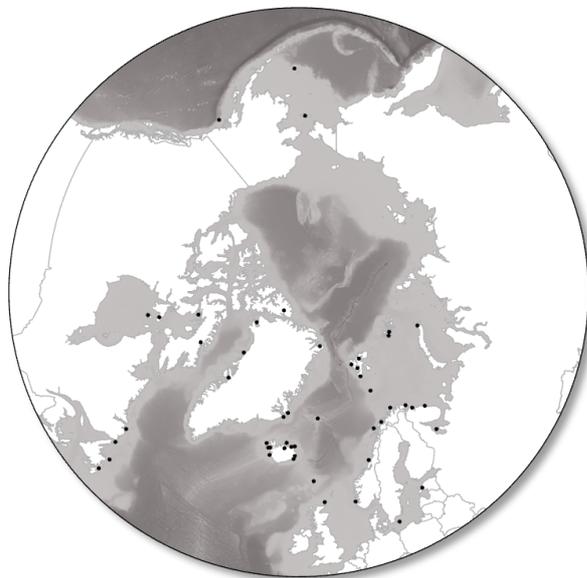


Figure 2.1: Colonies (black dot) sampled for blood included in the present PhD thesis.

From 2015 to 2018, 21 species distributed in 50 breeding colonies were sampled during the breeding season (Fig. 2.1 – more details in **Chapter 3**). Blood samples of 0.5-1 ml were collected on adults from the brachial vein or the

jugular (common eiders - Fig. 2.2), added to a vial with ~1 ml of ethanol 70% and stored until analyzed in the laboratory.



Figure 2.2: Blood sampling on the brachial vein of a little auk (left picture - © Samuel Perret) and b) on the jugular of the common eiders (right picture - Kongsfjorden, 2008) (with the courtesy of Børge Moe and Sveinn-Are Hanssen)

1.1.2. Feather sampling

1.1.2.1. Hg excretion into feathers

Seabirds excrete Hg through the excrements, the eggs, in the case of the females, and the feathers. This last path has been proposed to be the main path of excretion as ~70-90% of the Hg available in the bloodstream and internal tissues is excreted into the plumage during molt (Honda et al. 1986, Braune 1987, Agusa et al. 2005). Hg bounds to disulphide bridges of feather keratins (Crewther et al. 1965) and becomes stable once the feather is fully grown (Appelquist et al. 1984). Hence, Hg concentrations in feathers represent the Hg accumulated between two molts and inform about the Hg body burden at the molting time (Furness et al. 1986, Agusa et al. 2005). As a consequence, Hg concentrations in feathers are not affected by the age of the individual (Honda et al. 1986, Bustamante et al. 2016). The Hg excreted into feathers is mainly under its organic and toxic MeHg form (>80%) (Thompson & Furness 1989, Bond & Diamond 2009, Renedo et al. 2017). This means that measuring total Hg concentrations in seabird feather samples provides good information about their contamination and exposure to MeHg.

Some Arctic seabirds molt once a year only, i.e. post or pre-breeding molt (female anatids or ivory gulls, respectively) while others (most of the alcids, larids, and male anatids) molt twice a year (**Paper 1**). For instance, at the end of the breeding season, alcids complete a total molt (body, head and flight feathers) leading to their wintering plumage. At the end of the winter, they only molt their cheek, head and neck feathers, leading to their nuptial plumage. Hence, head feathers of most alcid and larid

species (Annex 1 of the **Paper 1**) provide information about Hg contamination during the non-breeding period (Fort et al., 2014, [Fig. 2.3](#)) and body feathers about the breeding period. For species having only one molt per year, the body feathers provide information about Hg contamination throughout the year ([Fig. 2.3](#)).

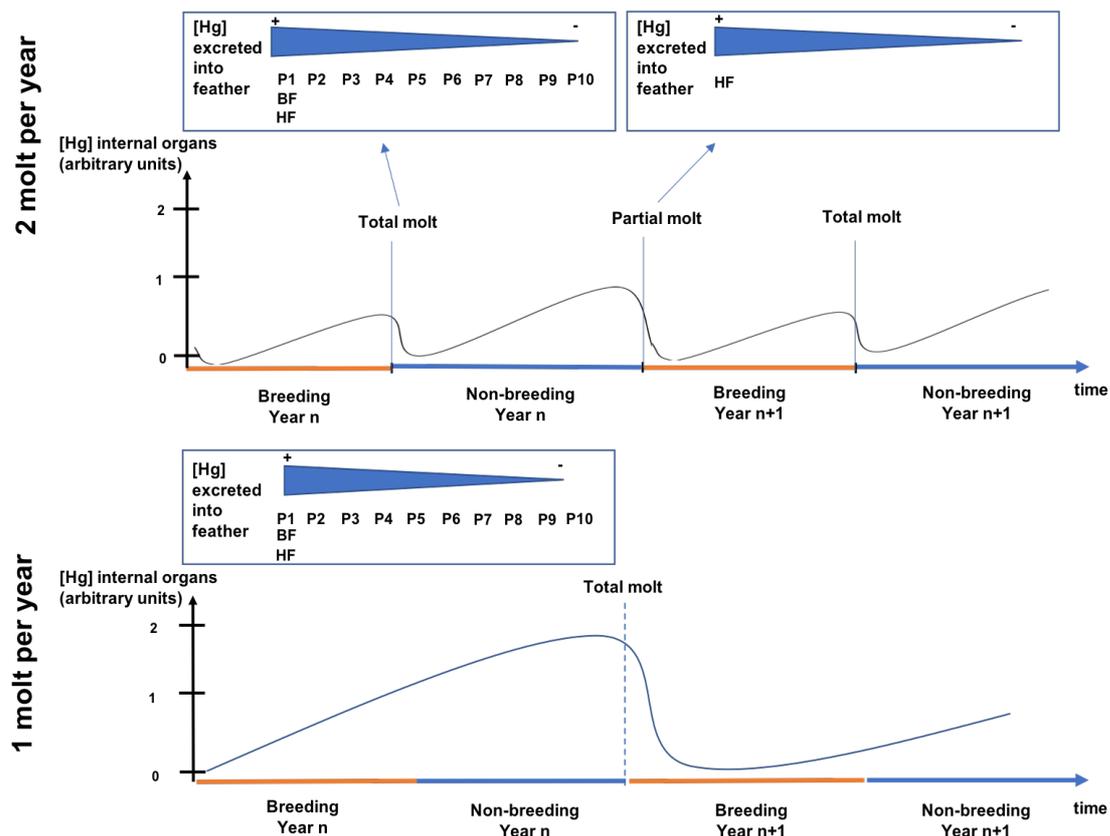


Figure 2.3: Extracted from **Paper 1**. Schematic representation of Hg excretion from internal tissues and organs to body, head and primary feathers for seabirds with one and two molts per year.

1.1.2.2. Types of feathers sampled

From 2015 to 2018, 23 species breeding in 51 colonies were sampled during the breeding season ([Fig. 2.1](#) – more details in **Chapter 3**), placed in plastic bags or paper envelopes and stored until analyzed in the laboratory. For Alcids, both body (belly and back) and head (cheek, head, neck – [Fig. 1.4](#)) feathers were sampled. For the Anatids, Larids, Procellariids and Stercorariids, only body (belly and/or back) feathers were sampled.

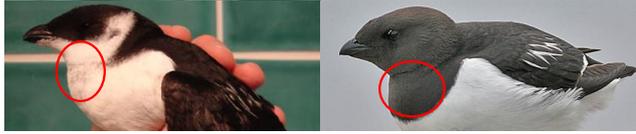


Figure 1.4: Red circles represent the area where head feathers should preferentially be sampled. extracted from the ARCTOX protocol, example here on the little auk on wintering plumage (left) and nuptial plumage (right).

1.2. Mercury (Hg) laboratory analysis

1.2.1. Blood and feathers samples preparation

Prior to analysis, both blood and feathers samples were dried and homogenized. For the blood samples, it consisted in being dried for 72 hours at ambient temperature to remove the ethanol, lyophilized for 48 hours and then homogenized. For the feathers, we first cleaned them to remove external contamination. To do so, we first removed the down and the bulk to only analyze the main part of the feathers. Then, the feathers were placed into a 2:1 chloroform:methanol solution in an ultrasonic bath for three minutes. The next step consisted in rinsing the feathers in two consecutive methanol baths. Then, the feathers were placed in an aluminium sheet and dried at 45°C in an oven for 48 hours (Fig. 1.5).

1.2.2. Hg analysis

Total mercury concentrations were measured in blood and feathers at the Littoral Environnement et Sociétés laboratory (LIENSs, La Rochelle, France). Prior to each set of analyses, we first ran blanks. To ensure the accuracy of the measurements, certified referenced materials were used (dogfish liver DOLT-5, NRC, Canada; reference value = 0.44 ± 0.18 ; mean measured value = 0.334 ± 0.04 – lobster hepatopancreas TORT-2, NRC, Canada; reference value = 0.27 ± 0.06 ; mean = 0.26 ± 0.01 $\mu\text{g/g}$ of dw) every 15 samples. Total Hg analyses were performed on a subsample of ~ 0.2 mg of blood and ~ 0.1 to 1.0 mg of feathers using an Advanced Mercury Analyser spectrophotometer (Altec AMA 254 – detection limit of 0.05 ng of dry weight - dw) (Fig. 1.5).

Samples preparation

FEATHERS

1 Cleaning

- Feathers are plunged in a 2:1 chloroform:methanol solution in a 3-minute ultrasonic bath.
- Feathers are rinsed twice in a methanol solution

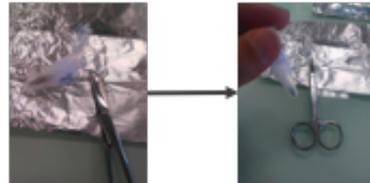


2 Drying

- 48 hours at 45° C

3 Homogeneisation

- To reduce homogeneity, three feathers are reduced into powder



4 Hg analysis

Advanced Mercury Analyser spectrophotometer (Altec AMA 254)



Detection limit of 0.05 ng of dry weight – dw

4 Hg analyses

3 Homogeneisation

2 Lyophilisation

1 Ethanol evaporation

BLOOD

Figure 1.5: protocol of sampling (blood and feathers) preparation

2. SEATRACK

SEATRACK is an international network aiming at tracking the winter distribution of Arctic seabirds from the North, Barents and Norwegian seas with the use of geo-tracking devices. Large populations of Arctic seabirds breed in the North Atlantic and their ecology at this time of the year is well known. However, information about their non-breeding period is scarce. As the winter period is harsh for seabirds, it is crucial to know where they winter to understand their ecology and the potential threats they might face. The network is composed of international researchers from Russia, Norway, Great Britain, Canada, France, the Faroe Islands and Iceland, leading Arctic seabird's assessment programs. Following a standardized protocol, they proceed to the deployment and the recovery of the geo-tracking devices on the field. All the geo-tracking devices are provided and analyzed following a standardized method (Bråthen et al. 2020) by the staff members of SEATRACK. To date, the program includes the study of 11 species breeding on 40 sampling sites.

Winter distribution of Arctic seabirds was monitored with light logger technology, also called GLS (Light Level Geolocator) loggers, weighting less than 1% of seabird weight. Breeding individuals were caught and equipped during the breeding season. To do so, GLS were attached to the seabirds' leg with the use of color-coded rings, cable-ties and glue (Fig. 1.6). Once started, the GLS registered the light level every five or ten minutes. The light level informed about the time of the sunrise and sunset, as well as the duration of the day and the night, which allowed the calculation of longitudes and latitudes of the birds, respectively. Two points per day were extracted, with an accuracy of ~180 km. These devices have two main limitations. First, birds needed to be recaptured the following year to retrieve the GLS and the recorded data. Second, as these geo-tracking devices depend on light to calculate positions, we could calculate their position when days and nights were the same length (i.e. equinox) and during the midnight sun or Polar night.

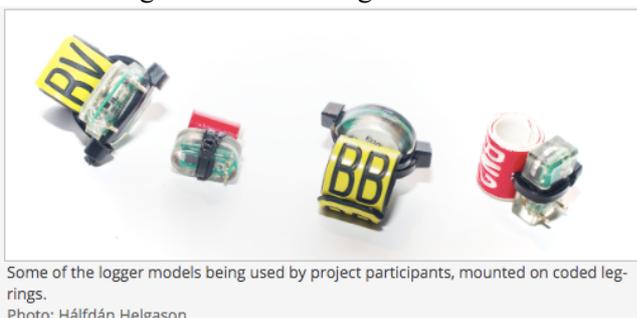
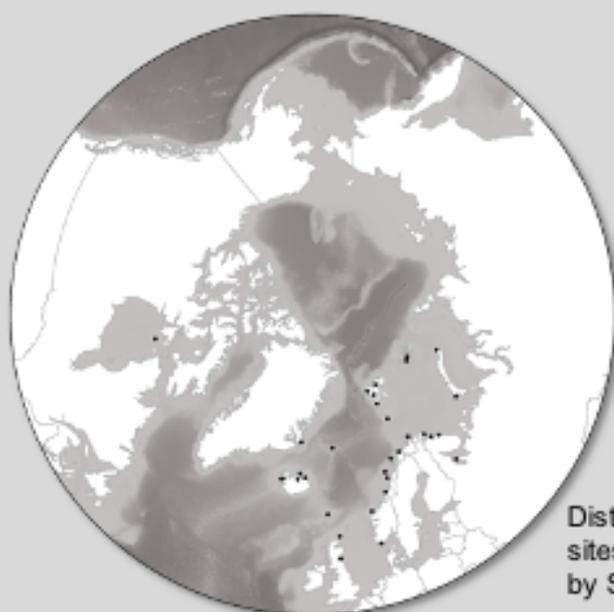


Figure 1.6: extracted from the SEATRACK website www.seapop.no/en/seatrack/about/

SEATRACK Network

SEATRACK is an international network started in 2014 and animated by a project staff currently including nine researchers and technicians from the Norwegian Polar Institute (NPI), the Norwegian Institute for Nature Research (NINA) and the Norwegian Environment Agency (NEA). It is composed of collaborators from Norway, Russia, Great Britain, the Faroe Islands and Iceland. The leaders of the study sites deploy and retrieve the geotracking devices on the field that are provided by the SEATRACK project group.

More information at www.seapop.no/en/seatrack/



Distribution of the sampling sites (black dots) covered by SEATRACK to date

Alcids

Atlantic puffin (*Fratercula arctica*)
Brünnich's guillemot (*Uria lomvia*)
Common guillemot (*Uria aalge*)
Little auk (*Alle alle*)



Larids

Black-legged kittiwake (*Rissa tridactyla*)
Glaucous gull (*Larus hyperboreus*)
Herring gull (*Larus argentatus*)
Lesser black-backed gull (*Larus fuscus*)



Phalacrocoracids

European shag (*Phalacrocorax aristotellus*)



Anatids

Common eider (*Somateria mollissima*)



Procellarids

Northern fulmar (*Fulmarus glacialis*)



3. ARCTOX and SEATRACK collaborations

ARCTOX and SEATRACK networks have been collaborating since 2015 with the aim of mapping Hg contamination during the non-breeding period in the North-Atlantic Arctic. To do so, during field seasons (breeding periods), each bird equipped with a SEATRACK GLS logger is sampled for both blood and feathers. The present PhD thesis is based on the recovery of 1,586 birds of nine species from five families equipped with GLS and analyzed for Hg concentrations in blood and/or feathers (Fig. 1.7).

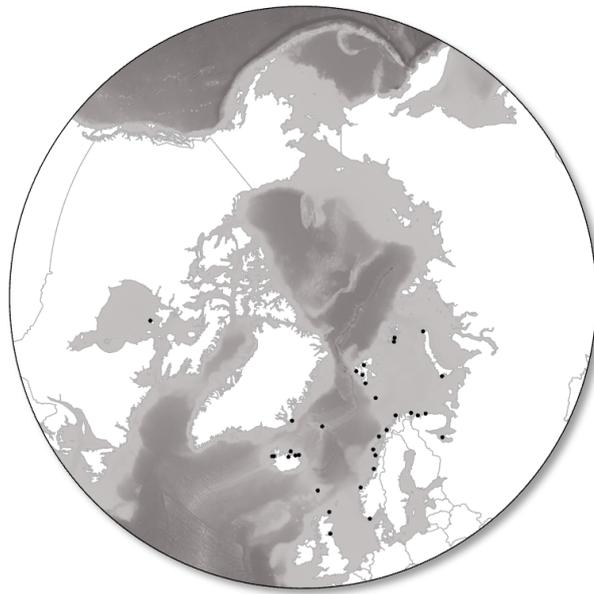


Figure 1.7: Study area of the ARCTOX and SEATRACK collaboration

Chapter 3

Are Hg contaminations seasonally constant?



Before studying the role of migration on Hg contamination of Arctic seabirds, it appears important to study Hg contamination during both breeding and non-breeding periods. For this reason, we first reviewed the existing knowledge about Hg concentrations measured in both blood and feathers of Arctic seabirds throughout the year (**Paper 1**). We found that most of the current information regarding Hg concentrations in the Arctic focused on the breeding period. During the non-breeding period, seabirds mostly migrate to southerly areas and in open seas where it is therefore more complicated (and expensive) for researchers to reach and follow the birds. But with the development of tracking devices and the use of selected biological samples, it is now possible to access seasonal information about Hg contamination in Arctic seabirds (see **Chapter 2**).

In the present chapter, I will summarize our research on large-scale Hg contamination during the breeding period, the non-breeding period, in addition to the potential Hg seasonality. I will also introduce the role of seabirds as bio-indicators for large-scale Hg studies, this being possible with an adjustment of blood Hg concentration by seabirds' trophic position. This research is based on a multi-species and multi-colony approach. First, I will describe large-scale Hg contamination during the breeding period with the use of both blood and body feathers, and their importance for large-scale Hg studies (**Paper 2**). I will also discuss the results in comparison to the findings of the review of the literature (**Paper 1**). Then, I will use seabirds as bio-indicators for large-scale Hg studies. Thus, I will show how Hg concentrations in blood can be adjusted by bird trophic position with the use of nitrogen stable isotopes ($\delta^{15}\text{N}$). Preliminary results and discussions will be introduced to show the potential of such a method (**Paper 2**). Then, I will describe large-scale Hg contamination during the non-breeding period and the potential Hg seasonality in Arctic seabirds, with the study case of the Alcid family (**Paper 3**).

1. Hg contamination during the breeding period

Large-scale seabird Hg contamination in the Arctic has been investigated and reviewed in previous studies (AMAP, 2018, Provencher et al., 2014, **Paper 1**), suggesting spatial variations with higher Hg concentrations measured in the West Atlantic compared to the East Atlantic. These studies also suggested higher Hg concentrations in species' feeding at a high trophic level. However, some

species and areas are still understudied. Based on a review of the literature, we showed that the multiple studies about Arctic seabirds did not necessarily use the same tissues and/or methods to measure Hg concentrations (**Paper 1**). The comparison of Hg concentrations between different tissues and even analyzed with different methods is possible by transforming the values with published equations (for example blood to feather, see Ackermann et al., 2016, or wet weight to dry weight, see Eagles Smith et al., 2016). However, they probably added uncertainties due to interspecific and/or spatial variations in Hg concentrations. Within the ARCTOX network, blood and feathers are largely sampled through the pan-Arctic area, following a standard protocol of sampling and laboratory analyses. Hence, and based on the ARCTOX network sampling, in this section we will focus on large-scale Hg contamination with the use of blood and body feathers sampled from 2015 to 2018 on 20 species distributed on 50 breeding sites. Those results will be compared with the existing knowledge regarding the use of blood and feathers, reviewed in **Paper 1**. We will also compare and discuss those results with the known threshold of risks proposed in the literature for both blood and feathers. **We hypothesized the existence of interspecific and spatial variations in Hg concentrations and predicted 1/ higher Hg concentrations in piscivorous and omnivorous species, 2/ higher Hg concentrations in the West Atlantic.**

1.1. Interspecific variations

1.1.1. Blood

Blood informs about relatively local and short-term Hg contamination (Bearhop et al. 2000, Monteiro & Furness 2001). In the present PhD thesis, blood was collected during incubation or chick rearing, and therefore informed about Hg contamination during this period and the few weeks before (see **Chapter 2** and **Paper 1** for more details). We tested for differences of blood Hg concentrations (n = 3,603) between species and regions with a linear mixed model. The pan-Arctic area, based on a cluster analysis, was divided into eight regions for the blood, being the “Baltic sea”, the “Barents Sea”, the “Greenland Sea”, “North-Norway and Svalbard”, “Pacific region”, “South Norwegian Sea”, “Northwest-Atlantic Arctic” and “Northwest-Atlantic sub-Arctic” (see detailed map in *Annex 2*). The breeding colonies are presented in *Annex 2*. This model included species (n = 20) and regions (n = 8)

as fixed effects. Because we combined data collected during multiple years and breeding colonies, but also because some individuals have been monitored for several years, the sampling years ($n = 4$),

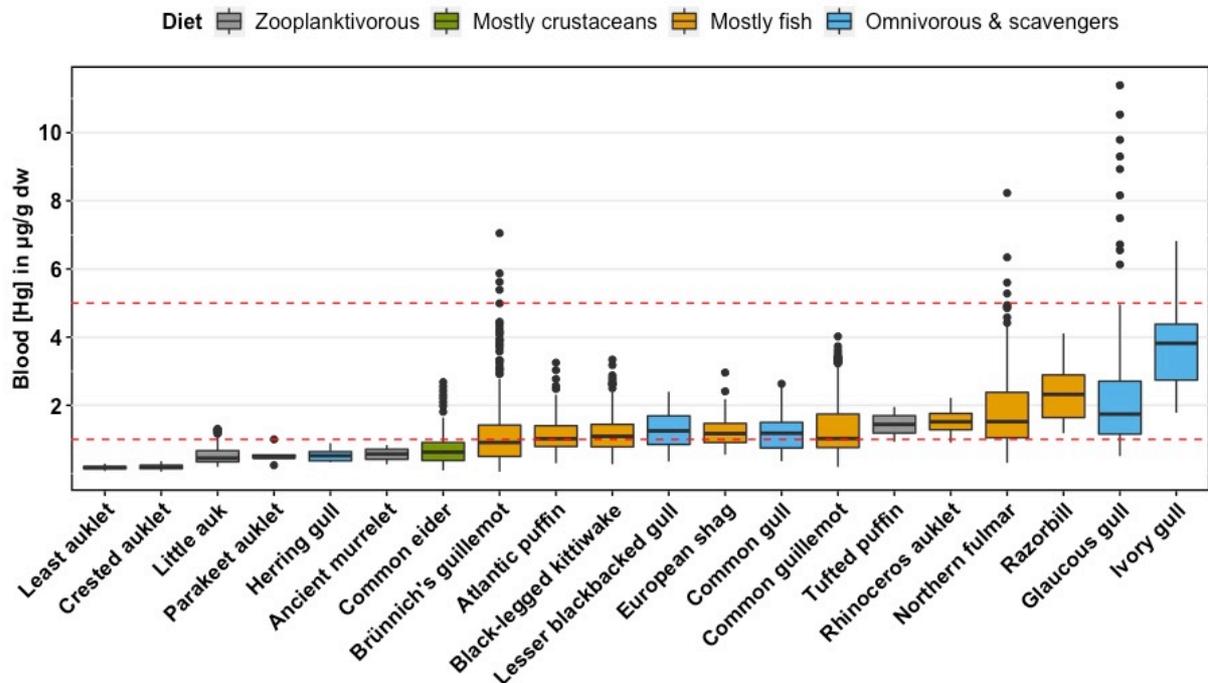


Figure 3.1: [Hg] in blood in $\mu\text{g/g dw}$, ranged by mean Hg concentrations per species. Red dotted lines represent low and moderate risks of toxicity at 1.00 and 5.00 $\mu\text{g/g dw}$, respectively (see Ackermann et al., 2016). The boxplots are colored by species diet during the breeding period.

individuals ($n = 3,040$) and breeding colonies ($n = 50$) were added as random factors. The mixed model indicated an effect of species ($F_{19,109.19} = 42.48$, $p < 0.001$), regions ($F_{7,28.41} = 4.22$, $p < 0.01$) and the interaction between these two variables ($F_{28, 139.68} = 45.62$, $p < 0.001$).

Following our predictions, piscivorous and omnivorous species presented some of the highest Hg concentrations (i.e. ivory gull *Pagophila eburnea* – mean \pm SD = $3.81 \pm 1.71 \mu\text{g/g dw}$ - Fig. 3.1). On the other hand, zooplanktivorous species were among the species with the lowest Hg concentrations (i.e. least auklet *Aethia pusilla* – $0.17 \pm 0.06 \mu\text{g/g dw}$ - Fig. 3.1). The results regarding the ivory gulls are consistent with previous studies highlighting their high Hg contamination, probably due to their diet (scavengers, opportunistic) (Bond et al. 2015). However, results regarding the ivory gull do not follow what we found in **Paper 1**, where their Hg concentrations were similar to values measured in species mostly relying on crustaceans (Fig. 3.2). This is probably explained by spatial differences. Indeed, in

the present section, individuals were sampled in High Arctic in E. Greenland (Station North - Fig. 3.1), while the individuals from **Paper 1** were sampled in E. Svalbard (Barentsøya - Fig. 3.2 - Lucia et al., 2016). This follows the spatial variations found on ivory gulls' eggs where higher Hg concentrations were measured in Station North (E. Greenland – median = 0.24 µg/g ww) than in Svenskøya (E. Svalbard - median 0.14 µg/g ww - Lucia et al. 2015).

We also found interspecific variations in Hg concentrations that might be explained by spatial differences (Fig. 3.1). For instance, tufted puffins *Fratercula cirrhata* (only monitored in the E. Pacific - Talan Island, Sea of Okhotsk – 1.44 ± 0.72 µg/g dw), mostly feeding on zooplankton, and rhinoceros auklets *Cerorhinca monocerata* (only monitored in the W. Pacific - Middleton, N. Pacific Ocean – 1.51 ± 0.38 µg/g dw), mostly feeding on fish, have close Hg concentrations (*Annex 1* - Fig. 3.1). Such a result might suggest spatial variations in Hg concentrations with a higher exposition to Hg in the East than in the West Pacific. Overall, our results suggest that the interspecific variations in Hg concentration in blood could be explained by the diet, but also by spatial variations.

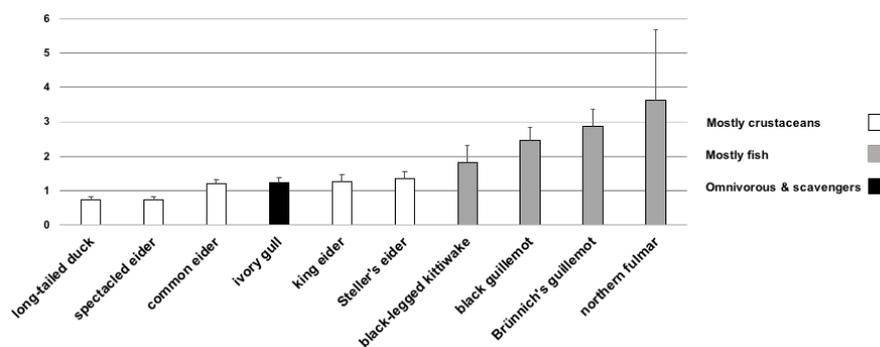


Figure 3.2: Extracted from **Paper 1**. Mean [Hg] (mean ± SD µg/g dw) measured in blood by species, in relation to bird diet during the breeding season

1.1.2. Body feathers

Body feathers inform about Hg contamination during an intermolt period, and more specifically during the breeding period for species molting twice a year (but see **Chapter 2, Paper 1** and *Annex 2* for more details). We tested for differences of Hg concentrations in body feathers (n = 3776) between species and regions with the use of a linear mixed model. The pan-Arctic area, based on a cluster analysis, was divided into seven regions for the feathers, being the “Barents Sea”, the “Greenland Sea”, “North-Norway and Svalbard”, “Pacific region”, “South Norwegian Sea”, “Northwest-Atlantic Arctic”

and “Northwest-Atlantic sub-Arctic” (see detailed map in *Annex 2*). The breeding colonies are presented in *Annex 2*. This model included the species (n = 20) and regions (n = 7) as fixed effects.

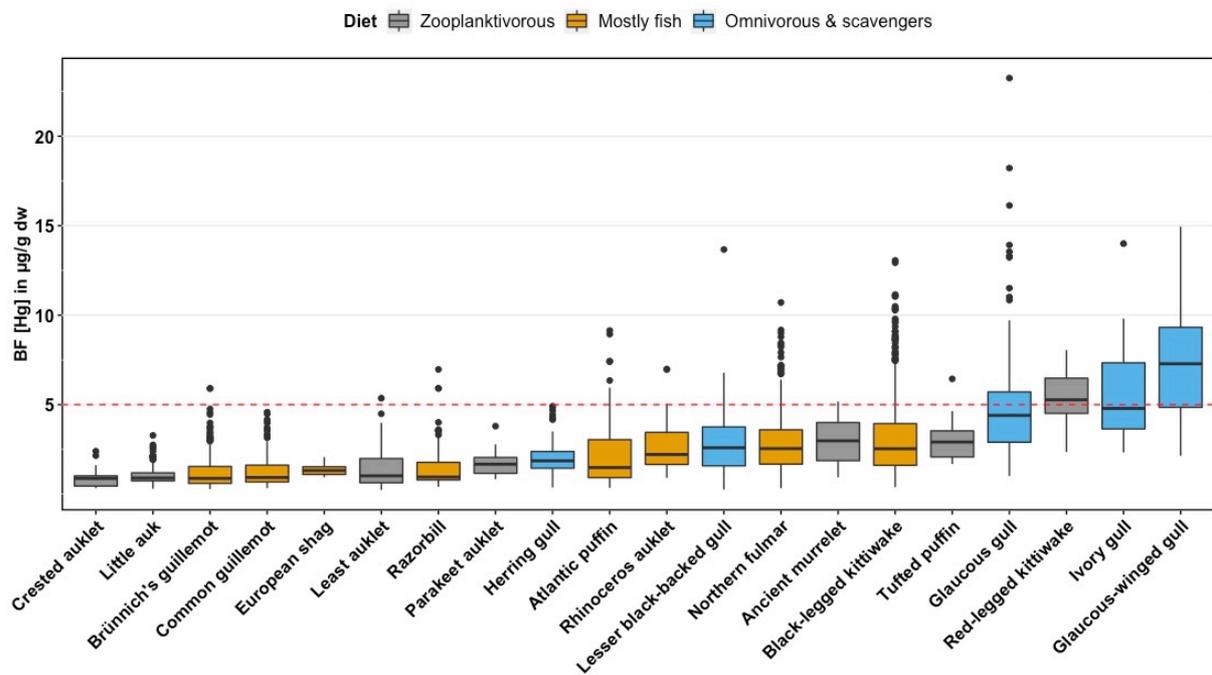


Figure 3.3: [Hg] in body feathers in µg/g dw, ranged by mean Hg concentrations per species. Red dotted line represents the risks at 5 µg/g dw (Eisler, 1987). The boxplots are colored by species diet during the breeding period.

Because we combined data collected during multiple years and at multiple sampling sites, but also because some individuals have been monitored for several years, the sampling years (n = 4), individuals (n = 3122) and breeding colonies (n = 51) were added as random effects. The mixed models indicated an effect of the species ($F_{19, 184.81} = 100.97$, $p < 0.001$), the regions ($F_{6, 36.49} = 15.48$, $p < 0.001$) and the interaction of these two variables ($F_{29, 448.12} = 20.58$, $p < 0.001$).

According to our predictions, piscivorous or omnivorous (e.g. gulls) presented some of the highest Hg concentrations (glaucous-winged gull *Larus glaucescens*: mean ± SD = 7.61 ± 3.88 µg/g dw), while planktivorous and/or piscivorous (mostly the alcids) were among the species with the lowest Hg concentrations (crested auklet *Aethia cristatella*: 0.91 ± 0.47 µg/g dw). As previously shown with the blood, ivory gulls had some of the highest mean Hg concentrations (6.02 ± 3.60 µg/g dw - Fig. 3.3), just behind the glaucous-winged gull. The glaucous-winged gull is a species endemic to the Pacific for which we only collected samples on Buldir Islands (Aleutian Islands). In the present study, we found

higher Hg concentrations (Fig. 3.3) than in the review of the literature ($3.61 \pm 0.59 \mu\text{g/g dw}$ - Fig. 3.4 - [Burger et al., 2009](#)), although birds were sampled in the close Kiska and Adak Islands colonies (Aleutian Islands – see **Paper 1**). Hence, the difference might not come from spatial variations, but most likely from temporal variations. Indeed, while individuals from the review were collected in 2004, individuals from the present study were collected in 2017. Additionally, we saw that Hg concentrations in herring gulls were much lower in the present study (Fig. 3.3), compared to the results from the review paper (Fig. 3.4). Herring gulls from the present study were sampled in five different breeding colonies from the Greenland Sea to the Barents Sea, with relatively constant Hg concentrations (mean \pm SD = $2.00 \pm 1.00 \mu\text{g/g dw}$). However, individuals from the review of the literature were sampled in Chaun ($6.08 \pm 4.06 \mu\text{g/g dw}$ - Russia – Fig. 3.4 - [Kim et al., 1996a, 1996b](#)), in 1993. Therefore, this suggest or temporal or spatial differences between those two studies. Surprisingly, red-legged kittiwakes *Rissa brevirostris*, mostly relying on zooplanktons were among the species with the highest Hg concentrations. In our study, this species was only sampled on St George Island (Pribilof Island – South of the Bering Sea). Therefore, these results suggest either a high exposure to Hg concentrations in this area, or the use of a specific habitat (e.g. surface, glacier) presenting higher concentrations.

Hence, as for the blood, our results show interspecific variations that could probably be explained by the diet, but with an intraspecific variation probably explained by spatial variations in Hg concentrations through the pan-Arctic. Overall, blood and body feathers seem to show the same interspecific variations in Hg concentrations with the piscivorous and/or omnivorous species being among the most contaminated and the zooplanktivorous species being among the least contaminated. Some variations occur, probably explained by spatial variations, which will be the focus of the following section.

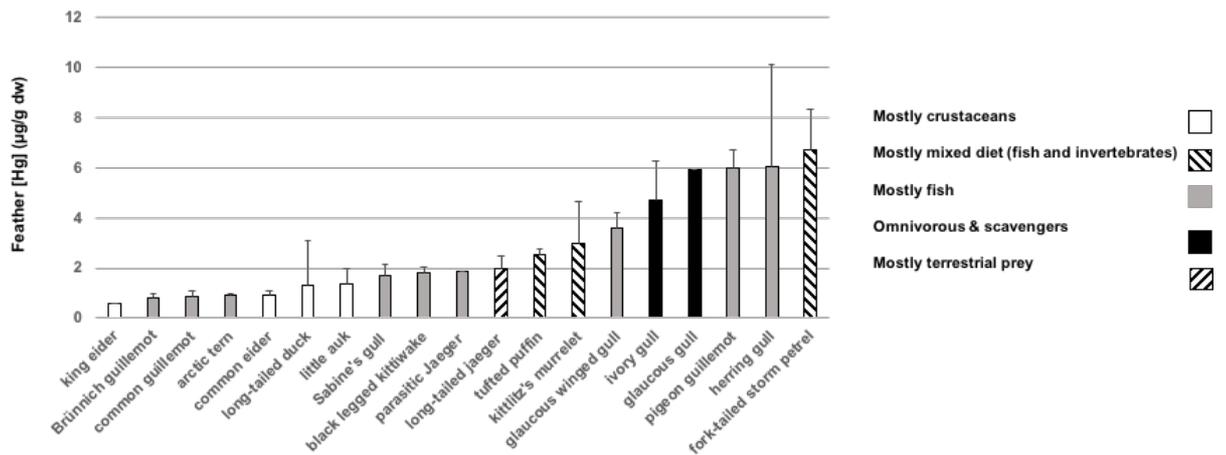


Figure 3.4: Extracted from **Paper 1**. Mean Hg concentration (mean \pm SD $\mu\text{g/g dw}$) measured in body feathers, by species, in relation to bird diet during the breeding season.

1.2. Spatial variations

Because of the high number of species presented in this section, we decided to focus the present sub-section on Brünnich's guillemots and black-legged kittiwakes only, two species with a circumpolar distribution and for which we have the greatest spatial coverage. All boxplots regarding the other species are presented in *Annex 2* and the mean Hg concentrations per regions, species and sampling sites are presented in *Annex 3*.

1.2.1. Brünnich's guillemots

1.2.1.1. Blood

Fig. 3.5 shows high spatial variabilities in Hg concentrations. For example, the region presenting the lowest mean Hg concentrations was the N. Norway – Svalbard (mean \pm SD = $0.36 \pm 0.23 \mu\text{g/g dw}$). The Pacific region also presented some of the lowest mean Hg concentrations ($0.65 \pm 0.56 \mu\text{g/g dw}$), followed by Barents ($0.84 \pm 0.18 \mu\text{g/g dw}$) and Greenland ($0.97 \pm 0.50 \mu\text{g/g dw}$) Seas. On the other hand, the highest Hg concentrations were measured in Northwest-Atlantic Arctic ($1.93 \pm 1.19 \mu\text{g/g dw}$) and sub-Arctic ($2.18 \pm 0.50 \mu\text{g/g dw}$). More specifically, we can see that the Baffin Bay population ($4.11 \pm 0.86 \mu\text{g/g dw}$ – Northwest-Atlantic Arctic), showed Hg concentrations ~ 20 times higher than the Bjørnøya one ($0.20 \pm 0.07 \mu\text{g/g dw}$ – North-Norway Svalbard). The east-west Hg concentrations increase found in the Atlantic region were in accordance with the results of previous studies (AMAP,

2018, Provencher et al., 2014). Similar patterns were found for other alcids studied in the present section (i.e. Atlantic puffins *Fratercula arctica* and common guillemots, *Annex 2*).

Brünnich's guillemots mostly feed on fish during the breeding season and forage at different distances of their colonies depending on their breeding origin (Gaston and Jones, 1998). Additionally, even though they mostly are piscivorous during the breeding season, invertebrates are also part of their diet (*Annex 2*). Hence, it is possible that the interpopulation variations in Hg concentrations observed between eastern (i.e. Barents Sea, N. Norway Svalbard or Greenland Sea) and western (i.e. West Atlantic or in the Pacific) colonies reflect a spatially different range of prey during the breeding season. This variation could also be explained by spatial differences in Hg contamination at their breeding ground.

1.2.1.2. Body feathers:

The spatial variabilities found for both blood and body feathers Hg concentrations presented the same trend with the lowest values on the East Atlantic and Pacific, and the highest on the West Atlantic (*Fig. 3.6*). Indeed, for the body feathers, we found the lowest Hg concentrations in the Barents Sea (mean \pm SD 0.71 ± 0.20 $\mu\text{g/g dw}$) or N. Norway – Svalbard (0.73 ± 0.37 $\mu\text{g/g dw}$). These concentrations were close to the ones measured in the Pacific (0.75 ± 0.29 $\mu\text{g/g dw}$). Intermediate Hg concentrations were measured in the Greenland Sea (1.54 ± 0.30 $\mu\text{g/g dw}$), while the highest concentrations were measured in Northwest-Atlantic Arctic (2.23 ± 0.78 $\mu\text{g/g dw}$) and sub-Arctic (2.39 ± 0.95 $\mu\text{g/g dw}$). As we observe relatively close spatial trend between both blood and feathers, this confirm that these tissues inform about the same period.

1.2.2. Black-legged kittiwakes

1.2.2.1. Blood

Fig. 3.7 shows that the spatial variations in Hg concentrations between the different regions were lower for the black-legged kittiwakes than for the Brünnich's guillemots (the colony with the lowest concentrations (0.61 ± 0.11 $\mu\text{g/g dw}$ - Kippaku) being ~ 4 times less contaminated than the one with the highest concentrations (2.37 ± 0.56 $\mu\text{g/g dw}$ – St George)). As for the Brünnich's guillemots (*Fig. 3.5*), and based on previous knowledge (AMAP, 2018), we expected some of the highest concentrations in the Northwest-Atlantic Arctic and sub-Arctic. Surprisingly, we found some of the

lowest Hg concentrations in the West-Atlantic Arctic (e.g. Kippaku, Ritenbenck – mean \pm SD for the region = $0.91 \pm 0.40 \mu\text{g/g dw}$), followed by the N. Norway – Svalbard ($1.02 \pm 0.49 \mu\text{g/g dw}$) and Barents Sea ($1.12 \pm 0.31 \mu\text{g/g dw}$). On the Atlantic region, the highest concentrations were measured for the Northwest Atlantic sub-Arctic ($1.21 \pm 0.39 \mu\text{g/g dw}$) and Greenland Sea ($1.23 \pm 0.47 \mu\text{g/g dw}$), but overall, the highest mean Hg concentrations were measured in the Pacific region (mean \pm SD = $1.57 \pm 0.51 \mu\text{g/g dw}$).

The intrapopulation variability is also much higher than in the case of the Brünnich's guillemots. Black-legged kittiwakes, as Brünnich's guillemots, mostly rely on fish (but also incorporate invertebrates' preys – *Annex 1*) during the breeding season and forage in the same range of km as Brünnich's guillemots (Daunt et al. 2002). The main difference between the two species is that Brünnich's guillemots are pursuit-divers while black-legged kittiwakes are surface feeders. Therefore, we suggest that the differences between the two species could be explained by differences in the diversity of preys they feed on, differences in Hg concentrations in the column water according to where the two species forage (e.g. < 10 m for black-legged kittiwakes, 100 -150m for Brünnich's guillemots), differences in the diversity of habitat they use or by physiological differences (e.g. excretion pathways, demethylation rate).

1.2.2.2. Body feathers

As for the Brünnich's guillemots, Hg concentrations in feathers and blood of black-legged kittiwakes follow the same spatial trend (Fig. 3.8). Therefore, we could also suggest that those two tissues represent the same period. However, we can see that the intrapopulation variation in Hg concentrations of the black-legged kittiwakes is much higher in feathers than in the blood Hg concentrations. The Hg concentrations measured into the feather represent contamination over an inter-molt period, while the blood represents Hg contamination within the previous few weeks (see **Paper 1**). This might therefore suggest a relatively constant diet and Hg exposure during the breeding period when blood is sampled, but a much more variable diet, habitat used and therefore Hg exposure during the inter-molt. As with the blood, we found some of the lowest Hg concentrations in W. Greenland, which was not consistent with the results found for the Brünnich's guillemots. Therefore, as proposed for the blood, we suggest that the differences between the two species might be explained by differences in the

diversity of preys they feed on, habitat differences (e.g. black-legged kittiwakes are surface feeders, while Brünnich's guillemots are pursuit divers), or physiological differences (e.g. excretion pathways, demethylation rate).

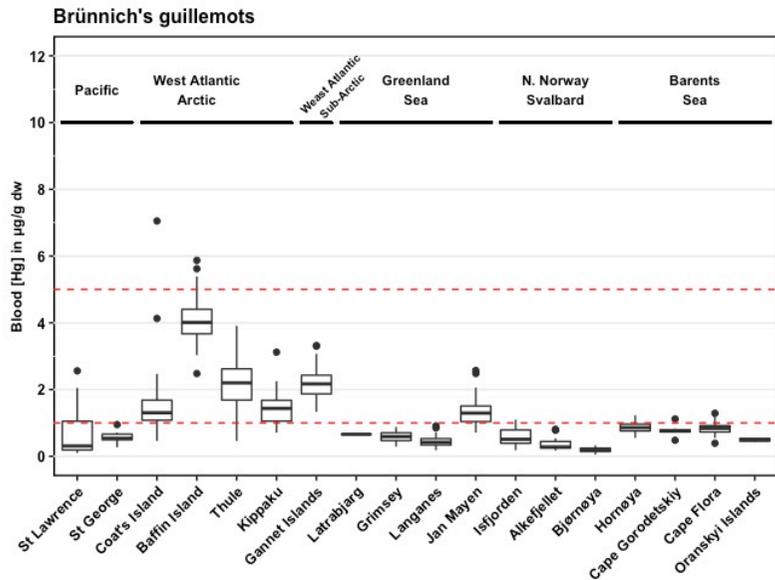


Figure 3.5: [Hg] in blood ($\mu\text{g/g dw}$) of the Brünnich's guillemots, by regions and breeding colonies. Red dotted lines represent the risks at 1 and 5 $\mu\text{g/g dw}$ (Ackermann et al., 2016).

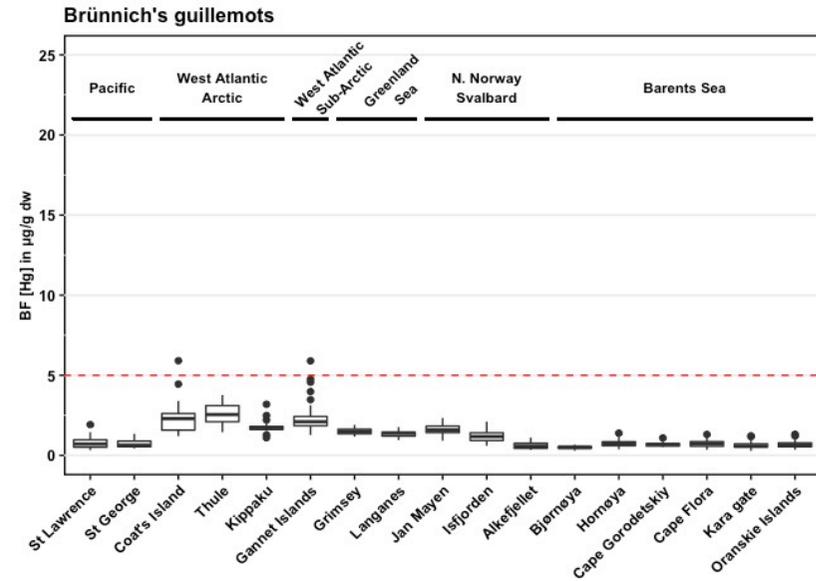


Figure 3.6: [Hg] in body feathers ($\mu\text{g/g dw}$) of the Brünnich's guillemots, by regions and breeding colonies. Red dotted lines represent the risks at 5 $\mu\text{g/g dw}$ (Eisler, 1987).

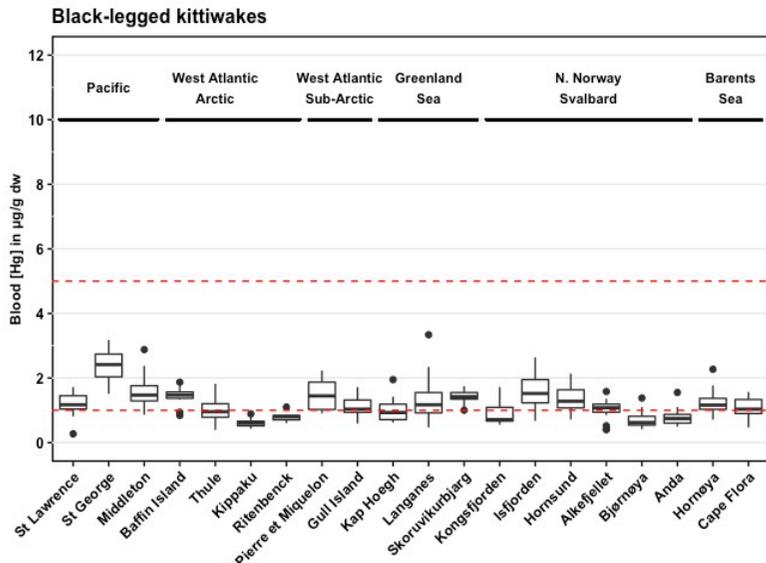


Figure 3.7: [Hg] in blood ($\mu\text{g/g dw}$) in black-legged kittiwakes, by regions and breeding colonies. Red dotted lines represent the risks at 1 and 5 $\mu\text{g/g dw}$ (Ackermann et al., 2016).

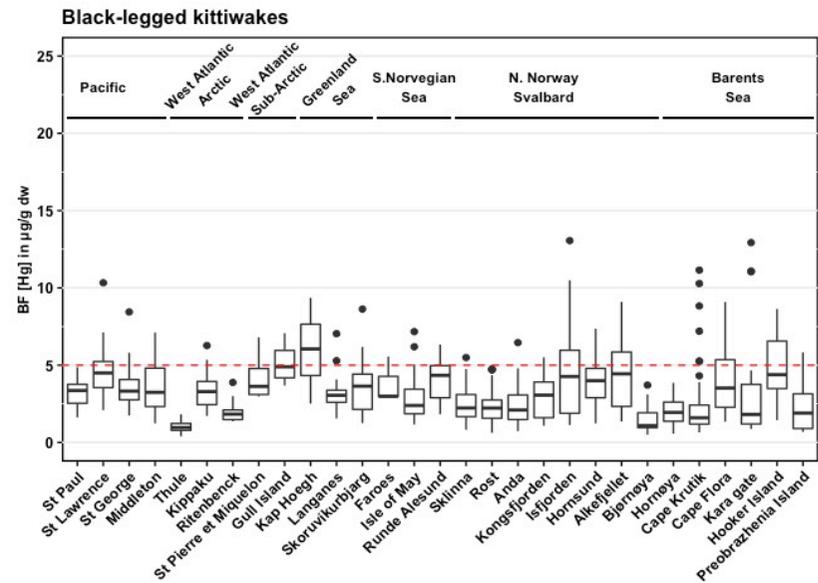


Figure 3.8: [Hg] in body feathers ($\mu\text{g/g dw}$) in black-legged kittiwakes, by regions and breeding colonies. Red dotted lines represent the risks at 5 $\mu\text{g/g dw}$ (Eisler, 1987).

1.3. Toxicity and risks

To study Hg toxicity from the values measured in blood of Arctic seabirds, we used the thresholds proposed by Ackerman et al (2016). Indeed, based on a review of the literature about avian Hg contamination across the western North Atlantic, they proposed different levels of risk:

- No risk: below 0.2 $\mu\text{g/g}$ ww (or 1.0 $\mu\text{g/g}$ dw).
- Low risk: above 0.2 $\mu\text{g/g}$ ww (or 1.0 $\mu\text{g/g}$ dw), with effects on health and physiology
- Moderate risk: above 1.0 $\mu\text{g/g}$ ww (or 5.0 $\mu\text{g/g}$ dw), with substantial effects on health, physiology, reproduction and behavior.
- High risk: above 3.0 $\mu\text{g/g}$ ww (or 14.0 $\mu\text{g/g}$ dw), severe impairment to health and reproduction
- Severe risk: above 4.0 $\mu\text{g/g}$ ww (or 19 $\mu\text{g/g}$ dw), reproduction failure and death.

In the present work, in order to have comparable values, we used dry weight (dw) values. To study Hg toxicity from the values measured in body feathers of Arctic seabirds, we used the thresholds proposed by Eisler (1987) who found the first deleterious effects at 5 $\mu\text{g/g}$ dw.

1.3.1. Interspecific variations

The study of Hg concentrations in blood revealed that most of the species (13 out of 20 species) and individuals (47% of the studied individuals) studied in the present work were, at least, at low risk (Fig. 3.1). The only species for which all individuals were not at risk were the least and crested auklets, herring gulls *Larus argentatus* and ancient murrelets *Synthliboramphus antiquus*. All the other species had at least few individuals above this first toxicity threshold. However, only Brünnich's guillemots (0.67% of the studied Brünnich's guillemots), northern fulmars (1.13%), glaucous gulls (6.10%) and ivory gulls (14.29%) had few individuals with Hg concentrations falling in the "moderate risk" category.

The study of Hg concentrations in the feathers revealed that most of the glaucous-winged gulls (67% of the studied glaucous-winged gulls), ivory gulls (50%), red-legged kittiwakes (62%) and glaucous-gulls (39%) presented Hg concentrations exceeding the thresholds of 5 $\mu\text{g/g}$ dw, being consistent with the raw blood values. Few other species presented individuals with Hg concentrations above this threshold being the black-legged kittiwakes or the northern fulmars. But when comparing with the blood, less species seemed at risk. Blood and body feathers should reflect the same period.

Thus, to explain this difference, we might consider the possibility that the threshold proposed by Eisler should be updated, and maybe specifically for Arctic seabirds. Indeed, more research regarding effects of Hg on Arctic seabirds are available and should therefore be reviewed, updated and refined (see Dietz et al., 2018). It might also be possible to propose blood-equivalent Hg concentrations for body feathers based on the relationship between concentrations measured in these two tissues, and thereby update this feather threshold. A dataset as the one used in the present doctoral work might actually be really helpful for such an analysis.

Overall, this interspecific study showed intraspecies variation in term of risk, probably explained by spatial variations, which will be presented for the Brünnich's guillemots and black-legged kittiwakes.

1.3.2. Brünnich's guillemots

For Brünnich's guillemots, we saw spatial variations in Hg concentrations in blood between regions. Only few individuals from Greenland Sea (only Jan Mayen, n = 68 individuals), N. Norway-Svalbard (only Isfjorden, n = 2), Barents Sea (n = 21) or Pacific (n = 13) exceeded the "Low risk" threshold. However, most individuals from the West-Atlantic Arctic (n = 123) and sub-Arctic exceeded (n = 41) this threshold, with even individuals above the "Moderate risk" threshold (West-Atlantic Arctic – n = 4). Therefore, with the use of the blood, our results suggest that some populations might be more at risk than others and should therefore be carefully monitored regarding Hg exposure and associated risks. However, with the use of the feathers, our results indicated that only two individuals of Brünnich's guillemots (one in Coat's Island, one in Gannet Islands) were above the threshold. Even though it is only two individuals, it is important to note that they both came from the most contaminated areas of the West Atlantic (Arctic and sub-Arctic).

1.3.3. Black-legged kittiwakes

For the black-legged kittiwakes, we also saw spatial variations, with most of the colonies exceeding the "Low risk" threshold when studying blood Hg concentrations. For instance, in the Pacific, all populations are above the "Low risk" threshold. Except for Kippaku and Ritenbenck colonies (West-Atlantic Arctic), Bjørnøya and Anda (N. Norway-Svalbard), all the other colonies have most of their individuals above this threshold. No individual was found above the "Moderate risk" threshold. When

looking at the body feather values, we saw a lot of heterogeneity with individuals from many colonies and most regions above the 5 µg/g dw threshold. For example, most individuals from Kap Hoegh (Greenland Sea) had concentrations above 5 µg/g dw, following the results for the blood Hg concentrations. In the Barents Sea, excepted for Hornøya, all the other sites comprised individuals with Hg concentrations above this threshold. This does not follow the results found with the raw blood values as most individuals from Hornøya were above this threshold. Such a heterogeneity in toxicity exposition might be explained by different time of incorporation into the tissues, or a problem of the threshold in feathers, as previously suggested.

2. Large-scale Hg contamination in the Arctic: seabirds as bioindicators of the marine ecosystem

2.1. Hg concentrations in blood adjusted by $\delta^{15}\text{N}$.

The contamination by Hg is closely related to the trophic level of an individual (biomagnification), which is commonly studied with the use of nitrogen stable isotope ($\delta^{15}\text{N}$). Within the Arctic, birds' diet and thus their trophic position vary between species, populations, but also between individuals. We also showed in the present doctoral work spatial variations in Hg concentrations with for example higher Hg concentrations in seabirds breeding in the West-Atlantic than in the East-Atlantic. If Hg concentrations are spatially the same, then spatial variations found in seabirds might be due to different trophic levels. However, if Hg concentrations are spatially different, then seabirds might feed on similar trophic levels that are differently contaminated. Therefore, if we want to understand Hg spatial variation at a large scale, we can use seabirds as bio-indicators of their environment. In order to take into account any potential change in trophic position and allow temporal and spatial comparisons, previous studies suggested to adjust Hg concentration by $\delta^{15}\text{N}$ (Braune 2007, Burgess et al. 2013, Braune et al. 2016, see **Papier 1**) with the following equation:

$$Hg_{adjusted} = Hg_{measured} + A[(\delta^{15}\text{N})_{average} - (\delta^{15}\text{N})_{measured}]$$

where A is the regression coefficient for $\delta^{15}\text{N}$ (or biomagnification slope), $\delta^{15}\text{N}_{average}$ are the mean values by species and $\delta^{15}\text{N}_{measured}$ are the values measured for each individual.

Blood represents relatively local and short-term diet, and therefore Hg exposure. In the present sub-section, in order to make large-scale comparison of Hg concentrations in blood, we therefore

propose to use seabirds as bio-indicators of their environment and to adjust their blood Hg concentrations with individual $\delta^{15}\text{N}$ and thus trophic position. Also, $\delta^{15}\text{N}$ values at the base of the food chain vary spatially. Subsequently, the $\delta^{15}\text{N}$ values will also vary at the top of the food chain. Therefore, individuals from a same species coming from two different areas can feed at the same trophic level but might have different $\delta^{15}\text{N}$. To take into account this variation, we used $\delta^{15}\text{N}$ values measured in common eiders (assumed to feed at a trophic level of two, whatever the breeding location) in each region as a baseline to infer such a spatial variation and adjust Hg concentrations. Hg adjusted concentrations per individual were thus calculated as follow:

$$Hg_{adjusted} = Hg_{measured} + A[(\delta^{15}\text{N})_{average} - (\delta^{15}\text{N})_{measured}]$$

where A is the slope of the linear regression of the relation between $\log_e(\text{Hg})$ and $\delta^{15}\text{N}$ (or the biomagnification slope) for each regions previously defined, $\delta^{15}\text{N}_{average}$ is the mean $\delta^{15}\text{N}$ measured in common eiders for each region previously defined and $\delta^{15}\text{N}_{measured}$ is the $\delta^{15}\text{N}$ measured for each individual (see maps blood and feathers on *Annex 2* for details on the regions).

In the present sub-section, we will therefore focus on spatial variations of Hg concentrations in blood adjusted for $\delta^{15}\text{N}$. Therefore, all species have been pooled within each sampling site. Hereafter, we will use the term “raw blood Hg concentrations” for the non-adjusted values and “adjusted blood Hg concentrations” for the adjusted values.

2.2. Results and discussion

We tested for blood adjusted [Hg] variations ($n = 3,491$) depending on the regions with the use of a linear mixed model. This model included the regions ($n = 8$) as fixed effects. Because we combined data collected during multiple years and at multiple sampling sites, but also because some individuals have been monitored for several years, the sampling year ($n = 4$), individuals ($n = 2,950$) and sampling sites ($n = 48$) were added as random effects. The mixed model indicated an effect of the regions ($F_{7,39.94} = 7.76$ $p < 0.001$).

First, and as suggested in the blood section, in the Pacific, St George seemed to be highly contaminated (Fig. 3.9). In this region, St Lawrence (North) and St George (South) are the only two sampling sites in the Bering Sea. Therefore, we can suggest that seabirds, and therefore other wildlife,

could be exposed to higher Hg concentrations in the south of the Bering Sea than in the North of the Bering Sea. In addition, when comparing Pacific and Atlantic, it seemed that Pacific presented higher concentrations.

Within the Atlantic region, our results showed higher contamination in Northwest Atlantic Arctic (ln(blood adjusted) mean \pm SD = $-0.16 \pm 0.58 \mu\text{g/g dw}$) and Sub-Arctic ($-0.28 \pm 0.33 \mu\text{g/g dw}$), in addition to Greenland Sea ($0.12 \pm 0.47 \mu\text{g/g dw}$) and lower contaminations in S. Norwegian Sea ($-0.70 \pm 0.47 \mu\text{g/g dw}$), N. Norway – Svalbard ($-0.76 \pm 0.57 \mu\text{g/g dw}$) and Barents Sea ($-1.22 \pm 0.43 \mu\text{g/g dw}$). Therefore, those results suggested an east-west increase in Hg concentrations, as already proposed in the present part of the thesis. This also supports the result found in previous research (AMAP, 2018, Provencher et al., 2014). Additionally, the Baltic seemed to have concentrations close to the ones measured in the West Atlantic ($-0.02 \pm 0.42 \mu\text{g/g dw}$), therefore suggesting higher concentrations in this area too. Overall, by adjusting blood Hg concentrations by the trophic level, we have been able to use seabirds as bio-indicators of the contamination during the breeding period, confirming the results of previous research suggesting an east-west increase gradient in wildlife. Indeed, such a method allowed us to confirm that the spatial variations in Hg concentrations are not due to the diet or the trophic chain,

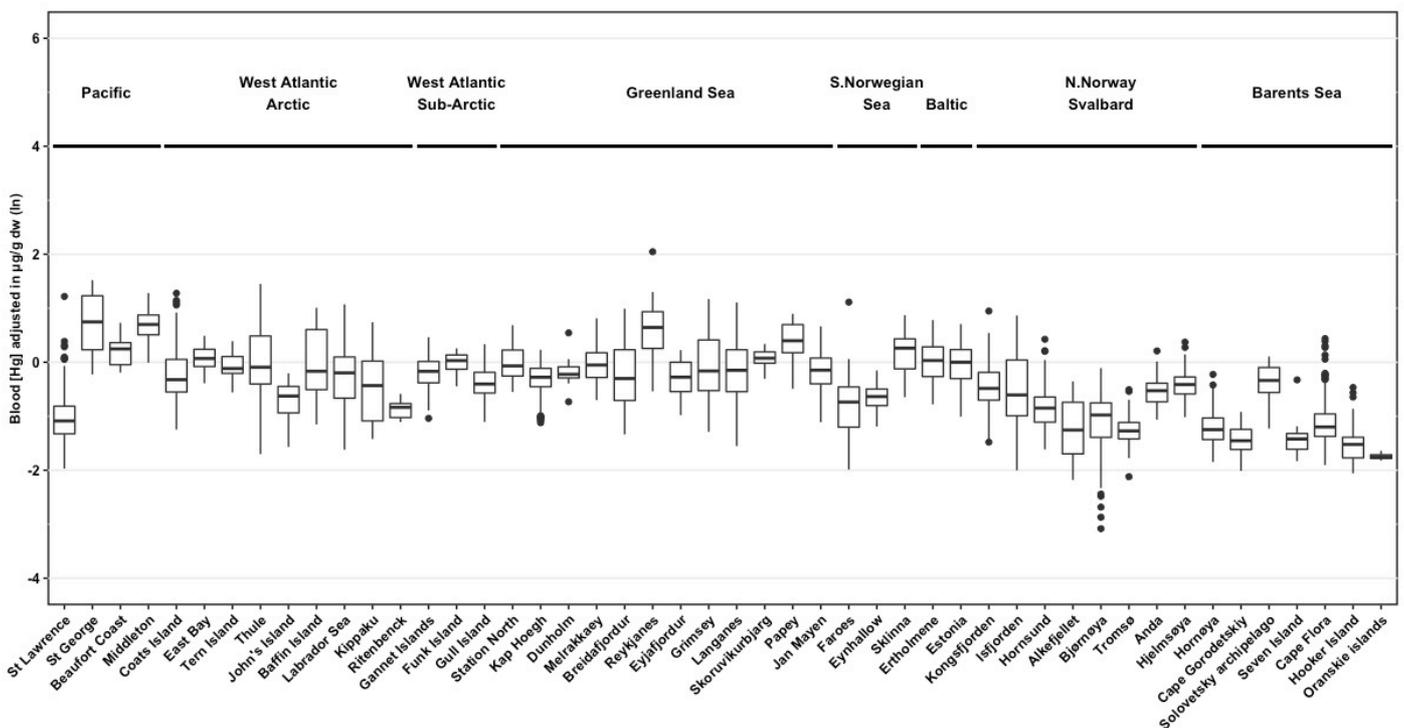


Figure 3.9: blood [Hg] adjusted by $\delta^{15}\text{N}$ presented by sampling sites and regions, following a longitudinal gradient. Each species is pooled within each site

but are due to a spatial variation of Hg at a large spatial scale within the trophic chains. Overall, these results also show the great potential of using such a method at a large scale in blood (or eggs – see Braune et al., 2007).

2.3. Conclusion

In the present section, our results showed high heterogeneity in Hg concentrations between and within species, in addition to spatial variations. **Following our hypothesis, our results suggested spatial variations in seabird Hg contamination during the breeding period.** As predicted, these variations follow previous studies, with higher Hg concentrations in the West Atlantic than in the East Atlantic. In addition, our results highlighted different Hg contaminations between the Pacific and the Atlantic regions, with higher concentrations in the Pacific. **They also follow our second prediction, with higher concentrations in piscivorous or omnivorous species.** In comparison, we covered more species into the review (n = 24), than in the present work (n = 20). On the other hand, we covered more breeding colonies in the present study (n = 50) than in the review (n = 20). However, we did not cover all the same species (e.g. king eiders, arctic terns, only 11 species common to both studies) or breeding colonies (e.g. Chaun – Russia; Kiska Islands – Aleutian Islands, only 4 breeding colonies common to both studies), making these two studies complementary. Regarding the Russian Arctic, we covered the western part, but information is still needed for the eastern part of the Russian Arctic. Overall, both blood and feathers are complementary tissues. As both tissues follow the same trend, this suggests that the feathers, when blood cannot be collected, are good proxy for studying Hg concentrations during the breeding period. Indeed, even if blood gives access to more precise and local information about Hg contamination, feathers can represent an intermolt period and therefore cover more parts of the breeding period. Finally, the use of seabirds as bio-indicators showed that large-scale spatial variations in Hg concentrations are mostly driven by a heterogeneous distribution of Hg in the Arctic environment, and not necessarily by their diet.

3. Large-scale Hg contamination during the non-breeding period and Hg seasonality: study case of the Alcids family

3.1. Introduction, hypotheses and predictions

The alcid family (Charadriiformes: Alcidae) comprises the most numerous seabirds in the Arctic and shows a circumpolar distribution, with species endemic to the Pacific [murrelets *Brachyramphus*, auklets *Aethiini* and tufted puffin *Fratercula cirrhata*], the Atlantic (little auks – or dovebies - and razorbills *Alca torda*] or common to both regions [black guillemots *Cepphus grylle*, common guillemots (or common murres) and Brünnich’s guillemots (or thick-billed murres - Gaston and Jones 1998). In the present section (based on the methods and results presented in **Paper 3**), we used body (representing the breeding period) and head feathers (representing the non-breeding period) to study seasonal variations in Hg concentrations from nine species breeding in 28 colonies through the pan-Arctic (Fig. 3.11).

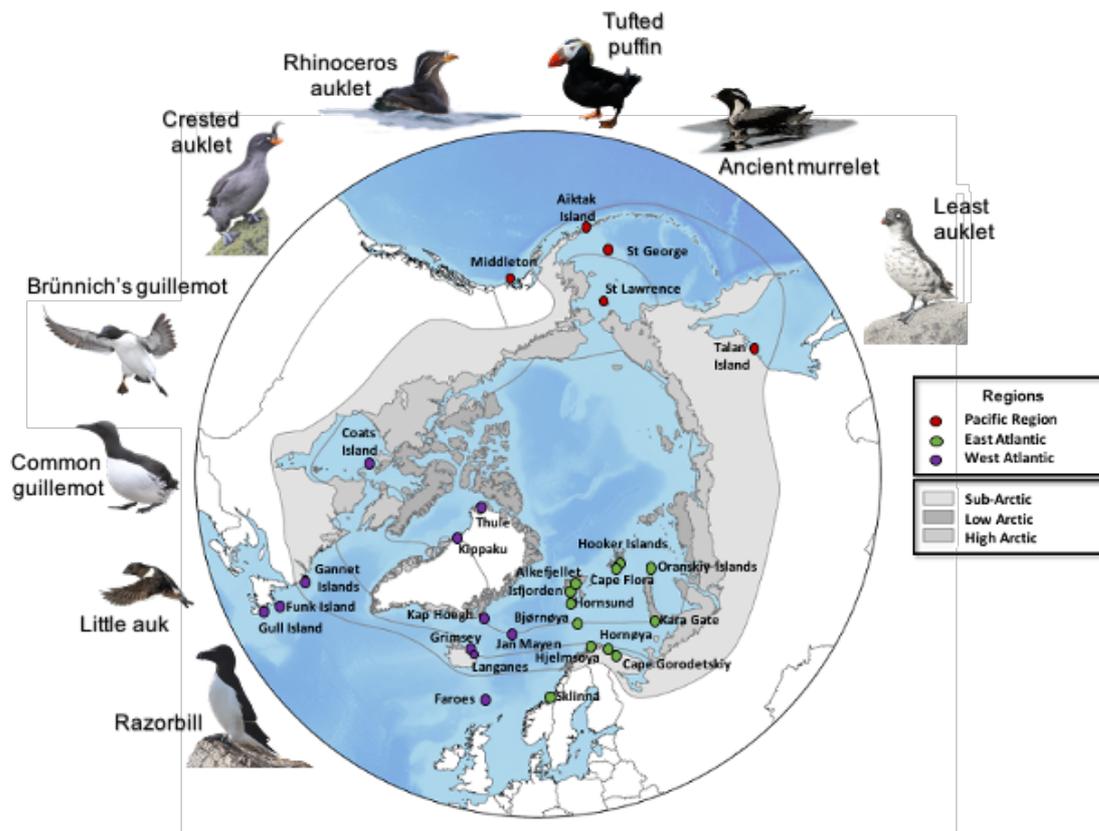


Figure 3.11: colonies where alcids have been sampled for the need of the present part (**Paper 3**). The study sites were classified in three regions. The name of the colonies was added in the map and colored depending on the areas they fall on. Sub, Low and High Arctic are represented following the Conservation of Arctic Flora and Fauna delimitations (CAFF, 2001)

Based on previous studies suggesting seasonal variations in Hg concentrations in Arctic seabirds (Fort et al. 2014) in addition to spatial variations in Hg through the Arctic (AMAP, 2018 – see **Chapter 1**), we hypothesized seasonal variations in Hg concentrations and predicted i) higher Hg concentrations during the non-breeding period than during the breeding period, ii) spatial and interspecific differences in Hg seasonality.

3.2. Results and discussion.

3.2.1. *Hg concentrations during the non-breeding season.*

In this section, the pan-Arctic area was divided in three regions being the “Pacific Region”, “East Atlantic” and “West Atlantic” (Fig. 3.11). On average we found Hg concentrations during the non-breeding period to be 3.62 ± 2.43 $\mu\text{g/g dw}$. The highest mean Hg concentrations during the non-breeding period were observed in the West Atlantic (5.42 ± 2.52 $\mu\text{g/g dw}$) followed by the Pacific (2.64 ± 2.07 $\mu\text{g/g dw}$) and the East Atlantic (2.52 ± 1.46 $\mu\text{g/g dw}$ – Fig. 2.12A). The lowest mean Hg concentration during the non-breeding period was measured in crested auklets (1.45 ± 0.58 $\mu\text{g/g dw}$) while the highest was in rhinoceros auklets (6.89 ± 2.04 $\mu\text{g/g dw}$ – Fig. 2.12B). These first results suggest large spatial and interspecies variations in [Hg] during the non-breeding period. However, both inter and intraspecies variations also suggest either different diets or different exposure depending on the location of the bird’s wintering ground (see below).

3.2.2. Hg seasonality

We tested for seasonal and regional differences in Hg concentrations. Our results indicated that differences between breeding and non-breeding Hg concentrations varied according to regions. The largest seasonal Hg difference was found in the West Atlantic (ratio summer to winter: $1.13 \pm 0.02 \mu\text{g/g dw}$) and the East Atlantic ($1.13 \pm 0.02 \mu\text{g/g dw}$) followed by the Pacific ($0.57 \pm 0.04 \mu\text{g/g dw}$) (Fig. 2.12A). We also tested for seasonal and species variation in Hg concentrations. Our results indicated that differences between breeding and non-breeding Hg concentrations varied according to species. The largest seasonal differences were found in Brünnich's guillemots (estimate \pm SE: $1.15 \pm 0.02 \mu\text{g/g dw}$), common guillemots ($1.12 \pm 0.02 \mu\text{g/g dw}$), little auks ($0.82 \pm 0.03 \mu\text{g/g dw}$) and razorbills ($1.50 \pm 0.07 \mu\text{g/g dw}$); while no significant seasonal variation in Hg concentrations was found in ancient murrelets ($0.36 \pm 0.21 \mu\text{g/g dw}$), crested auklets ($0.41 \pm 0.13 \mu\text{g/g dw}$), least auklets ($0.23 \pm 0.09 \mu\text{g/g dw}$), rhinoceros auklets ($0.75 \pm 0.11 \mu\text{g/g dw}$) and tufted puffins ($0.29 \pm 0.14 \mu\text{g/g dw}$) (Fig. 2.12B). **Hence, the study of Hg seasonality in alcids revealed spatial and interspecies variations. As predicted, we found clear seasonal differences in Hg concentrations for most of study sites and species, confirming higher Hg exposure during the non-breeding season.** These results also highlight

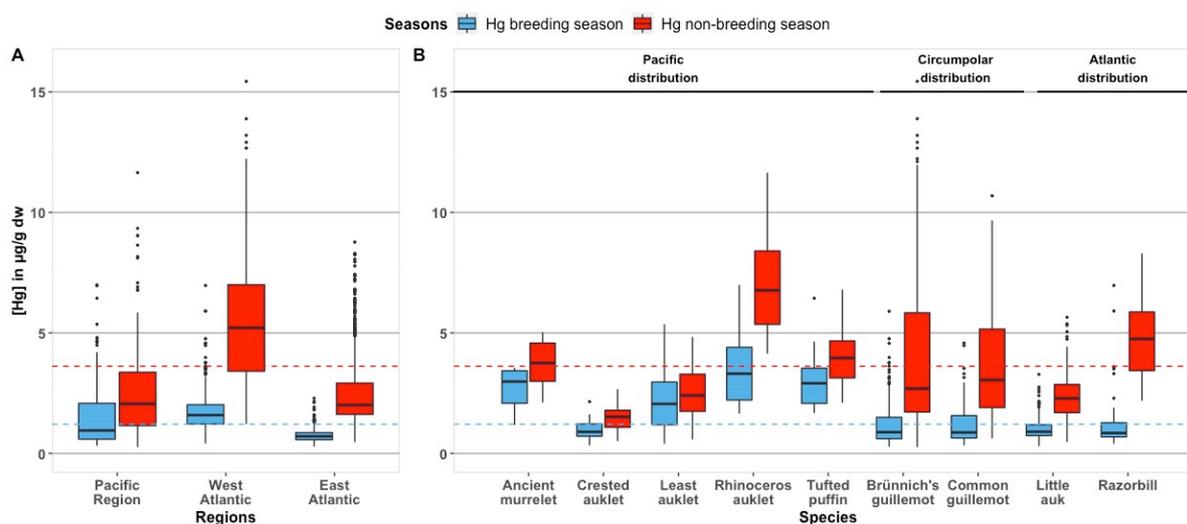


Figure 2.12: Mean Hg concentrations during the breeding season (blue - represented by the body feathers) and non-breeding season (red – represented by the head feathers) per A) regions and B) species. The species are sorted out by their distribution, and then alphabetically. Mean Hg concentrations for BF ($1.21 \pm 0.83 \mu\text{g/g}$, blue) and HF ($3.62 \pm 2.43 \mu\text{g/g}$, red) are represented by dashed lines. Mean Hg concentrations and SD per species are presented in Paper 3, Table 1 and Table S1. Boxplots show the median (horizontal black line within the boxes), 1st and 3rd quartiles, the minimum (1st-1.5x interquartile range) and maximum (3rd-1.5x interquartile range) concentrations (lower and upper whiskers) and outliers.

differences between the Pacific and the Atlantic regions. Three non-exclusive hypotheses could explain Hg seasonal variations in migratory species: 1) the occurrence of a dietary shift between seasons, 2) different food chain lengths on breeding and non-breeding sites, 3) contrasting environmental contamination between sites.

Dietary shift or food-chain length to explain Hg seasonality and higher [Hg] during non-breeding period?

A seasonal dietary shift towards higher trophic level prey should lead to higher Hg concentrations. For example, our results showed that for several of our studied species, higher Hg concentrations were found during the non-breeding period when we would have expected lower concentrations due to a dietary shift to lower trophic level (e.g. Brünnich's guillemots) or a constant Hg concentration due to a relatively constant diet throughout the year (e.g. little auks, common guillemots, razorbills – *Annex 1* for diet information). A few studies investigated food web structure in the Arctic (e.g., Baffin Bay; Hobson and Welch, 1992; Linnebjerg et al., 2016) and subarctic marine systems (e.g., continental shelf off Newfoundland; Sherwood and Rose, 2005). These findings suggested slightly decreasing food chain lengths and again contrasted with higher Hg contamination in birds during the non-breeding period, suggesting other processes. In only one case, a change in diet between breeding and non-breeding periods could explain the enhanced Hg contamination during winter (e.g. rhinoceros auklets). Overall, our results suggest that the diet changes alone cannot explain the seasonal changes and increased winter

Hg contamination we found for most alcid species and areas. They rather suggest a dominant role of bird migration strategies and distribution, both during summer and winter.

Contrasting environmental contamination to explain Hg seasonality?

Our results highlighted spatial differences in seabird Hg contamination during the non-breeding period. We found higher mean winter Hg concentrations in seabirds populations breeding in the Northwest Atlantic (which are likely overwintering in the Labrador Sea, [McFarlane Tranquilla et al., 2014](#)) compared to the Northeast Atlantic seabirds, overwintering farther East (Fort et al., 2013; Stein et al., 2013). Such results are in agreement with the earlier reported higher Hg concentrations in little auks overwintering off Newfoundland (Fort et al. 2014) and suggest a Hg hotspot in the Labrador Sea. In a few cases only, we found low or no seasonal variations in Hg concentrations, in populations where Hg contamination was constant year-round. This almost exclusively occurred in Pacific species and populations (i.e. ancient murrelets, crested and least auklets) as well as in one little auk population breeding in northwest Greenland (i.e. Thule). This absence of seasonal variation in the Pacific could be due either to a similar exposure on both their breeding and non-breeding sites or to a resident strategy of birds staying close to the breeding grounds year-round (Gaston and Jones, 1998).

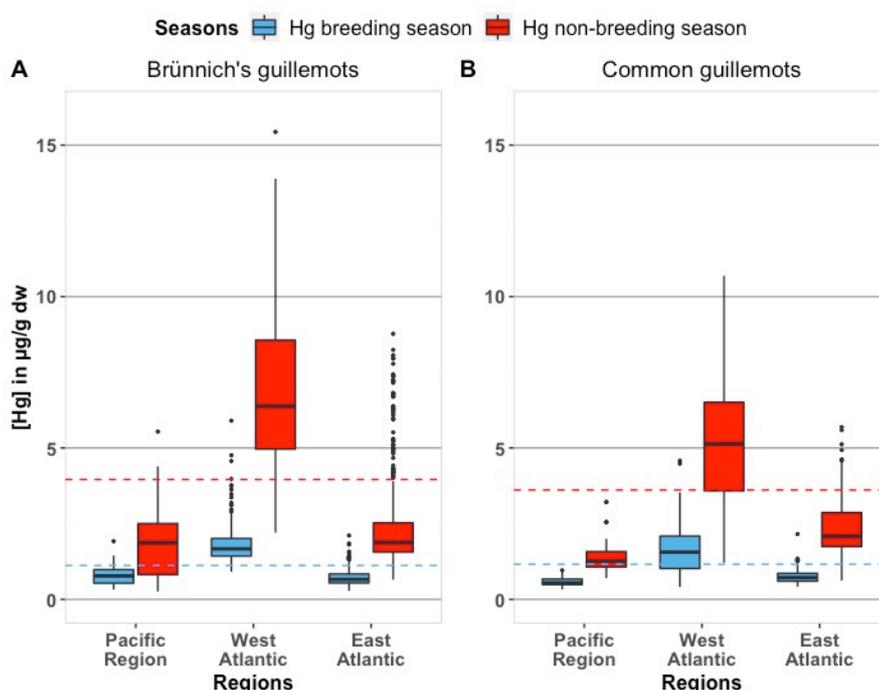


Figure 3.13: Mean Hg concentrations during the breeding season (blue - represented by the body feathers) and non-breeding season (red - represented by the head feathers) for the A) Brünnich's guillemots and the B) common guillemots. Mean Hg concentrations for Brünnich's guillemots (BF = 1.12±0.70 and HF = 3.96±2.91 µg/g dw) and common guillemots (BF=1.16±0.70 and HF = 3.61±2.05 µg/g dw) are represented by blue dashed lines for BF and red dashed lines for HF. Boxplots show the median (horizontal black line within the boxes), 1st and 3rd quartiles, the minimum (1st-1.5x interquartile range) and maximum (3rd-1.5x interquartile range) concentrations (lower and upper whiskers) and outliers.

3.2.3. *Pacific vs Atlantic*

Populations of ancient murrelets, crested and least auklets, and tufted puffins from the Pacific presented low or no seasonal variation in their Hg concentrations, while we found significant seasonal variation in most of the Atlantic populations (excepted the little auk population from Thule, West Greenland). We believed these variations were due to spatial differences in environmental contamination. To study specifically this point, we analyzed seasonal variations for the Brünnich's (Fig. 3.13A) and common guillemots (Fig. 3.13B), the two species being sampled in all three regions. When testing for spatial variations in Hg seasonality, we found a similar pattern, with the lowest seasonal differences and lower non-breeding Hg concentrations in the Pacific compared to the Atlantic regions (Fig. 3.13), suggesting differences in Hg environmental contamination between both regions. The Arctic Ocean is a sink for contaminants such as Hg, with inflows from both atmospheric and oceanic currents, as well as rivers (Sonke et al. 2018). Nevertheless, important outflows of Hg also occur from the Arctic at lower latitudes (AMAP 2011). Because the Atlantic region is widely opened with more complex and numerous atmospheric and oceanic fluxes than the Pacific region, oceanic outflows of Hg (e.g. through the Arctic Canadian Archipelagoes and Fram Strait; Outridge et al., 2008) could partly explain the higher concentrations in Hg found in species from the Western Atlantic region compared to the Pacific region.

3.3. Conclusion

Our large-scale study of Hg revealed spatial differences in Hg seasonality, with higher exposition during the non-breeding period compared to the breeding period. As seabirds feeding at a low trophic level had unexpected higher Hg concentrations, we suggested that seasonal Hg variations cannot be explained by the diet alone but are rather explained by seabirds' seasonal migrations and distribution. Overall, this revealed the importance of the winter period on seabirds' contamination to Hg.

In the present chapter the study of Hg concentrations during the breeding period allowed us to highlight spatial variations in Hg concentrations, following results of previous studies. We also found a

seasonality in Hg concentrations, with higher concentrations during the non-breeding period. This reveals the importance of this period in terms of contaminations and the need to study spatial variations in Hg concentrations during the non-breeding period. Indeed, this will be essential in order to see if spatial variations are similar throughout the year, to therefore extend this knowledge to larger marine areas, acknowledge areas at risk for wildlife and thus set up appropriate conservation measures throughout the year to protect wildlife from pollutants.

4. Summary of Chapter 3

Breeding period

- Hg concentrations vary spatially, increasing following an east-west gradient. These spatial variations are in accordance with previous studies on seabirds and other wildlife.
- Blood and body feathers both inform about the breeding period, but with a different integration period (blood: few weeks, feathers: intermolt).
- We filled gaps by covering 20 species, in areas not previously monitored with the use of blood and feathers.
- Overall, using seabirds as bio-indicators by adjusting blood Hg concentrations by $\delta^{15}\text{N}$ give similar spatial trend as with raw blood or feathers and showed that large-scale spatial variations in Hg concentrations are mostly driven by a heterogeneous distribution of Hg in the Arctic environment, and not necessarily by their diet.

Non-breeding period

- The use of specific feathers can inform about Hg contamination during the non-breeding period in Arctic seabirds, when they are physically not accessible.
- Only one study found seasonality in Hg contamination in one population of little auks breeding in the High Arctic. Our results allow us to extend and generalize this finding at the Pan-Arctic scale for multiple species.
- A diet switch alone cannot explain the seasonal variations and increased winter Hg contamination we found for most alcid species and areas. This suggests a dominant role of bird migration strategies and distribution, both during summer and winter.
- This Hg seasonality varies spatially with the highest concentrations and variations in the West Atlantic.
- Atlantic and Pacific Hg variations might be explained by environmental structure differences and fluxes (e.g. inputs, outputs)

Chapter 4

Does seabird migration play a role on Hg exposure at the individual level?



In the previous chapter, we highlighted seasonal variations in Hg concentrations with higher concentrations measured during the non-breeding period, revealing the importance of this period. As seabirds feeding at a low trophic level had unexpected high concentrations, we suggested that these variations could not be explained by the diet alone. We proposed that it might rather be explained by seabirds' migrations and distributions. Such results call for further studies linking seabirds' individual movements and their Hg contaminations, which will be studied in the present chapter. This chapter is based on the methods and results of **Paper 4**.

1. Introduction, hypotheses and predictions

Seabirds are typically migratory species, many travelling hundreds to thousands of kilometers every year to reach their wintering grounds (e.g. Egevang et al., 2010, Frederiksen et al., 2016; McFarlane Tranquilla et al., 2014). However, these migratory strategies can be highly variable between individuals, conditioning their distribution and thus their exposure to Hg. First, different individuals from a same population can use contrasting wintering areas. For instance, red-legged kittiwakes breeding at Saint George Island (Pribilof Islands - Pacific) overwinter in different areas, ranging from the Sea of Okhotsk to the Bering Sea, with higher Hg concentrations measured in seabirds wintering in the southern areas (Fleishman et al. 2019). Second, each individual can itself have different interannual migratory strategies and thus contrasting fidelity to its wintering grounds, with species showing high (e.g. common eider, Atlantic puffin, European shag *Phalacrocorax aristotelis*; Guilford et al. 2011, Petersen et al. 2012, Grist et al. 2014), low (e.g. thin-billed prion *Pachyptila belcheri*; Quillfeldt et al. 2010) or variable (e.g. Brünnich's and common guillemots, northern fulmar; Hatch et al. 2010, McFarlane Tranquilla et al. 2014, Orben et al. 2015) individual fidelity to wintering grounds.

In the present chapter, we combined data from individual geolocators with parallel analyses of Hg in the feathers of six seabird species using Arctic and sub-Arctic wintering areas. This study has been performed on black-legged kittiwakes, Brünnich's and common guillemots, common eiders, little auks and northern fulmars. This multi-species approach allowed us to extend our study to species using different habitats and environment, experiencing different migratory strategies and thus probably facing different exposure to Hg. **Hence, we studied interannual seabird winter distribution in relation to**

winter Hg contamination on multi-years tracked individuals (Fig. 4.1) and tested the hypothesis that winter migration and wintering ground fidelity affect their contamination to Hg. More specifically, we predicted that individuals with low fidelity to their wintering grounds from one year to the other show more variable Hg concentrations in comparison to faithful ones, reflecting the influence of the wintering grounds location.

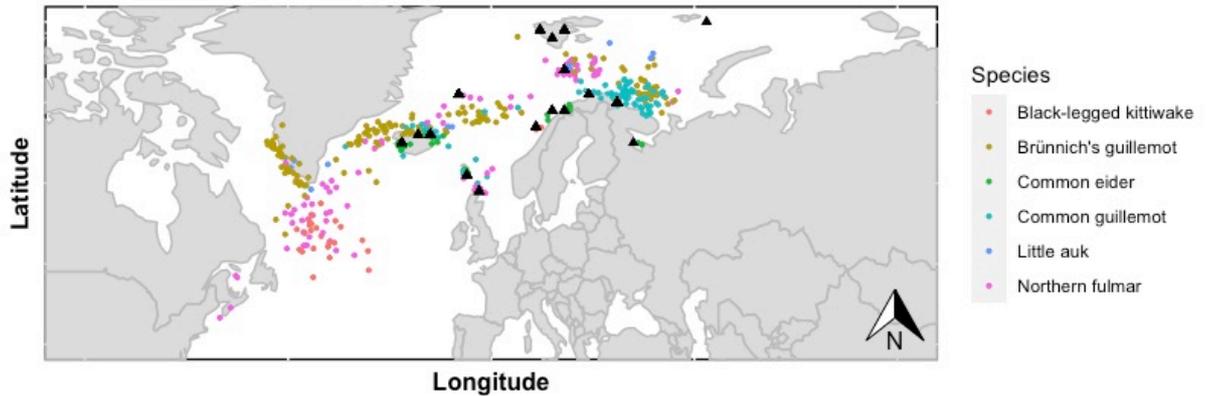


Figure 4.1: Winter distribution of black-legged kittiwakes, Brünnich's guillemots, common eiders, common guillemots, little auks (median position from November to January) and northern fulmars (median position from November to December). Sampling sites (Table S1) are identified by (▲).

2. Results and discussion

2.1. How does faithfulness to wintering grounds impact mercury contamination?

Following the methods described in **Chapter 2**, Hg concentrations representing the non-breeding period were measured in body feathers for northern fulmars (belly), common eiders (belly) and black-legged kittiwakes (back) and head feathers for alcids. In addition, we extracted for each individual a winter median position for the winter period, reflecting its winter distribution and defined from November to December for northern fulmars and from November to January for Brünnich's and common guillemots, little auks, common eiders and black-legged kittiwakes.

We first defined individuals with low or high fidelity to their wintering grounds. Phillips and colleagues (2004) showed that the accuracy of seabird positions using GLS is 186 ± 114 (mean \pm SD) km. Given this accuracy, the scale of the study area, and that the different study species can be more or less mobile during winter, we considered the limit of 372 km to discriminate individuals with high or

low fidelity to their wintering grounds. Based on this, individual birds fell into the “high fidelity” group when their consecutive winter median positions were <372 km apart. Conversely, birds were considered to belong to the “low fidelity” group when they wintered in distinct geographical areas between years (i.e. winter median positions >373 km apart) (see **Paper 4** for detailed methods). To study the variation in Hg exposure from one year to the other, we used the difference of Hg concentrations from one year to the other (Δ [Hg]).

The common eider was the only species to consistently show high fidelity to its wintering ground. This was reflected by low variations in the Δ Hg concentrations (hereafter Δ [Hg] - absolute value) (Fig. 4.2).

Table 4.1: Mean [Hg] (mean \pm SD $\mu\text{g/g dw}$) during the non-breeding period, including the number of observations (n), in addition to the number of individuals having a high, low, or both fidelities for their wintering ground from one winter to the other.

Species	Mean [Hg] \pm SD (n obs)	Individuals (n)	High fidelity (n)	Low fidelity (n)	Variable fidelities (n)
Black-legged kittiwakes	3.10 \pm 1.67 (26)	13	5	8	0
Brünnich’s guillemots	4.09 \pm 2.24 (157)	65	40	18	7
Common guillemots	2.73 \pm 1.18 (89)	39	18	15	6
Little auks	2.93 \pm 1.23 (12)	6	3	3	0
Northern fulmars	2.34 \pm 1.16 (83)	38	13	21	4
Common eiders	0.81 \pm 0.29 (41)	20	22	0	0

Individuals of all other species showed both a mixture of high and low fidelity (Fig. 4.2) and individuals tracked for more than two years even presented a mixture of low or high fidelity (Table 4.1). Overall, we found that Δ mean [Hg] (absolute) was significantly different between the high (mean \pm SD, with eiders = 0.80 \pm 0.81 $\mu\text{g/g dw}$; without eiders = 0.89 \pm 0.84 $\mu\text{g/g dw}$) and low (1.19 \pm 0.98 $\mu\text{g/g dw}$) fidelity groups (Fig. 4.2), with (Wilcoxon’s test: $W = 5649.5$, $p = 0.0002$) or without ($W = 5294.5$, $p = 0.009$) including common eiders. Individuals experiencing high fidelity to their wintering grounds showed less inter-annual variations in [Hg] (mean absolute Δ [Hg] \pm SD, including eiders = 0.80 \pm 0.81 $\mu\text{g/g dw}$; excluding eiders = 0.89 \pm 0.84 $\mu\text{g/g dw}$) than individuals with low fidelity to their wintering grounds (1.19 \pm 0.98 $\mu\text{g/g dw}$ - Fig. 4.2). The lower variations in Hg concentrations for individuals with high winter ground fidelity were observed in all species but the little auk (Fig. 4.2). This difference between low and high-fidelity individuals was mostly pronounced in black-legged kittiwakes and

common guillemots (Fig. 4.2). **Following our prediction, these results suggest that, regardless of common eiders only experiencing high fidelity for their wintering grounds, Hg concentrations of individuals tend to be stable when consistently migrating to the same wintering ground, while they vary when migrating in different areas from one year to the other.**

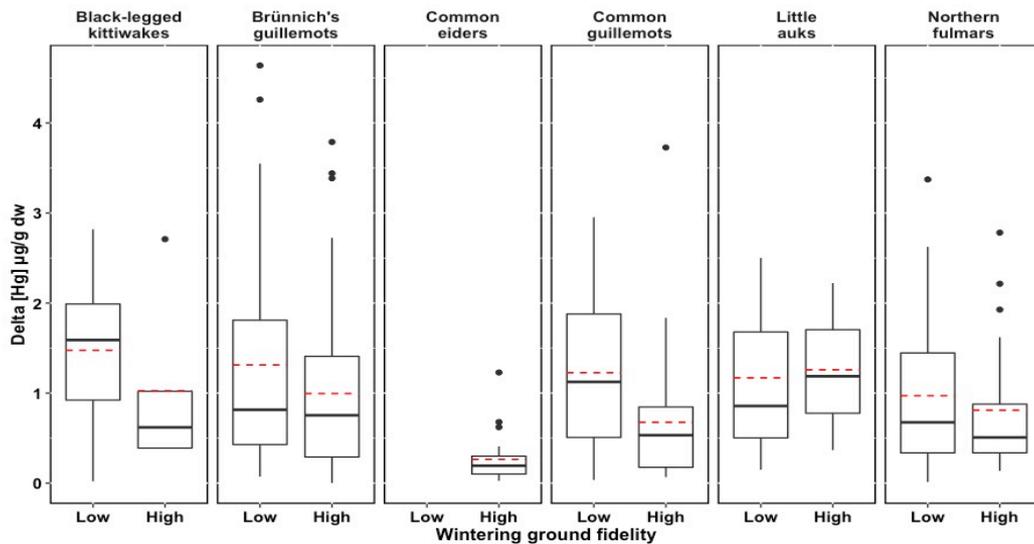


Figure 4.2: Δ [Hg] (absolute) from one year to the other for birds with low (<372km) or high fidelity (>372km) for their wintering grounds, per species. The mean Δ [Hg] is represented by a red dotted line. Boxplots show the median (horizontal black line within the boxes), 1st and 3rd quartiles, the minimum (1st-1.5x interquartile range) and maximum (3rd-1.5x interquartile range) concentrations (lower and upper whiskers) and outliers.

Seabirds are exposed to various environmental threats during the winter period (e.g. oil spills, storms, contaminants; Frederiksen et al. 2012, Petersen et al. 2012, Fort et al. 2014, Guéry et al. 2017) which might impact their condition, survival or subsequent reproduction (Votier et al. 2005, Mesquita et al. 2015, Anker-Nilssen et al. 2016). The large-scale distribution of Hg in the marine environment is assumed to be relatively stable over the years. Hence, high fidelity to their wintering ground likely expose birds to repeated threats and constant pollution issues associated to this specific area (e.g. human industries), possibly impacting their short-term survival and reproduction with long-term impacts on their population dynamics (Guéry et al. 2017, 2019). On the other hand, seabirds wintering constantly in an area with a low level of such threats might only be exposed to limited acute pollution events, which could have severe effects on their survival and subsequent reproduction but limited long-term impacts on their populations. Conversely, seabirds with low fidelity to wintering grounds exploit contrasting wintering environments with different Hg concentrations years after years (Quillfeldt et al., 2010) which might lower long-term risks associated with Hg contamination.

2.2. How does inter-annual variation in Hg contamination relates to spatial variations in migratory grounds?

For species experiencing low fidelity to their wintering grounds (all species, excepted the common eider – Fig. 4.3), we used generalized linear models (GLMs) to test Hg variations depending on the directions (North-West, North-East, South-West, South-East) and the distance (in km) travelled from one winter to the other. First, our results showed that the Δ mean [Hg] were higher for the Brünnich's guillemots than for the other species (Fig. 4.3). This might be explained by the fact that they winter in more contrasted areas from one winter to the other compared to the other species. Then, when including all species into the GLM analyses, we found that the inter-annual variation in [Hg] varied among species and were associated with both the directions and the distance (in km) between two median winter positions (Paper 4, Table 2). These results showed that Hg concentrations increased when individuals changed their winter distribution to the northwest direction and decreased when individuals changed to the southwest direction, and these effects were similar among species (Fig. 4.4A & B).

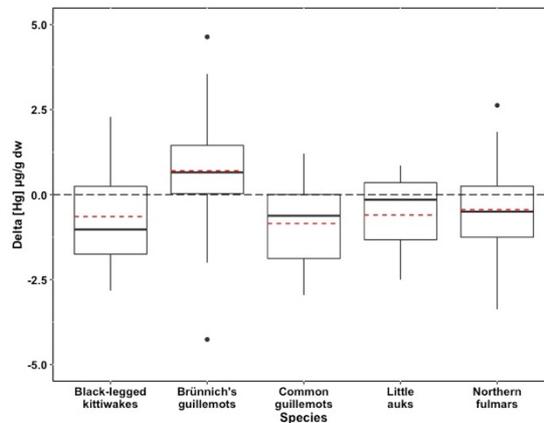


Figure 4.3: Δ [Hg] for the five species experiencing low fidelity for their wintering grounds. The mean Δ [Hg] is represented by a red dotted line. The zero axis is represented by a black dotted line. Boxplots show the median (horizontal black line within the boxes), 1st and 3rd quartiles, the minimum (1st-1.5x interquartile range) and maximum (3rd-1.5x interquartile range) concentrations (lower and upper whiskers) and outliers.

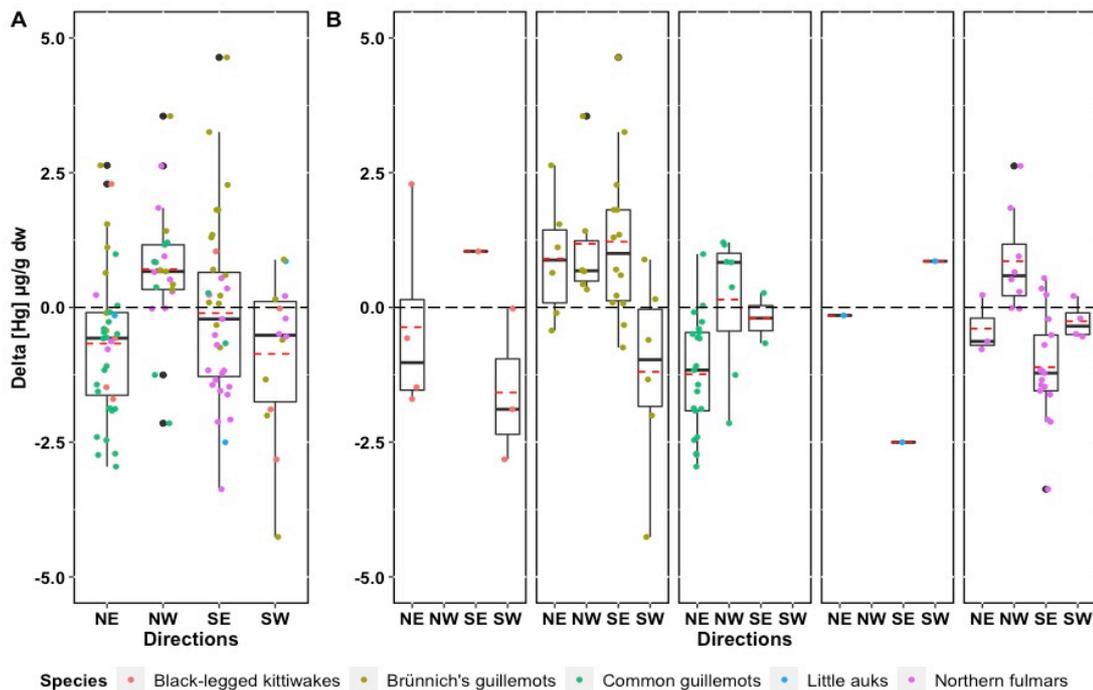


Figure 4.4: Δ [Hg] in the fourth different directions (NE=North-East, NW=North-West, SE=South-East, SW=South-West) in species and individuals with low fidelity for their wintering grounds for A) all species and B) per species. The mean Δ [Hg] is represented by a red dotted line. The zero axis is represented by a black dotted line. Boxplots show the median (horizontal black line within the boxes), 1st and 3rd quartiles, the minimum (1st-1.5x interquartile range) and maximum (3rd-1.5x interquartile range) concentrations (lower and upper whiskers) and outliers.

The absence of interaction between the species and the distance in the GLM analyses indicated that the effect of the distance on [Hg] variation varied among species. More specifically, our results indicated that Δ [Hg] was positively correlated with the distance for black-legged kittiwakes (estimate \pm SE = $-1.62 \pm 1.02 \mu\text{g/g dw}$) and Brünnich's guillemots ($-0.07 \pm 0.47 \mu\text{g/g dw}$) and negatively correlated with common guillemots ($-0.52 \pm 0.45 \mu\text{g/g dw}$), northern fulmars ($-0.29 \pm 0.42 \mu\text{g/g dw}$) and little auks ($1.19 \pm 1.00 \mu\text{g/g dw}$) (Fig. 4.5). This suggests that more Brünnich's guillemots and black-legged kittiwakes migrate far from their previous wintering ground, more their Hg concentrations increase. On the other hand, this suggests that the more the little auks, common guillemots and northern fulmars migrate far from their previous wintering grounds, the more their Hg concentrations decrease (Fig. 4.5). However, the low sampling size regarding little auks (three interannual observations only) might be interpreted carefully and might require more investigation with a higher sampling size. **Overall, this suggest a heterogeneous distribution of Hg in the marine environment at a large spatial scale.**

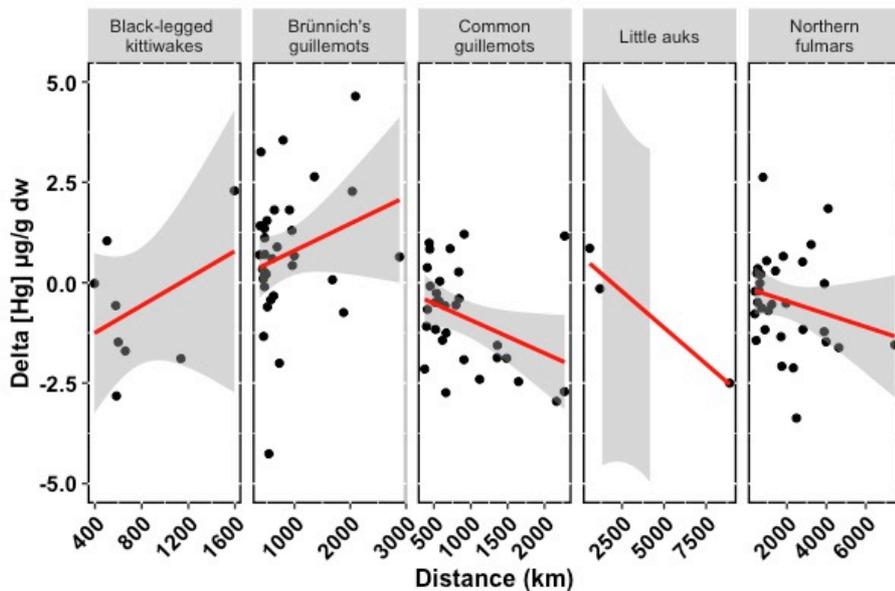


Figure 4.5: Δ [Hg] in relation to the distance (in km) for the five studied species and individuals with low fidelity for their wintering grounds.

3. Conclusion

In the present chapter, the different species had different fidelity to their wintering grounds which could differently impact their exposition to pollutants, and subsequently have different implications on their survival and population dynamics. In addition, wintering Brünnich's guillemots and northern fulmars covered a large part of the North Atlantic, while common guillemots, little auks and black-legged kittiwakes were distributed over a part of it only (Fig. 4.1). With such a large-scale study, seabirds found on the eastern (or western) areas for the first year of our assessment had more chance to migrate in the western (or eastern) areas the following years. Unfortunately, such a point was difficult to incorporate into the statistical analysis. But despite these spatial differences, our results showed that their Hg concentrations similarly varied between years according to changes in their distribution. **This supports the hypothesis of a heterogeneous distribution of Hg in the marine environment at a large spatial scale. Overall, these results confirm our hypotheses that seabird winter migration and wintering ground fidelity affect their Hg contamination at the individual scale.**

We also highlighted that winter distribution plays a role on seabirds' exposure to Hg and highlighted spatial differences in Hg concentrations through the North-Atlantic Arctic, with an increasing east-west

gradient, following the results presented in the previous chapter. Previous studies focused on the Arctic wildlife also suggested an increasing east-west gradient in Hg concentrations during spring and summer (breeding period), with species distributed in the Canadian Arctic being more contaminated than those in the European Arctic (Provencher et al. 2014, AMAP 2018, **Paper 1**). In the previous chapter, we also suggested the existence of this spatial pattern during the non-breeding period. Such a pattern could be explained by spatially contrasting Hg inputs in these regions (e.g. atmospheric, rivers, subsurface MeHg enrichment in global oceans; see AMAP 2018, Sonke et al. 2018, Wang et al. 2018). However, so far, little was known about spatial Hg distribution in wildlife during fall and winter. Studying such patterns and collecting samples is indeed logistically more complicated during these periods and had only been performed in two studies focused on Arctic seabirds (Fort et al. 2014, Fleishman et al. 2019). Our results suggested a similar pattern during the non-breeding period but calls for more large-scale ecotoxicological studies to confirm such a trend and highlight potential hotspots of Hg contamination. This will be the focus of the following chapter.

4. Summary of Chapter 4

- Individuals with low fidelity for their wintering grounds experience higher Hg concentrations variations than faithful individuals.
- The variations in directions and distances from one winter to the other affect seabirds' exposure to Hg, strengthening our hypotheses of the role played by winter movements and distribution in seabirds' contamination to Hg.
- We highlighted spatial differences in Hg concentrations through the North-Atlantic Arctic, with an increasing east-west gradient during the non-breeding period.

Chapter 5

Spatial ecotoxicology: determining the spatial origin of Hg contamination



In the previous chapters, we highlighted higher Hg concentrations accumulated by Arctic seabirds during the non-breeding period than during the breeding period. Subsequently, we hypothesized that seabird migration and wintering grounds played an important role on their exposure to Hg. This hypothesis was strengthened by our temporal variation study and the association we found between seabirds' wintering grounds and Hg variations. We also found an east-west increase in Hg concentrations, following previous studies focused on the breeding season (AMAP, 2018; Provencher et al., 2014). To go further, we will now study Hg distribution at the North-Atlantic Arctic scale, in addition to the spatial origin and sources of Hg contamination. To do so, we will first describe intraspecies variations in Hg contamination for seven different species during the non-breeding period. Then, we will use those seven species as bio-indicators to track Hg distribution through the North-Atlantic Arctic and the existence of potential hotspots. This part will be based on the results of **Paper 5**. Then, we will use an original method including seabirds' tracking and Hg isotopes to determine the source of Hg contamination. This part will be based on the results of **Paper 6**.

1. Are there hotspots of Hg?

1.1. Introduction, hypotheses and predictions

To protect the environment from Hg toxicity, the United Nations Environment Program (UN Environment) established the Minamata convention in 2013, inviting the signatory countries to decrease their emissions. To follow and improve the efficiency of such program, extensive and large-scale Hg studies are essential to understand Hg transportation, the origins of spatial contamination (Heimbürger et al. 2015, Pomerleau et al. 2016, Bowman et al. 2016, Wang et al. 2018, Evers & Sunderland 2019) and the associated risks for the humans and the wildlife (Dietz et al. 2013, AMAP 2018), but also to cover underrepresented areas such as the Arctic or the North-Atlantic marine areas (UN Environment Programme 2019).

As previously mentioned, high Hg concentrations have been measured in the Arctic, with the Canadian Arctic presenting some of the highest concentrations (AMAP, 2018). Additionally, global change has induced a rapid rise of temperature (ACIA 2004, AMAP 2011a), causing a fast decrease of

sea ice cover. This could induce an exacerbation of Hg release in the Arctic and subsequently to the North-Atlantic (i.e. outflux). However, and mostly because of logistical reasons, most of this knowledge is limited to the summer period, thus only covering areas relatively close to the breeding colonies and therefore neglecting a large part of the Arctic and pan-Arctic marine areas (but see Fort et al. 2014, Fleishman et al. 2019). Therefore, there is a need to closely monitor this pollutant at a larger spatial and temporal scale (e.g. marine areas, non-breeding period).

Here, we combined Hg analyses and biologging (Fort et al. 2014) to provide new information about winter Hg distribution at the North-Atlantic Arctic scale. For this purpose, we used Arctic seabirds as bio-indicators of their marine environment. In order to broadly cover marine environment, we chose seven species relying on different marine habitats and/or preys, being Atlantic puffins, Brünnich's and common guillemots, common eiders, little auks, northern fulmars and black-legged kittiwakes. These species overwinter from Canada to Russia, cover different ecologies (e.g. diets, feeding areas) and habitats (e.g. open sea, coastal areas), therefore allowing us to broadly cover Hg distribution in different wintering areas and trophic levels through the North-Atlantic Arctic. To do so, and in order to take into account Hg variability depending on the different variables, we used a method including both Generalized Additive Models (GAMs) and interpolation (kriging; Pebesma 2004), following the method used in previous studies (Bishop & McBratney 2001, Hengl et al. 2004).

Thus, we tested the hypotheses that there are hotspots of Hg and predicted that they occur in the Northwest Atlantic (Borgå et al. 2006, Provencher et al. 2014, AMAP 2018). To do so, we will present and describe intra and interspecific spatial variations. This section is based on the methods and results of **Paper 5**.

1.2. Results and interpretations

1.2.1. *Intraspecific analysis*

In the present part, each species was analyzed individually. Following the methods presented in **Chapter 4**, Hg concentrations were measured in body feathers for northern fulmars (belly, n = 124 individuals), common eiders (belly, n = 118) and black-legged kittiwakes (back, n = 120) and head

feathers for the alcids (Atlantic puffins, $n = 42$; Brünnich's guillemots, $n = 239$; common guillemots, $n = 131$; little auks, $n = 64$). The winter period was defined from November to December for northern fulmars and from November to January for Atlantic puffins, Brünnich's and common guillemots, little auks, common eiders and black-legged kittiwakes for tracking analyses. In order to take into account the entire wintering ground that plays a role in seabirds exposition to Hg, we extracted weekly median positions for each individual. Hence, the unique winter Hg concentration measured for each individual was related to several winter positions ($n = 2$ to 15) and therefore increased the number of observations per individual (northern fulmars, $n = 1977$ observations; common eiders, $n = 1917$; black-legged kittiwakes, $n = 1764$; Atlantic puffin, $n = 729$; Brünnich's guillemots, $n = 3842$; common guillemots, $n = 2322$; little auks, $n = 763$). The spatial variation in Hg concentrations was tested with Generalized Additive Models (GAMs). Winter median longitudes and latitudes were added as fixed effects, while individuals were added as a random effect to take into account repeated numbers of winter positions per individual. The results of the GAMs indicated a significant effect of each variable (excepted individuals for the black-legged kittiwakes, **Paper 5, Table 2**). The interpolation was done on the residuals of the GAMs. Then the prediction of both the GAMs and the interpolation were summed up in order to have the most accurate Hg predictions. The predictions and variances of Hg concentrations for each species are mapped on [Fig. 5.1](#).

Our results showed that, despite the fact that species do not all cover the same range of areas, similar patterns of Hg distribution occur. Indeed, for most of the species, we can see an east-west gradient in Hg concentrations increase, in addition to hotspots of Hg. For Atlantic puffins, hotspots of Hg concentrations were predicted in the south of Iceland and around Greenland, while coldspots were predicted from the Northern Iceland to the Northern Norway ([Fig. 5.1A](#)). For Brünnich's guillemots, we found the same pattern, extended to the East coast of Canada ([Fig. 5.1C](#)). For common guillemots, the results also showed an east-west positive gradient, but the highest concentrations started more East than for the Brünnich's guillemots. This species also demonstrated hotspots of Hg in the North of the UK and in the North Atlantic ([Fig. 5.1E](#)). Additionally, the lowest predictions in Hg concentrations were in the Eastern part of the Barents Sea, while for the Brünnich's guillemots, they covered most parts of the Barents Sea and only a part of the North Atlantic. We also found that even if little auks covered less

areas than the Brünnich's guillemots, both species followed the same pattern. The northern fulmars followed the same east-west gradient in Hg concentrations (Fig. 5.1M), in addition to a north to south increase in the western part of the Atlantic. Some of the highest predictions were found in the southern part of the North East Atlantic, but also along the East Coast of Canada (like Brünnich's guillemots, little auks) and in the north of UK (like common guillemots).

The common eiders winter in coastal areas and are faithful to their wintering grounds (see **Chapter 4**). Populations from the low Arctic spend their entire year in the same areas (Bustnes & Erikstad 1993), while populations from the high Arctic winter along Icelandic or Norwegian Coasts (Hanssen et al. 2016). Results showed that this species had some of the lowest Hg predictions (Fig. 5.1J). This is probably due to their low Hg concentrations and their low trophic level position as they feed on bivalves, in addition to the fidelity to their wintering grounds (see **Chapter 4**). Overall, our predictions showed that the lowest Hg concentrations are found in the North of Norway and on the Faroe Islands and the highest for populations wintering in Iceland and in the South of Norway. Black-legged kittiwakes also showed spatial variations in Hg winter contamination but did not exactly follow the east-west gradient previously described (Fig. 5.1K). For this species, we found some of the lowest predictions close to Greenland and some of the highest in the southern part of the Northwest-Atlantic, as found for the northern fulmars. In addition, some of the lowest predicted concentrations were found around the North of the UK and the Norwegian coast. However, it is important to note that few individuals were found close to coastal areas highlighted as hotspots for other species (e.g. Brünnich's guillemots). In addition, the black-legged kittiwake is the only species for which we did not find a significant effect of the variable "individual" and presented high values of variance (Fig. 5.1L). The black-legged kittiwake is a surface feeder species while most of the others are pursuit-divers. Therefore, this could suggest that this species covers a different habitat compared to the other species, in which higher variations occur. This could also suggest that each individual uses larger wintering areas within which accumulated Hg might vary spatially. Additionally, we used one Hg measurement for several wintering locations. Thus, each individual might cover a larger distribution of marine area during winter compared to the other species, therefore influencing their exposition to Hg and create such a high variance.

Black-legged kittiwakes are surface feeders, relying on both fish and invertebrates (Hatch et al. 2020). Brünnich's guillemots, common guillemots and Atlantic puffins are pursuit-diver species, relying mostly on fish, but also on invertebrates (Gaston & Jones 1998), with some regional dietary differences (Moody & Hobson 2007, Karnovsky et al. 2008, Linnebjerg et al. 2013). The little auks are also divers, but they rely on zooplankton (Gaston & Jones 1998), while northern fulmars are omnivorous, and are able to feed at different compartments (i.e. surface feeding, pursuit-diving) (Mallory et al. 2020). Finally, common eiders rely on benthic preys (e.g. crustaceous, bivalves - Goudie et al., 2020). **Even if those species cover relatively different ranges of habitats and preys, the results showed a relatively similar pattern of Hg distribution throughout the North-Atlantic Arctic with an east-west increase in Hg concentrations.**

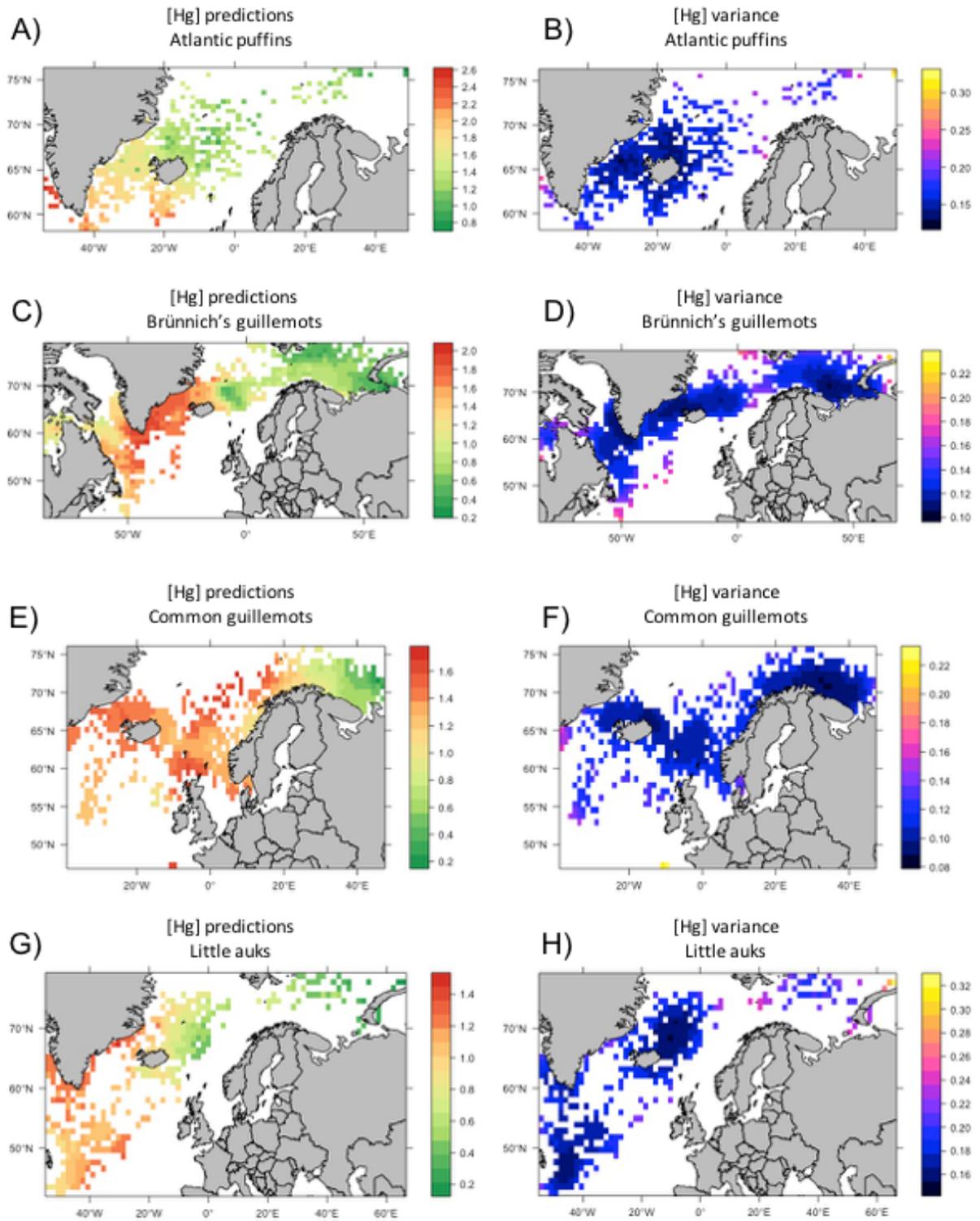


Figure 5.1: Hg prediction and variances for the A), B) Atlantic puffins, the C), D) Brünnich's guillemots, E), F) common guillemots and G) H) little auks.

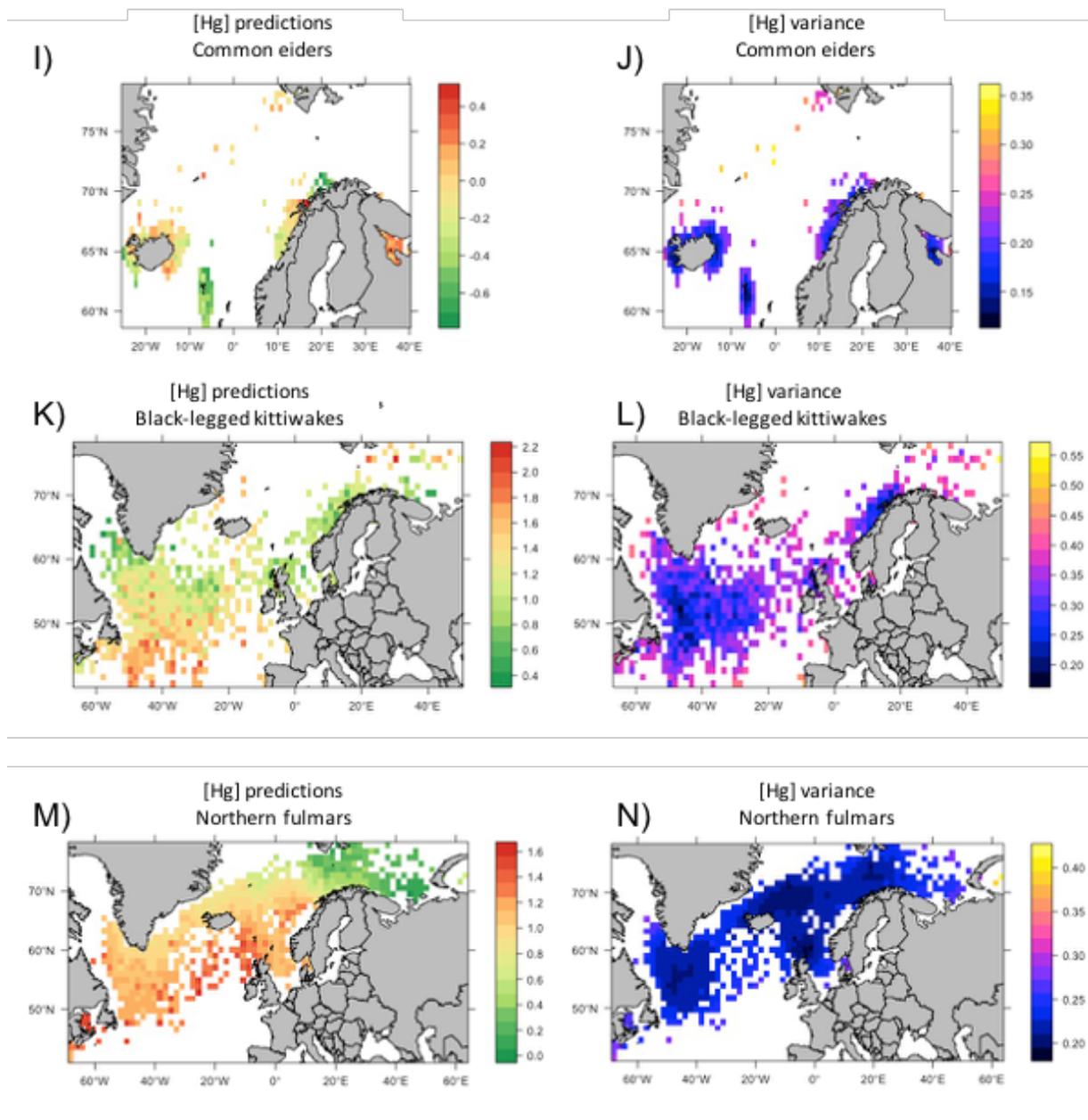


Figure 5.1 (suite): Hg prediction and variances for the I), J) common eiders, the K), L) black-legged kittiwakes, M), N) northern fulmars.

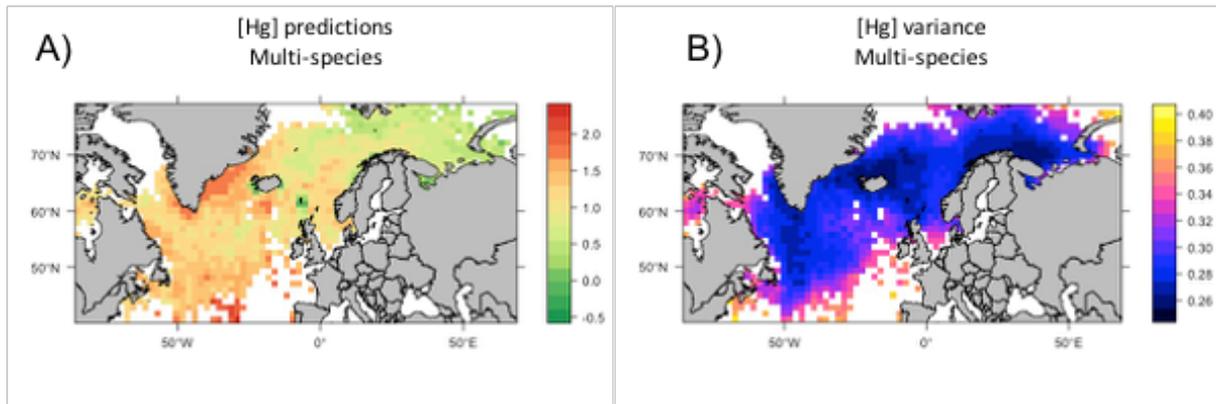
1.2.2. Multi-species analysis

In order to have a broad picture of Hg distribution in biota throughout the North-Atlantic Arctic, we included all the seven species into the same analysis to use them as bio-indicators. In order to take into account their different ecologies (*Annex 1*), we included species as random effects in addition to individuals (see previous section 1.2.1.) in the GAMs analysis. The winter median longitudes and latitudes were both added as fixed effects. The results of the GAMs indicated a significant effect of each

variable (**Paper 5**, *Table 1*). The interpolation of Hg concentrations was done on the residuals of the GAMs. Predictions of both GAMs and interpolation were summed up in order to take into account most of the variability induced by this analysis and therefore have the most accurate results. Because of the high variability in Hg predictions found in the black-legged kittiwakes and a pattern that looks a bit different compared to the other species, we ran this multi-species analysis with or without this species (**Fig. 5.2**). The predictions (**Fig. 5.2A, 5.2C**) and the variance (**Fig. 5.2B, 5.2D**) of Hg concentrations were mapped on **Fig. 5.2**. The results indicated an increase in Hg concentrations following an east-west gradient, with or without the black-legged kittiwakes. Overall, according to our results, including or not this species does not change the general pattern. Therefore, and also because they allow us to cover more areas and a slightly different habitat, we decided to keep them into the multi-species analyses.

Our results support previous findings focused on the breeding season, therefore mostly focusing on the Arctic (AMAP, 2018; Provencher et al., 2014, **Paper 1**). Seabirds mostly feed in the vicinity of their colony during the breeding season. Hence, by focusing on the contamination of breeding seabirds, these previous works informed about the spatial distribution of Hg in marine areas relatively close to the breeding colonies. But Arctic seabirds are migratory species, and most of them migrate outside of the Arctic during the non-breeding period. By using seabirds as bio-indicators, we therefore extend these results to a large part of the North-Atlantic Arctic marine areas, which also confirms that these spatial variations cover the entire year. We thereby provide a detailed map allowing us to highlight hotspots of Hg in marine food webs. **Indeed, and following our predictions, hotspots were found on the East and Southwest coast of Greenland, in addition to the south of Iceland. To a lesser extent, other hotspots were found alongside the East Coast of Canada.** This confirms previous results highlighting hotspots of Hg contamination in the little auks breeding in the East-coast of Greenland and wintering off the East Coast of Canada (Fort et al., 2014). Overall, the results found within the multi-species analysis confirm the trend presented in the intraspecies description.

With black-legged kittiwakes



Without black-legged kittiwakes

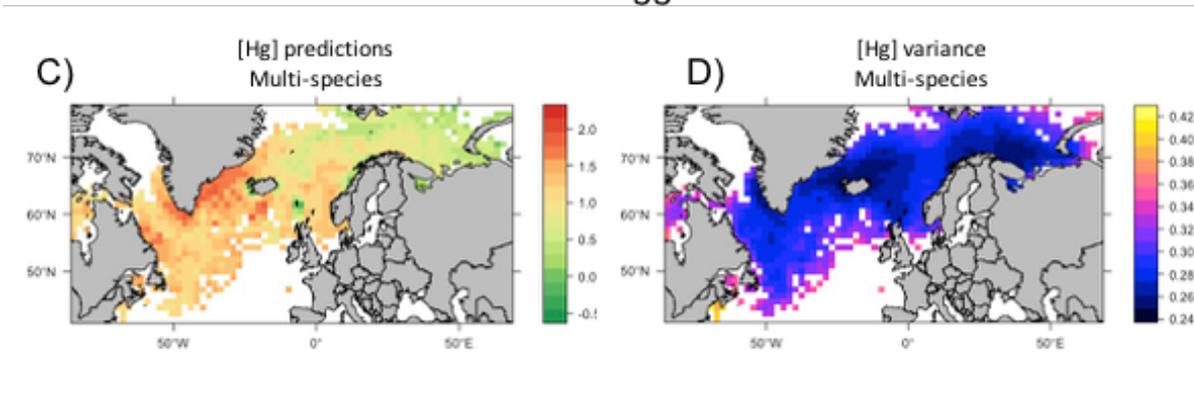


Figure 5.2: spatial representation of the [Hg] predictions and variances A), B) with or C), D), without black-legged kittiwakes.

1.3. Comparison with abiotic monitoring

Hg cycle and transportation is complex and involves many mechanisms. Therefore, the spatial patterns of Hg are not necessarily the same between the environment (e.g. oceanic or atmospheric environment) and biota (e.g. trophic chain). Indeed, Hg found in the trophic chain comes from bacteria methylation and such methylation is driven by environmental factors (e.g. light, temperature). Thus, high Hg concentrations can be measured in the ocean, but in an area with a low rate of methylation, it is most likely that Hg concentrations in the biota will be low. Therefore, there is a need to monitor this pollutant in different compartments (biotic and abiotic) in order to have a holistic vision of its distribution and of the environmental risks associated (Evers et al., 2016). Previous works modeled Hg distribution at a large scale through the study of air deposition and oceanic current. When comparing our results with the atmospheric depositions we see that on the four models presented in the AMAP

(2011), only one seems to follow our trend (Fig. 5.3a), but only alongside the Greenlandic coast (Dastoor et al., 2008, GRAHM, net depositions). But it is important to note that those models were performed in 2005 while our study goes from 2015 to 2018. We also compared our results with the oceanic currents. For instance, Zhang et al (2020) modelled MeHg at different marine layers. We compared our results with their surface models (0-100m – Fig. 5.4) where most of our studied species feed (although some seabirds dive deeper than 100m), but only similar Hg hotspots are found in a few areas only (off East Greenland). Overall, the differences between our work and the oceanic and atmospheric studies could be explained by different period of measurements, or by a lag between Hg deposition and its incorporation into the food chain (see Foster et al. 2019). This could also suggest an actual difference in Hg concentrations between those different compartments, strengthening the idea that all compartments need to be studied at a large scale to have a holistic idea of environmental contamination and properly evaluate the area at risk for wildlife.

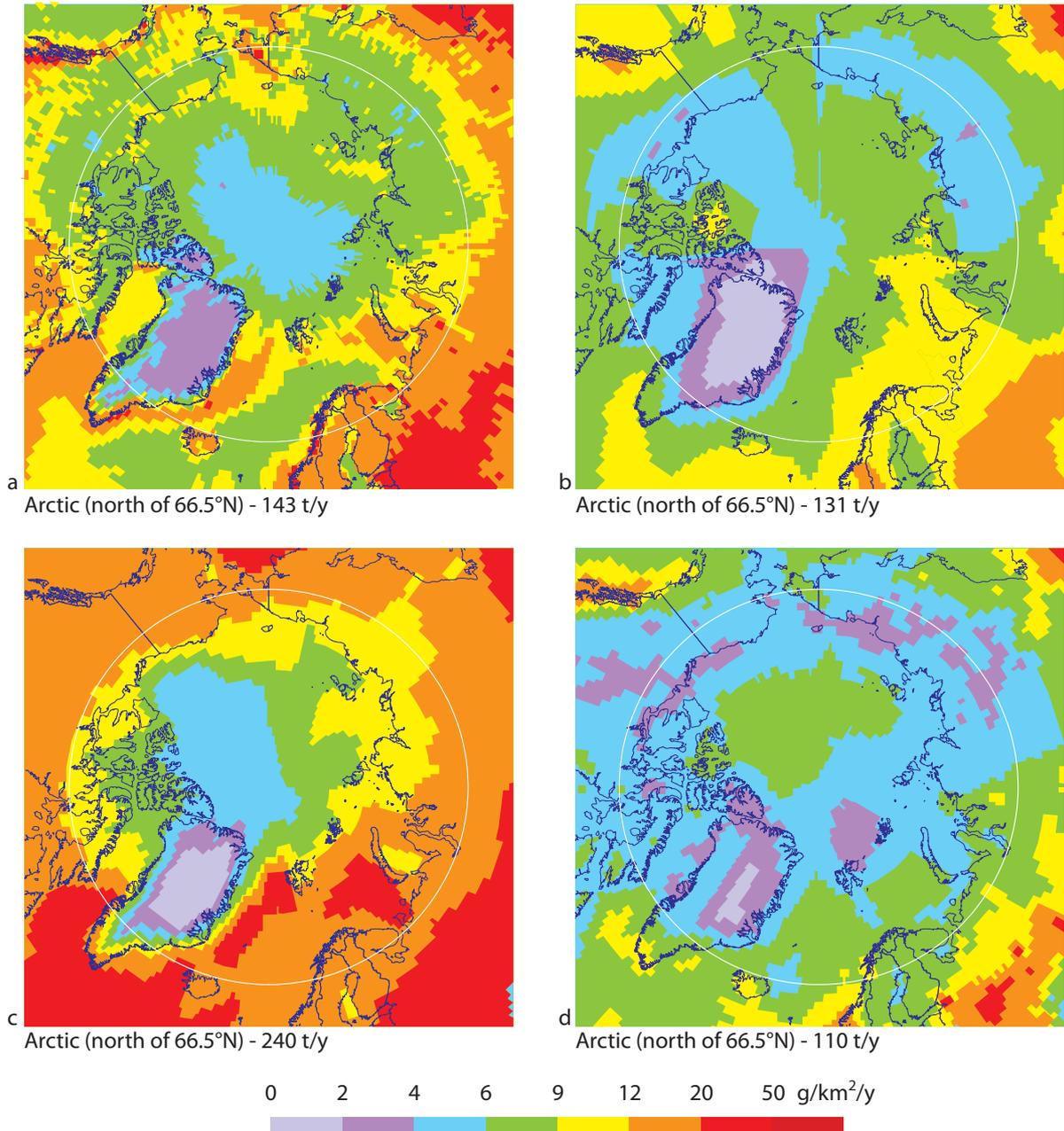


Figure 5.1: Extracted from AMAP, 2011, p. 28. Spatial distribution of total Hg deposition to the Arctic simulated by a) GRAHM (net), b) GLEMOS (gross/net), c) GEOS-Chem (gross) and d) DEHM (gross/net), in 2005.

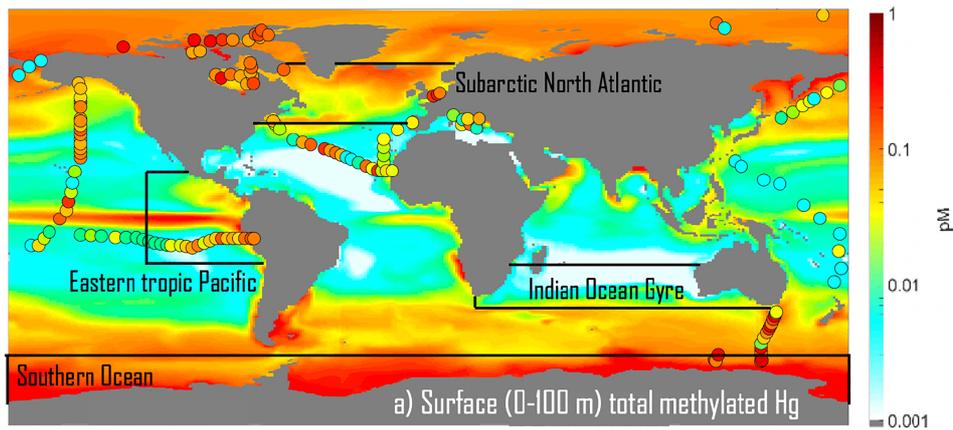


Figure 5.2: Extracted from Zhang et al., 2020

1.4. Conclusion

Seabirds are commonly used as bio-indicators to monitor the health of their environments (Furness & Camphuysen 1997, Burger & Gochfeld 2004). In the present study, we used them to map the distribution of Hg concentrations at a large spatial scale. The use of a multi-species and multi-colony approach was essential to cover a large part of the marine environment throughout the North Atlantic Arctic, but also to cover different habitats and highlight hotspots of Hg. **Following our hypotheses and predictions, our results show that for both intra and interspecies analyses, Hg concentrations spatially vary through the North-Atlantic Arctic with an east-west increase in Hg concentrations; also suggesting the existence of hotspots of Hg concentrations in the East and Southwest coast of Greenland, the West coast of Canada and the South of Iceland.** This result also confirms what was found in previous studies and extend this knowledge to larger marine areas.

2. What is the origin of Hg and its spatial variability in the North-Atlantic Arctic?

2.1. Introduction, hypotheses and predictions

The existence of hotspots of Hg contamination questions about their sources. To study this point, we used an original approach consisting in the combination of isotopic analyses (Hg, C) and tracking of little auks breeding in five different colonies throughout the North-Atlantic Arctic. This section is based on the methods and results of **Paper 6**.

2.1.1 *Hg isotopes: what, why, how?*

Hg has seven stable isotopes (196 to 204) and fractionates dependently and independently of the isotopic masses (Fig. 5.5). The combined use of Hg isotopic mass-dependent (MDF, $\delta^{202}\text{Hg}$) and mass-independent (MIF, $\Delta^{199}\text{Hg}$) fractionation enables the quantification of processes and the identification of sources and pathways of Hg in marine ecosystems (Point et al. 2011). Mass-dependent fractionation (hereafter MDF) of Hg isotopes occurs during many physical, chemical or biological processes (Zheng et al. 2007, Kritee et al. 2008, 2009, Kwon et al. 2013). However, large Hg mass-independent fractionation (hereafter MIF) of odd isotopes (199 and 201) is observed during light-induced reactions (i.e. inorganic Hg photoreduction, MeHg photodemethylation). Hg MIF signature is not affected by biological or trophic processes, so it is conserved up to the food web (Kwon et al. 2012), which represents a significant advantage to trace Hg marine sources. Hg MIF of even Hg isotopes (reported as $\Delta^{200}\text{Hg}$) has been hypothesized to occur during complex atmospheric mechanisms (i.e. photo-oxidation in the tropopause - Chen 2012) and is therefore useful to identify major potential Hg sources of atmospheric origin (Demers et al. 2013, Enrico et al. 2016, Obrist et al. 2017).

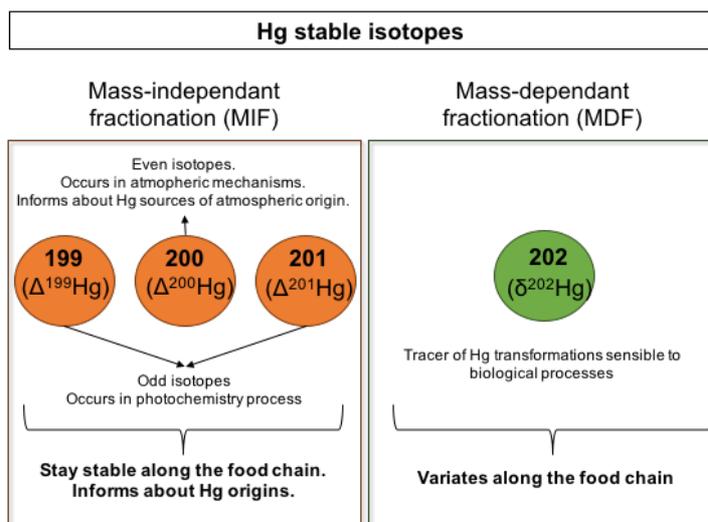


Figure 5.3: representation of four of the seven stable isotopes used to track spatial sources and origins of Hg contamination

Due to the different combinations of the processes involving Hg transformations in the environment, Hg isotopes fractionate differently and with different degrees of magnitude in every environmental compartment. Thus, the analysis of Hg stable isotopes in environmental samples provides information about the different Hg transformations and specific reservoirs

of ecosystems. Hg isotopes of mobile predators such as Arctic seabirds can give access to interesting information about MeHg trophic sources at large scales of the Arctic Ocean and neighboring water bodies. The combination of carbon ($\delta^{13}\text{C}$) stable isotopes with Hg stable isotopes is also a useful tool for the identification of Hg sources and the associated geochemical processes in the different marine compartments (Point et al. 2011, Day et al. 2012, Renedo et al. 2018). Therefore, its use can help understanding Hg exposure pathways and sources in wildlife.

2.1.2. Hypotheses and predictions

Here we propose an original approach consisting in the combination of isotopic analyses (Hg, C) and wildlife tracking to provide new information about MeHg exposure and its spatial origin. For this purpose, we focused on the little auk, species with several ecological advantages for their use as a bio-indicator model. 1) They reflect MeHg accumulation in a short food web that is strongly dependent on sea ice abundance and seawater temperature (Grémillet et al. 2012). 2) They exhibit colony specific wintering areas (Fort et al. 2013a), then reflecting wide-ranging spatial variability of Hg exposure (Fort et al. 2016). In this study, head feathers (see **Chapters 2 & 3**) isotopic signatures (Hg and C) and wintering migration of five distant colonies of little auks (n=10 per colony) from the Arctic Ocean were studied in order to track sources of Hg contamination in the North-Atlantic during the non-breeding

period. **We tested the hypotheses of spatial variations in Hg concentrations coming from different sources, and predicted higher concentrations in the West Atlantic.**

2.2. Tracking the source of Hg contamination: results and discussion

2.2.1. Hg variations

As previously demonstrated in **Chapters 3 and 4**, Hg concentrations increased following an east-west gradient. Indeed, head feather concentrations were higher in populations wintering in western zones (3.48 $\mu\text{g/g dw}$, East Greenland population) and decreased gradually and significantly ($H=20.13$, $p=0.001$) in those wintering in northeast areas (1.74 $\mu\text{g/g dw}$, Franz Joseph Land population).

2.2.2. MDF ($\delta^{202}\text{Hg}$)

Seabirds from Franz Joseph Land, the most northeastern population, exhibited significantly heavier $\delta^{202}\text{Hg}$ values relative to the other four populations during the non-breeding period ($H=29.42$, $p<0.0001$ – Fig. 5.6). Spatial variations of MDF can be used as a proxy of MeHg sources and transformations between different oceanic compartments (e.g. inshore-offshore spatial grounds), including biological processes (e.g. transformation through the trophic chain). Therefore, different $\delta^{202}\text{Hg}$ values in Franz Joseph Land birds suggest potentially distinct Hg sources or dynamics in the wintering grounds of this population relative to the others. However, as MDF is sensible to biological processes, the geographical variations of $\delta^{202}\text{Hg}$ values are generally difficult to distinguish due to confounding metabolic processes. For example, it is known that little auks from Franz Joseph Land are morphologically bigger than populations from Svalbard due to more severe climate conditions in this area (Stempniewicz et al. 1996). Thus, potentially different biological characteristics associated to their bigger size could contribute to higher feather $\delta^{202}\text{Hg}$ values for individuals from this

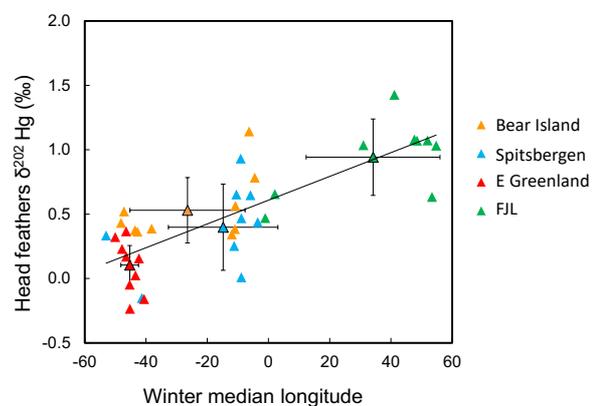


Figure 5.6: Hg MDF ($\delta^{202}\text{Hg}$) of little auk head feathers (winter) as a function of the median longitude of their winter grounds.

colony. Consequently, we cannot discern spatial MeHg sources related to different winter distribution of the different populations only by the interpretation of their $\delta^{202}\text{Hg}$ signatures.

2.2.3. Odd-MIF ($\Delta^{199}\text{Hg}$)

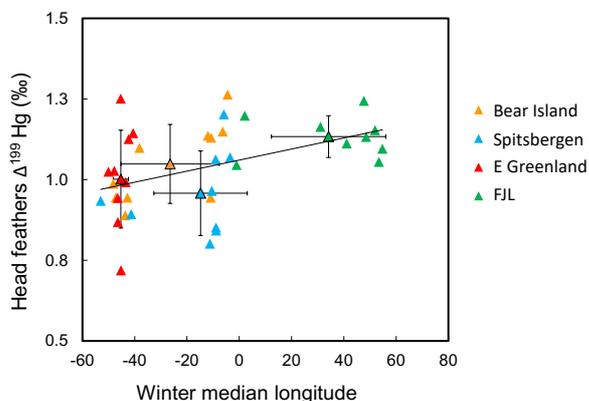


Figure 5.7: Hg odd-MIF ($\Delta^{199}\text{Hg}$) of little auk head feathers (winter) as a function of the median longitude of their wintering grounds.

A positive and significant relationship was observed between head feather $\Delta^{199}\text{Hg}$ values and longitude ($R^2=0.22$, $p<0.0001$, Fig. 5.7), and a positive and weak relationship between head feather $\Delta^{199}\text{Hg}$ values and latitude ($R^2=0.07$, $p=0.06$), although not significant in winter. As for the MDF, Franz Josef Land population showed higher $\Delta^{199}\text{Hg}$

values ($1.13 \pm 0.06\text{‰}$) compared to other studied colonies ($1.00 \pm 0.12\text{‰}$). This suggested higher extent of Hg photochemical processes in wintering areas of Franz Josef Land birds. However, head feathers are assumed to integrate Hg during late autumn and winter seasons when this region is highly covered by sea ice and under scarce or even inexistent light availability. Therefore, we would expect lower $\Delta^{199}\text{Hg}$ values in little auks wintering in these northern zones compared to southern zones where the sea ice extent is lower or absent. Previous studies on Alaskan seabirds revealed that the presence of sea ice impedes light penetration and influences Hg marine photochemistry. Indeed, they found $\Delta^{199}\text{Hg}$ lower values in ice-covered northern regions ($0.53 \pm 0.15\text{‰}$; 68°N) than in low-ice covered southern oceanic areas ($1.13 \pm 0.16\text{‰}$; $56\text{-}58^\circ\text{N}$ - Point et al. 2011). Even if we observed weaker differences on $\Delta^{199}\text{Hg}$ values between FJL (80°N) and other colonies ($70\text{-}77^\circ\text{N}$), it appears that our north to south trend is inversed to the one observed in Alaska. Therefore, this opposed latitudinal trend of $\Delta^{199}\text{Hg}$ values between the Eastern and Western Arctic Ocean regions reveals different Hg dynamic systems in terms of Hg accumulation in food webs, which might be explained by MeHg photomethylation processes. Indeed, previous work found that, compared to open ocean basins, the methylation by bacteria occurs in much shallower depths in the Arctic. Therefore, the sun light is more involved in the methylation and

could enhance the MeHg concentration input in the food chain, even in iced areas (Heimbürger et al., 2015).

2.2.4. Even-MIF ($\Delta^{200}\text{Hg}$)

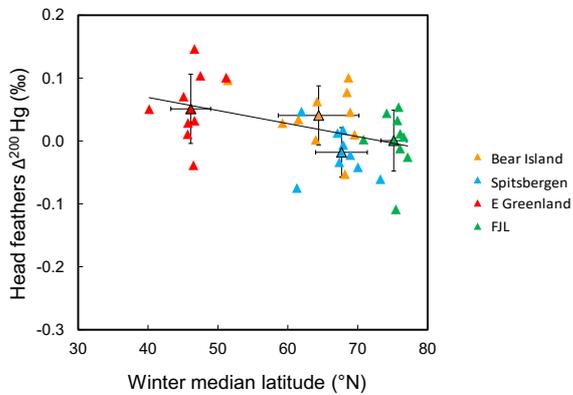


Figure 5.8: Hg even MIF ($\Delta^{200}\text{Hg}$) of little auk head feathers (winter) as a function of the median wintering latitude.

No relationship was observed between head feathers $\Delta^{200}\text{Hg}$ values and winter longitude ($p=0.15$). However, head feathers $\Delta^{200}\text{Hg}$ values were negatively and significantly related to winter latitude ($R^2=-0.13$, $p=0.016$, Fig. 5.8), decreasing from East Greenland ($0.02 \pm 0.06\text{‰}$) to Spitsbergen ($-0.05 \pm 0.04\text{‰}$) individuals, suggesting a

latitudinal trend of Hg atmospheric sources. Atmospheric deposition in sea ice or boreal soil, in addition to riverine inputs in the Arctic are believed to be the major drivers of Hg originating from industrialized countries (Kirk et al. 2012, Soerensen et al. 2016, Sonke et al. 2018). Although we could suppose a relative contribution of Hg released from snow in seabirds wintering in Arctic northern regions, $\Delta^{200}\text{Hg}$ values in feathers were in general more positive than those previously documented in Arctic snow impacted by AMDEs (Atmospheric Mercury DEpletions; -0.12‰ - Sherman et al. 2010a). The low $\Delta^{200}\text{Hg}$ values observed in boreal soils ($-0.02 \pm 0.03\text{‰}$ - Obrist et al. 2017a) and Arctic Ocean's sediments ($0.00 \pm 0.03\text{‰}$ - Gleason et al. 2017) matched the close-to-zero mean $\Delta^{200}\text{Hg}$ values registered in feathers of little auks wintering in northern regions. This could suggest a predominance of sedimentary/riverine inputs over the Arctic and North Atlantic Oceans as suggested by recent observations (Soerensen et al. 2016, Sonke et al. 2018). However, the high degree of intra-population uncertainty did not allow us to make conclusive deductions about the contribution of atmospheric, sea ice melting or continental Hg input across the wintering sites of little auks.

2.2.5. Carbon ($\delta^{13}\text{C}$) and Hg stable isotopes: terrestrial vs marine Hg inputs

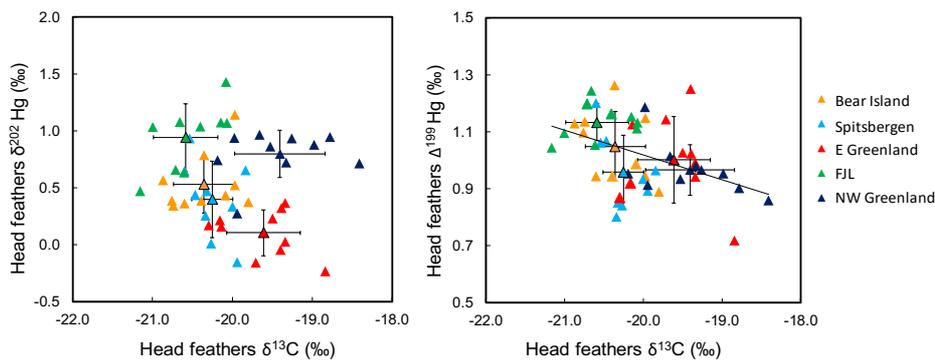


Figure 5.9: Carbon ($\delta^{13}\text{C}$) vs MDF and MIF Hg signatures for winter (head feathers) periods.

Carbon ($\delta^{13}\text{C}$) is commonly used as a proxy to determine foraging habitats (e.g. coastal vs pelagic - Cherel & Hobson 2007). The combination of $\delta^{13}\text{C}$ and Hg stable isotopes allows to go further in such an investigation as they can be used as proxies to discriminate the contribution of terrestrial or marine MeHg sources (Renedo et al. 2018). However, it is important to note that while Hg isotopes represent the winter period, the $\delta^{13}\text{C}$ represents the molt period only (end of the winter period when feathers grow). Head feather $\delta^{13}\text{C}$ values of little auk populations overwintering in western areas of the North Atlantic Ocean were statistically different from those wintering in north-eastern areas ($H=26.28$, $p<0.0001$). No relationship was observed between $\delta^{202}\text{Hg}$ and $\delta^{13}\text{C}$ values of head feathers ($R^2=-0.025$, $p=0.863$) but the linear relationship was significant for $\Delta^{199}\text{Hg}$ and $\delta^{13}\text{C}$ values ($R^2=-0.350$, $p=0.017$ - Fig. 5.9), suggesting only a link between the foraging habitat and the photochemistry process in Hg concentrations.

Head feather $\delta^{13}\text{C}$ values of little auks wintering in north-eastern Arctic regions (Franz Josef Land (-20.59‰) and the Bear Island (-20.35‰), are close to the values of the Arctic sediments (i.e. the Yermak Plateau (-23.0‰), the central Arctic Ocean (-21.4‰) - Schubert & Calvert 2001). **Such results suggest a supply of terrestrial Hg for individuals wintering in north-eastern Arctic regions.** Indeed, the high Arctic areas are impacted by currents going from north to south (i.e. Makarov and Arctic currents), potentially transporting terrigenous organic matter from river discharge and from coastal erosion by sea ice and glaciers (Winkelmann & Knies 2005). This suggestion is strengthened by the recent observations reporting higher concentrations of total dissolved Hg in the Arctic Ocean compared to the North-Atlantic Ocean (Cossa et al. 2017). However, head feather $\delta^{13}\text{C}$ values of little auks from

North West (-19.41 ‰) and East Greenland (-16.61‰) are closer to marine carbon endmembers (i.e. particulate organic matter of meltwater ponds and Arctic ice-cores (-15.3 to -20.6‰) - Schubert & Calvert 2001a). **This suggest a contribution of Hg from deep water masses for individuals wintering in the north-western Atlantic regions.** Indeed, these populations winter in regions near Newfoundland (Fort et al. 2013b), where the North Atlantic Current enhances the supply of marine organic matter (Fontela et al. 2016) and Hg (Lamborg et al. 2014) from deeper layers, known to be enriched in Hg as a result of the incorporation of anthropogenic inputs (Lamborg et al. 2014).

3. Conclusion

Our results show the great potential of using both tracking data and head feathers isotopic signatures to track Hg sources during the non-breeding period. **By linking different methods, and following our hypotheses, we highlighted spatial differences in Hg inputs. Indeed, Hg concentrations in seabirds wintering in the High Arctic could be explained by terrestrial Hg inputs, while for seabirds wintering in the North-Atlantic region Hg concentrations could mostly be explained by Hg marine inputs coming from Hg-enriched deep waters.** Tracking Hg sources is essential to improve the knowledge about the risks encountered by wildlife and to subsequently set up appropriate conservation actions and Hg mitigations programs.

Overall, the use of seabirds as bio-indicators allow us to show spatial variations in Hg concentrations throughout the North-Atlantic Arctic with an increase Hg concentration following an east-west gradient. This confirm and generalize the knowledge found on previous chapters. More precisely, in the present chapter, we highlighted hotspots of Hg on the west part of the North-Atlantic Arctic, that seems to follow the descendant oceanic currents going from the Arctic to the West of the North-Atlantic. Regarding the finding of the second part of this chapter, the results suggest that the spatial differences found in Hg concentrations might potentially result from different sources of contamination. Indeed, the origin of contamination in the East Atlantic, where we find the lowest concentrations, might mostly come from terrestrial or riverine sources. On the other hand, the origin of the contamination in the West Atlantic, where we find the highest concentrations, might mostly come from deep water masses known to be

enriched in Hg as a result of the incorporation of anthropogenic inputs (Lamborg et al. 2014). This suggests that Hg concentrations found in biota might result from Hg inputs in the ocean, with deep water masses being more loaded by anthropogenic Hg on the West Atlantic than the terrestrial and riverine inputs in the East Atlantic.

4. Summary of chapter 5

- The use of seabirds as bio-indicators of Hg distribution show:
 - Spatial variations in Hg distribution through the North-Atlantic Arctic, with a positive east-west Hg gradient
 - Hg hotspots were found alongside the Canadian coast, the south-east Greenlandic coast and the South of Iceland.

- The use of tracking information and Hg isotopes reveals spatial differences in Hg sources throughout the North-Atlantic Arctic:
 - High supply of Hg marine inputs by Hg-enriched deep waters in North-Atlantic regions.
 - High supply of terrestrial Hg in northern east Arctic environments.

- Using seabirds as bio-indicators indicates spatial variations in Hg concentrations through the North-Atlantic Arctic which are probably the result of differences sources of contamination.

Chapter 6

General discussion, limitations and perspectives



1. Highlights of the doctoral work

The starting point of the present doctoral work was to study the exposure of Arctic seabirds to pollutants and the role played by migratory movements and non-breeding distributions. The thesis was based on a multi-species and multi-colony approach, comprising more than 20 species, distributed in 50 different colonies, covering the pan-Arctic area. This large-scale approach seemed essential to take into account different ecologies and environments, but also to extract a large-scale pattern. With this work, we highlighted:

- 1) a seasonality in Hg concentrations with higher exposure during the non-breeding period than during the breeding period, revealing the importance of the non-breeding period in terms of contamination to Hg
- 2) migratory movements and distribution play a role on seabirds Hg contamination at both individual and population scales, which has been linked to spatial variations in Hg distribution.
- 3) spatial variations in Hg distribution throughout the North-Atlantic Arctic, with the existence of hotspots of Hg contamination

2. Seasonality in Hg contaminations

In **Chapter 3**, we found seasonal variations in Hg concentrations, revealing the importance of the winter period. In the Arctic, only one study of a little auk population breeding in the Northeast of Greenland found seasonal variations in Hg concentrations with higher concentrations during the non-breeding period, that they linked with carryover effects on the following breeding period (Fort et al., 2014). Because of these deleterious effects, it was therefore crucial to extend such a knowledge to multiple species at a large spatial scale. In the present doctoral work, we found, for most of the species and colonies, a higher contamination during the non-breeding period. We can therefore extend and confirm the importance of the winter period in terms of contamination to Hg for multiple species, at a large spatial scale. Therefore, such work highlights the importance to take into account the non-breeding period when studying Hg contamination, its effects and risks on wildlife, at a short and long term.

3. Migratory movements of Arctic seabirds and their role in Hg exposure

In **Chapters 4 and 5**, we focused on spatial ecotoxicology during the non-breeding period. More specifically, we studied the link between seabird Hg contamination and migratory movements. The different approaches (i.e. inter-annual variability, inter- and intraspecies variability, seabirds as bio-indicators) allowed us to highlight spatial variations and hotspots of Hg concentrations throughout the North-Atlantic Arctic during the non-breeding period. **More specifically, our results allowed us to suggest that, beyond the migration, it is their distribution and the areas where they decide to winter (or breed) that affect their contamination to Hg.** By extension, the breeding area also plays a role in seabirds Hg contamination (**Chapter 3**), but regarding the main focus of this PhD work and the extent of knowledge regarding this period, I decided to only discuss the non-breeding period and the different sources of Hg that could explain the concentrations in seabirds wintering areas.

In the present doctoral work, we found an east-west increase in Hg concentrations during the non-breeding period, following previous studies focused on the breeding season (AMAP, 2018; Provencher et al., 2014). As seabirds mostly feed close to their colony during the breeding season, only a restricted part of the marine areas was covered. With the present doctoral work, we support this result and extend it to the non-breeding period and the North-Atlantic Arctic marine areas. We highlighted hotspots of Hg concentrations alongside the Canadian Coast in addition to the East coast of Greenland (Labrador Sea, Newfoundland). During the breeding season, the Canadian Arctic and the West coast of Greenland were highlighted as the most contaminated areas (AMAP, 2018), which was mostly attributed to atmospheric depositions coming from Asia (AMAP, 2011, but see Wang et al., 2018). The Arctic Ocean is a sink for contaminants such as Hg, with inflows from both oceanic and atmospheric currents, as well as rivers (Sonke et al. 2018). Nevertheless, important oceanic outflows of Hg also occur from the Arctic to lower latitudes through the Arctic Canadian Archipelagoes and Fram Strait (Outridge et al., 2008). The Hg hotspots found in the present thesis are following the Greenlandic (East and West) and Labrador oceanic currents and could therefore partly explain those high Hg concentrations. On the contrary, the lowest concentrations found in the Barents Sea might be explained by lower atmospheric

depositions and oceanic transportation in comparison to the West Atlantic. Indeed, the Minamata convention invited signatory countries to decrease their Hg emissions. Important decrease has been observed in Europe. Oceanic currents from the South of Europe to the Barents Sea may have seen their load in Hg concentrations decrease which might therefore explain the lower concentrations found in this area. Additionally, atmospheric depositions in this area seem to come from Russia and North America, which emit considerably less Hg than Asia (AMAP, 2011). This might explain the differences between these two regions. In **Chapter 5**, we compared our results to oceanic and atmospheric depositions and showed only a close pattern for the Canadian and Greenlandic areas, which we believe could be attributed to different periods of emission and/or lag of Hg incorporation into the food webs, or an actual difference between biotic and abiotic compartments.

The results of this doctoral work give new inputs about Hg contamination throughout the year in seabirds and wildlife (e.g. seabirds used as bio-indicators). This work can therefore be helpful for an adaptation, reinforcement and /or improvement of protective measures depending on the level of the contaminations. Previous studies based on multi-colony analysis show that the West part of the North-Atlantic was a key area for many seabird species during winter and played a role on population trends (Frederiksen et al. 2012, 2016, Fort et al. 2013b). For instance, while Brünnich's guillemot populations breeding in Canada and mostly wintering off Canada were stable, the populations breeding in Svalbard and mostly wintering off Greenland declined (Frederiksen et al. 2016). To explain such a decrease, direct human impacts like hunting have been proposed as one possible threat (Frederiksen et al. 2012). Our study also reveals the importance of winter by highlighting the existence of hotspots of Hg concentrations and the populations above toxicity thresholds. The fact that these hotspots occur in areas with high seabird densities during winter suggests that this contaminant should also be considered as a possible threat for populations survival and trends during this period. Seabirds studied in the present work winter in both national (e.g. Canada, Greenland, Europe) and international waters. Therefore, and knowing the deleterious impacts of this pollutant (and other pollutants) on wildlife, both national and international conservation measures about contaminants should be extended to the winter period in marine areas highly used by wildlife.

4. Limitations

Blood and feathers are non-invasive and complementary tissues that have shown their effectiveness in large spatial and temporal scale Hg assessments. But they have their limitations.

On remote fieldwork, electricity is not always available and can therefore prevent to freeze blood samples. Blood samples can be preserved in ethanol. However, even if this not an issue for Hg monitoring, this will be the case for other pollutants (e.g. HAP) or analyses (e.g. telomeres, oxidative stress). Regarding feathers, as Hg is stored and fixed in feather keratin (**Chapter 2, Paper 1**), it does not necessarily require to be frozen. As long as feathers are protected from external contamination or degradation (e.g. UV, humidity), they can therefore be more easily stored, no matter the fieldwork conditions (see Jaspers et al., 2019). Blood and feathers are both good proxies to monitor seabird's health and contamination status. However, even if the concentrations indicate the risks for the internal organs, they cannot inform about lesions *per se* that Hg could cause on internal organs.

Using feathers in Hg assessment requires to properly know seabirds molting pattern in order to select the feathers corresponding to the study period we are interested in. This is essential to make proper interpretations. For example, some species have a sequential molt of their body feathers over several months or an unknown molting pattern. The use of these species for feather Hg assessment is thus more complicated. This is for example the case of the Atlantic puffin (Gaston and Jones, 1998). Even if, like most of the alcids, they undergo two molts per year, this species presents unsynchronized post-breeding molts between individuals (Harris & Yule 1977, Gaston & Jones 1998). This is why we decided not to use this species in seasonal studies (**Paper 3**), but only in the non-breeding period Hg study (**Paper 5**). Additionally, it is the knowledge about molting pattern that allowed us to study seasonal variations in Hg concentrations of seabirds in the first place. And even if this study was restricted to the alcids, other seabird groups could allow such studies (e.g. gulls) at even more specific temporal scales. Indeed, by acknowledging the detailed molting pattern of a species (including primary feathers), it might be possible to narrow down the study of Hg contamination to more specific time periods (see Quinn et al. 2016).

Large scale movements such as seasonal migration can be associated to changes in diet, possibly affecting Hg intakes and therefore Hg concentrations measured in feathers or blood. But adjusting Hg

concentrations by the diet can allow large-scale assessments and comparisons. As presented in **Chapter 3**, this can be successfully realized when using blood. This is not the case for feathers. Indeed, Hg concentrations in feathers represent an intermolt period, while the nitrogen stable isotopes only represent the period of the feather growth. Therefore, as they do not provide information for the same periods, isotopic values cannot be directly compared to Hg measurements in feathers for species known to switch their diets between (or within) seasons (Bond 2010).

In the present doctoral work, feathers were used to study the non-breeding period, which was defined from November to January for most of the species. This choice was done based on the knowledge of molting patterns and the migration of the species. But also, because we needed to have homogenized periods for all species. Indeed, in the case of this large-scale study, we incorporated species breeding at different latitudes. However, it is for example known that species start breeding earlier in the Low Arctic than in the High Arctic, which subsequently influences migration and molting periods. In addition, we are aware that Hg concentrations into feathers represent an intermolt period and therefore include Hg contamination during migrations. But as the migration is a small part of the non-breeding period (see Fort et al., 2013, Frederiksen et al., 2011) and because during this period, seabirds will feed for the longest period on their wintering ground, we postulate that its influence on the Hg concentrations was minimum.

In conclusion of **Chapter 2**, we suggested that the diet alone could not explain the seasonality in Hg concentrations. However, the diet is the main source of Hg contamination, so it appears important to further discuss seabirds' prey in relation to migration and seasonality as a limitation. Seabirds rely on a wide range of prey species, like zooplankton or fish (larval to older stages). Zooplankton do not migrate over large areas as top predators (only diel vertical migration - Zaret and Suffern, 1976). Thus, they are representative of the contamination at their local water column. By extension, seabirds relying on zooplankton at their breeding or non-breeding grounds will also represent local contamination. Other prey like fish can migrate during their life cycle. Herrings *Clupea harengus* are important preys for top predators (see *Annex I*). The youngest stages are consumed by the chicks during the breeding season while the oldest stages are consumed by adults throughout the year. The youngest stages usually stay close to their spawning areas (see Trenkel et al., 2014). Therefore, Hg concentrations measured in chicks

will represent relatively local contamination. However, older stages are able to migrate up to 1,500 km between their spawning and wintering areas (see Trenkel et al., 2014 for a review) and might therefore represent Hg contamination from different regions. Capelins *Mallotus villosus* are also an important resource for seabirds throughout the year (see *Annex 1*). This species releases eggs in spawning areas that can drift because of currents and might therefore face different Hg exposures. However, older stages, representing an important prey for adults, stay in relatively close areas throughout the year (see Magnusson et al., 2005). Overall, seabirds and top predators are essential bio-indicator of the health of their environment. But it seems important to keep in mind that even if their prey migrate over smaller distances and areas than they do (see Magnusson et al., 2005), their migration should be taken into account while monitoring Hg contamination and other pollutants related to diet in piscivorous seabirds. That is why we believe that modelling analyses including migration and contamination of top predators' prey (from breeding and non-breeding periods) might be additional important knowledge to take into account when studying Hg contamination sources.

5. What about other pollutants?

In the present doctoral work, we focused on large-scale mercury contaminations, but many other pollutants affect wildlife health. This is for example the case of the POPs which induce deleterious effects on seabird's reproduction (Bustnes et al. 2003, Tartu et al. 2014). Large-scale assessment of those pollutants in Arctic seabirds is possible when using blood but will therefore be restricted to the breeding period. Previous studies used feathers to study POPs in birds (Jasper et al., 2006) and suggested that this tissue could be a good proxy to study POPs at a large spatial scale (see Michielsen et al. 2018 for a review). However, even if we know that POPs are excreted into the feather during the molt, the knowledge regarding the period of incorporation (i.e. intermolt as Hg, feather growth as nitrogen isotopes?) is unclear (e.g. remobilization from internal organs) and needs more investigation. Finally, few information on assessment of other pollutants with the use of feather is available and is not recommended without further studies (Jaspers et al. 2019). Therefore, the methods used in the present

doctoral work could only be extended to few other pollutants including some POPs and heavy metals at least for the breeding period (Jaspers et al. 2019).

6. What's next?

Seabirds have shown their usefulness for large scale Hg studies during both breeding and non-breeding periods. However, and based on the comparison made with biotic modelling (**Chapter 5**), it appears important to implement research including both biotic and abiotic measurements at a large scale. Such research will be essential to have more inputs on potential lags between Hg deposition and incorporation into the food webs (see Foster et al., 2019).

Research on long-term effects of Hg contamination and the associated risks at a large scale are also essential. Such studies are reviewed within the Arctic Monitoring and Assessment Program (AMAP) including multiple species, coming from different research programs and dataset. Therefore, they normalize the data to be able to compare them between each other (see Ackerman et al., 2016 for example). Large dataset analyzed within the same conditions like in the present doctoral work will be useful to reduce uncertainty due to transformations. In regard to the present doctoral work and the ARCTOX network, long-term and risks research at a large scale might also be conducted specifically for seabirds. We briefly described risks that seabirds might have to face in **Chapter 3**. But when we talk about risks, we should also talk about selenium. Indeed, this element is known to be an antagonist of Hg effects (Khan & Wang 2009). Thus, seabirds presenting high Hg concentrations, but also high selenium concentrations might actually be able to cope with the deleterious effects that Hg is responsible for (Khan & Wang, 2009). Therefore, additional large-scale selenium assessment might be extremely useful to have better insight regarding Hg contamination and its associated risks. It is also possible to implement large scale study on long-term effects of Hg contamination and the associated risks during the non-breeding period. This could potentially reinforce and extend previous studies highlighting the carryover effects of Hg from the winter period to the following breeding period (Fort et al., 2014). Previous studies have highlighted the risks on seabirds associated with long-term Hg contamination (Goutte et al., 2014). However, long-term effects of winter Hg contamination (and other pollutants) and

its effects on population dynamics are still scarce (but see Amélineau et al., 2019 and Leat et al., 2013) and should be improved at a more global scale. All of those points will be essential to inform about the efficiency of international programs of Hg reduction such as the Minamata convention. But this will also be essential to inform about the conservation of the most impacted species throughout the Arctic.

7. ARCTOX, what's next?

The impressive dataset used in the present doctoral work was made possible because of the numerous collaborators involved in the ARCTOX network. Beyond the sample collections, international collaborations are essential in polar research. First of all, we gain the expertise of many collaborators about different ecological domains, specialized on different geographical areas or species. Such broad expertise is crucial to extract essential information at such large-scale. Second of all, polar regions are interconnected with the rest of the planet. Therefore, all industrialized countries are responsible for the changes (e.g. pollutants, temperatures) in such remote areas. Hence, we should all be involved in international polar research. Arctic countries should not be the only ones to have to deal with it (financially and humanly speaking). Finally, the present doctoral work shows that such collaboration efficiently works and should therefore continue, be reinforced and supported in the long-term.

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ANNEX

Annex 1

species descriptions

In the present annex, I summarized ecological information about the species studied in the present thesis. Details on the different families will be introduced, but only the species studied in this present work will be detailed.

Alcids

Pacific distribution:

- Ancient murrelet (*Synthliboramphus antiquus*)
- Crested auklet (*Aethia cristatella*)
- Least auklet (*Aethia pusilla*)
- Parakeet auklet (*Aethia psittacula*)
- Rhinoceros auklet (*Cerorhinca monocerata*)
- Tufted puffin (*Fratercula cirrhata*)

Atlantic distribution:

- Atlantic puffin (*Fratercula arctica*)
- Little auk (*Alle alle*)
- Razorbill (*Alca torda*)

Pacific and Atlantic distribution:

- Brünnich's guillemot (*Uria lomvia*)
- Common guillemot (*Uria aalge*)

Anatids

- Common eider (*Somateria mollissima*)

Larids

Pacific distribution:

- Glaucous-winged gull (*Larus glaucescens*)
- Red-legged kittiwake (*Rissa brevirostris*)

Pacific and Atlantic distribution

- Black-legged kittiwake (*Rissa tridactyla*)
- Common gull (*Larus canus*)
- Glaucous gull (*Larus hyperboreus*)
- Herring gull (*Larus argentatus*)
- Ivory gull (*Pagophila eburnea*)
- Lesser black-backed gull (*Larus fuscus*)

Phalacrocoracid

- European shag (*Phalacrocorax aristotelis*)

Procelarids

- Northern fulmar (*Fulmarus glacialis*)

Stercorariid

- Great skua (*Stercorarius skua*)

Alcids (commonly called auks)

This family encounters 23 species in the Arctic, with a circumpolar distribution. Globally, they are migratory species laying one egg per year, presenting no sexual dimorphism. Some species are specialized on Pacific (e.g. ancient murrelet, crested auklet, least auklet), the Atlantic (e.g. razorbill, little auk) or both regions (e.g. common and Brünnich's guillemot). Species in this family undergo two molts per year: a pre-breeding molt during which only head feathers (head, cheeks, throat) are molted to switch to the breeding plumage; a post breeding molt during which all feathers (body, head, wings) are molted to switch to wintering plumage. This section is based on "the Auks" (Gaston and Jones, 1998). If otherwise, the reference is added to the text.

Pacific distribution:

The **ancient murrelet** (*Synthliboramphus antiquus*) is about 24-27 cm length (Harrison, 1983) and ~250 g (spatially different). No sexual dimorphism, but females tends to have longer wings.

Distribution: Colonies are found in the Aleutian Islands, the Sea of Okhotsk and Gulf of Alaska. During winter some birds are resident throughout the year while other migrate south. They are mostly found on continental shelf, slope or continental waters.

Diet: During the breeding season, they mostly rely on euphausiids, with some proportions of small fish, depending on their area (juvenile sandlance, shiner perch (*Cymatogaster*) and rockfishes, capelin, walleye pollock, rainbow smelt (*Osmerus mordax*) and sculpins (*Triglops spp*). It has been suggested that they dive in ~10-20 meters. During the non-breeding period, it has been suggested that they only feed on euphausiids.

Table 1: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the ancient murrelet, used in the present thesis.

	Blood	Feathers	GLS
Colonies	0	3	0
Individuals	0	34	0



Figure 1: Ancient murrelets colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **crested auklet** (*Aethia cristatella*) is about 18-27 cm length and 211-322 g, with females a little bit smaller than males.

Distribution: They are found in Bering and Okhotsk seas and in some areas of the North Pacific. Colonies have been referenced in Asia, Russia and Alaska breeding both in remote islands and coasts. During the non-breeding time, populations mostly migrate south of winter sea-ice.

Diet: During the breeding season, this species mostly relies on zooplankton (euphausiids, musids, hyperiids, gammarids, calanoid copepods), but can add larval fish and squid to its diet (Hunt et al., 1993). However, there are no information about their winter diet.

Table 2: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the crested auklets, used in the present thesis.

	Blood	Feathers	GLS
Colonies	1	3	0
Individuals	52	68	0

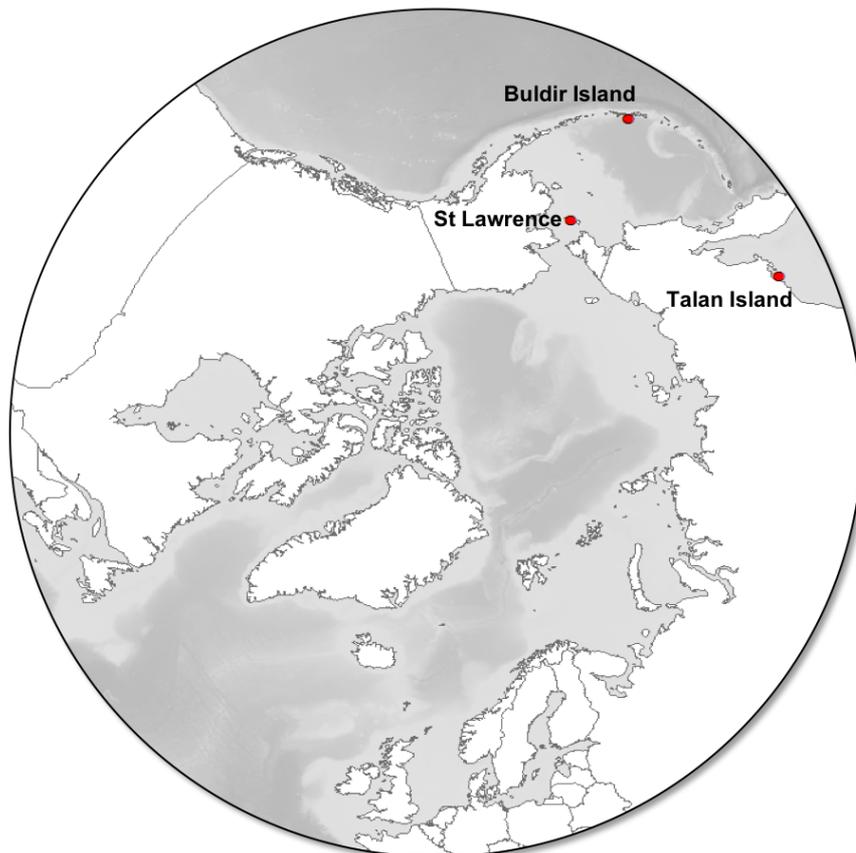


Figure 2: Crested auklets colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **horned puffin** (*Fratercula corniculata*) is about 38 cm length and 612 g.

Distribution: Colonies have been referenced in Asia, Russia, Alaska and Aleutian Islands. During the non-breeding period, there are found offshore, throughout the North Pacific.

Diet: During the breeding season, this species relies on fish and zooplankton while during the non-breeding period, they feed predominantly on myctophids and squids.

Table 3: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the horned puffins, used in the present thesis.

	Blood	Feathers	GLS
Colonies	0	2	0
Individuals	0	5	0

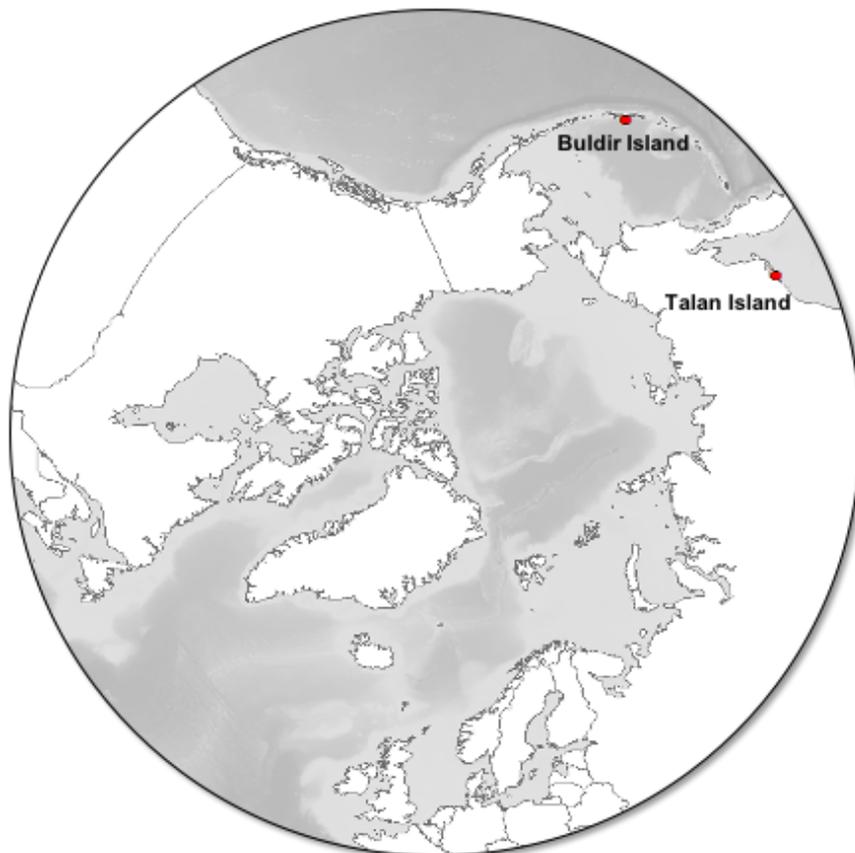


Figure 3: Horned puffins colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **least auklet** (*Aethia pusilla*) is the tiniest auk species being about 14 cm length and 70-98 g. This species is found in the same areas as the crested auklets during breeding season.

Distribution: During the non-breeding period, populations from the Bering Sea migrate south, in areas free of ice (*A. Will, personal comment*).

Diet: This species is mostly zooplanktonic during the breeding season (Calanoid copepods - mostly *N. plumchrus*). No information is available about their non-breeding diet. It has been suggested that they do not dive more than 25 meters (Hunt et al., 1993).

Table 4: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the least auklet, used in the present thesis.

	Blood	Feathers	GLS
Colonies	1	3	0
Individuals	27	99	0



Figure 4: Least auklets colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **parakeet auklet** (*Aethia psittacula*) is about 23-26cm length and 238-347 g.

Distribution: Colonies are found in the the Aleutian Islands, Bering Sea and the Sea of Okhotsk. During the non-breeding period, they are found in the Chukchi Sea and the Central part of the North Pacific.

Diet: The few information available about their diet suggest that they rely on zooplanktons during the breeding period. However, no information is available during the non-breeding period.

Table 5: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the parakeet auklet, used in the present thesis.

	Blood	Feathers	GLS
Colonies	1	0	0
Individuals	5	0	0



Figure 5 : Parakeet auklets colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **rhinoceros auklet** (*Cerorhinca monocerata*) is about 35-38cm length and 441-569 g for male and 389-616 g for female.

Distribution: While the least and crested auklets are specialized in Low and High Arctic, the rhinoceros auklet is mostly specialized in Pacific, boreal and sub-Arctic areas. Colonies are found in the south of the Aleutian Islands, Asia, Japan and Central California. During the non-breeding period, they disperse after the breeding season, before migrating south. The western populations stay close to their breeding colonies while eastern colonies migrate south to winter off California.

Diet: The few information available about their diet suggest that they are piscivorous during both breeding (sandlance, capelin and herring) and non-breeding periods (capelin and sandlance). They are pursuit divers in mid-waters and are able to dive up to 30 meters (Burger et al., 1993).

Table 6: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the rhinoceros auklet, used in the present thesis.

	Blood	Feathers	GLS
Colonies	1	3	0
Individuals	20	44	0



Figure 6: Rhinoceros auklets colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **tufted puffin** (*Fratercula cirrhata*) is a large alcid, measuring about 40 cm length and 775 g.

Distribution: During the breeding season, they are widely found in the North Pacific Ocean, including the Sea of Okhotsk, the Bering Sea, the North Pacific and the Chukchi Sea. During winter, they migrate south in the North Pacific and winter offshore.

Diet: During the breeding season, they mostly feed on invertebrates (e.g. euphausiids) and small fish (e.g. sandlance), while they mostly feed on squid and euphausiids during the non-breeding period. It has been suggested that they are able to dive up to 60 meters.

Table 7: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the tufted puffin, used in the present thesis.

	Blood	Feathers	GLS
Colonies	0	3	0
Individuals	0	18	0

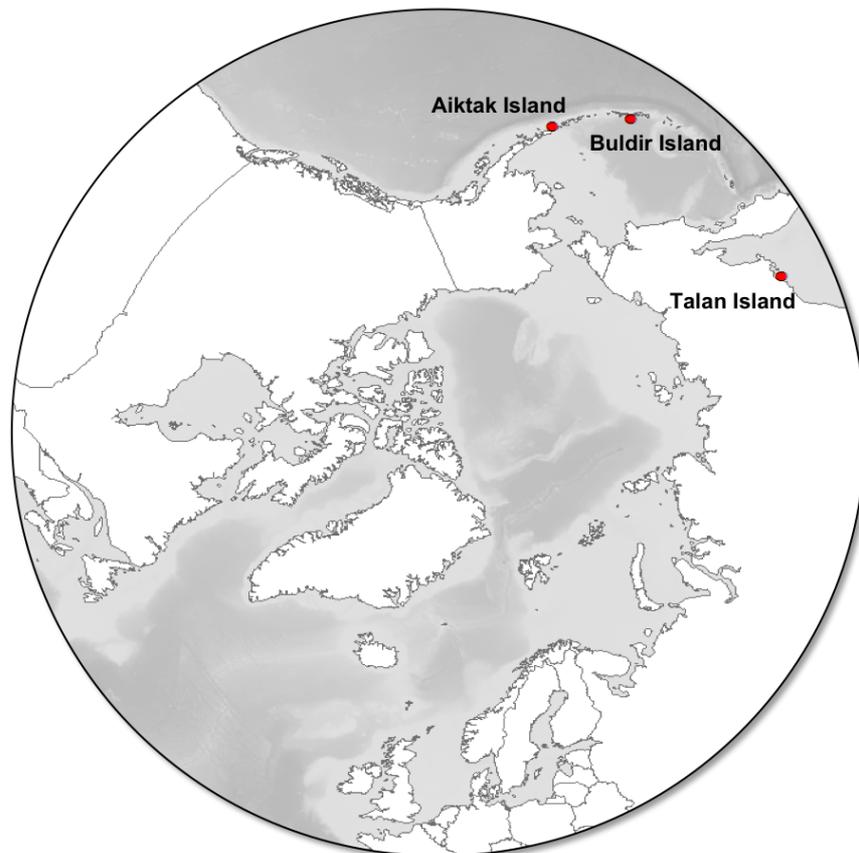


Figure 7: Tufted puffins colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis

Atlantic distribution:



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The **Atlantic puffin** (*Fratercula arctica*), is about 28-34 cm length and 387-710 g, with males slightly larger than females. The weight varies between colonies, with individuals from the High Arctic (subspecies *F. a. naumanni*) being bigger than individuals breeding in the Low Arctic (*F. a. arctica* & *grabae*) (Gaston and Provencher, 2012).

Distribution: breeding populations of Atlantic puffins are found in Svalbard, Jan Mayen, Novaya Zemlya (Russia), Norway, France, Faroes, UK, Greenland, Canada & Iceland. During the non-breeding seasons, they mostly winter south, in open sea.

Diet: Atlantic puffins rely on squids, zooplanktons (euphausiids, mysids, copepods) or small fish (4-14 cm; e.g. *Ammodytes sp.*, *Mallotus villosus*, juvenile cods) during the breeding season. During the non-breeding period, they mostly rely on zooplanktons and small fish (Falk et al., 1992).

Table 8: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the Atlantic puffin, used in the present thesis.

	Blood	Feathers	GLS
Colonies	9	13	1
individuals	374	323	55



Figure 8: Atlantic puffins colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **little auk** (*Alle alle*) (or dovekie), one of the most numerous seabirds in the Arctic, is about 17-20 cm length and 130-200 g.

Distribution: As the Brünnich's guillemot, this species is specialized of the High Arctic. They breed in rocky areas. This species is only found in the Atlantic region with breeding sites in Canada, Svalbard, Russia, Jan Mayen, Greenland. During the non-breeding period, they migrate over large distances in the open sea.

Diet: This species mostly feeds on zooplanktons (Copepods (*Calanus* spp.), ref) during the breeding period, with a shift to a higher trophic level preys (but still zooplankton) during the non-breeding period (J Fort et al., 2010a). This species can dive up to 40 meters (Harding et al., 2009).

Table 9: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the little auk, used in the present thesis.

	Blood	Feathers	GLS
Colonies	7	6	4
Individuals	366	273	79



Figure 9: Little auks colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **razorbill** (*Alca torda*), the least abundant auk, is about 40-45 cm length and 505-890 g (Lavers et al., 2009).

Distribution: razorbills breed in low Arctic, boreal and temperate coastal regions, into crevices in cliff areas. Colonies have been referenced in Norway, England, France, Iceland, Greenland and East Canada. During the non-breeding period, they migrate into open sea, but not as offshore as common guillemots. Populations from European Arctic are suspected to winter in south Europe and populations from East Canada suspected to winter off Newfoundland.

Diet: This species is mostly piscivorous during the breeding period (cod, herring, capelin) (Piatt and Nettleship, 1985), but can add crustaceans or polychaetes to his diet during the non-breeding period (krill, - Huettmann et al., 2005; Moody and Hobson, 2007). It usually dives around 25 meters but can dive up to 120 meters (Piatt and Nettleship, 1985).

Table 10: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the razorbill, used in the present thesis.

	Blood	Feathers	GLS
Colonies	3	2	0
Individuals	30	52	0



Figure 10: Razorbills colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis

Pacific and Atlantic distribution:

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The **Brünnich's guillemot** (*Uria lomvia*) (or thick-billed murre) is about 39-43 cm length and weighting from 700 to 1200 g (K. M. Kovacs and Lydersen, 2006).

Distribution: It preferentially reproduces in the High Arctic, on cliffs ledges. This circumpolar species breeds in both Atlantic (Svalbard, Greenland, the Canadian Arctic, Iceland, Jan Mayen and Russia) and Pacific regions (Alaska, the Bering Sea). During the non-breeding period, they migrate over large distances in the open sea.

Diet: This species is mostly piscivorous, (polar cod, capelin, herring; Gaston and Jones, 1998) but also feeds on crustaceans during the breeding season. During the non-breeding period, they prefer low trophic level prey (mostly invertebrates and fish in the Atlantic and mostly invertebrates in the Pacific, Gaston and Jones, 1998), although spatial differences might occur (Karnovsky et al., 2008; Linnebjerg et al., 2013; Moody and Hobson, 2007). This suggest an adaptation ability to changing environment. It is known to usually dive down to 50-100 meters, with records at 200 meters (K. M. Kovacs and Lydersen, 2006).

Table 11: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the Brünnich's guillemot, used in the present thesis.

	Blood	Feathers	GLS
Colonies	18	17	12
Individuals	601	669	325



Figure 11: Brünnich's guillemots colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **common guillemot** (*Uria aalge*) (or common murre) is about 38–43 cm length and weighting from 900 to 1300 g (K. M. Kovacs and Lydersen, 2006). This species comprises two morphs: the bridled and the non-bridled one. The bridled morphs consist as a white circle around the eye and continuing as a straight line being the eye. As the frequency of this morph increases with latitude, it has been suggested as a cold adaptation (Birkhead, 1984).

Distribution: It mostly reproduces in low Arctic and sub-Arctic (even found in lower latitude – North Europe), on cliffs. The shape of their eggs is adapted to this breeding area not to fall down. Numerous colonies are found in both Atlantic and Pacific regions. As Brünnich’s guillemots, they migrate over large distances in the open sea during the non-breeding period, but at a smaller extent.

Diet: common guillemots mostly rely on fish throughout the year (Gaston and Jones, 1998) and are able to dive up to 150 meters to forage (Hedd et al., 2009).

Table 12: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the common guillemot, used in the present thesis.

	Blood	Feathers	GLS
Colonies	11	12	6
Individuals	496	441	177



Figure 12: Common guillemots colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis

Anatids (commonly called ducks)

Anatids are found in every continent, excepted Antarctica and encompass 39 species in the Arctic (CAFF, 1996).



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The **common eider** (*Somateria mollissima*) is about 71 cm length and 1200 to 2800 g. This species has a sexual dimorphism with male molting into a white and black plumages during the breeding period. Female are greyish all around the year. They breed in relatively flat areas, lay four to six eggs into a nest mostly made of down feathers.

Distribution: populations are found on both Pacific and Atlantic regions, breeding or in Low Arctic or in High Arctic. Populations breeding in High Arctic migrate south to overwinter (Hanssen et al., 2016) while populations breeding in Low Arctic are relatively residents throughout the year (Bustnes and Erikstad, 1993).

Diet: common eiders mostly feed on benthic preys like crustaceans (e.g. crabs) and bivalves (e.g. mussels or clams) (Goudie et al., 2020).

Table 13: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the common eider, used in the present thesis.

	Blood	Feathers	GLS
Colonies	11	10	6
Individuals	373	446	131



Figure 13: Common eiders colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis

Larids (commonly called gulls).

This family encounter 102 species comprising gulls, terns, kittiwakes and skimmers. In the present thesis, we studied eight species of gulls (n=6) and kittiwakes (n=2). This family has a circumpolar distribution, with species specialized in the Pacific (e.g. red-legged kittiwakes, glaucous-winged gulls), the Atlantic (e.g. lesser-black-backed gull, ivory gull) or both regions (e.g. black-legged kittiwake, herring gull, glaucous gull, common gull).

Species in this family (excepted the ivory gulls) undergo two molts per year: a pre-breeding molt to switch to the breeding plumage; a post breeding molt to switch to the wintering plumage. Because of the interspecific variations, further details about their respective molts are provided for each species. This section is based on the website “The Birds of North America, birdsna.org”. If otherwise, the reference is added to the text.



Black-legged kittiwake (*Rissa tridactyla*) is a small gull about 40 cm and weighting from 365 to 400 g. They have two molts per year. A partial molt (some body feathers, and a few median and lesser covert) at the end of the wintering period on their non-breeding grounds or during the migration. A total molt occurs at the end or during the breeding period, occurring or during the migration or at their wintering grounds. They lay one to three eggs per year.

Distribution: Black-legged kittiwakes have circumpolar distribution, found from subarctic to Arctic areas and from the Pacific to the Atlantic Oceans. During non-breeding periods, they migrate south. On the Pacific region, they are found wintering between 41° and 55° latitude. On the Atlantic region, they are found in West and East Atlantic, Greenland Sea, but also even more south for European populations.

Diet: Black-legged kittiwakes are surface feeders, mainly piscivorous (e.g. sandlance, capelin or herring), but might also rely on invertebrates (e.g. euphausiids, amphipods squids, polychaetes). Little is known about their non-breeding diet, but it has been suggested they rely on the same prey throughout the year.

Table 14: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the black-legged kittiwake, used in the present thesis.

	Blood	Feathers	GLS
Colonies	19	28	17
Individuals	459	989	563



Figure 14 : Black-legged kittiwakes colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **common gull** (*Larus canus*) is about 41-46 cm length and weight around 360-415 g, with male a little bit heavier and longer than females. They have a complete molt after the breeding period and a partial molt (head and body) at the end of the non-breeding period.

Distribution: this species is found in both Pacific and Atlantic Oceans. In the Atlantic, they are found across North and south Europe and in Iceland. Individuals are also found in Svalbard and on the East coast of North America. On the Pacific Ocean, they are largely distributed on both American and Asian coasts. During winter, they migrate south of their breeding range.

Diet: The common gulls are omnivorous, feeding on large variety of fish, insects, eggs of other species, invertebrates or garbage. Quantities vary depending on their distribution. It appears that they adapt their diet on the food availability depending on areas and seasons.

Table 15: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the common gull, used in the present thesis.

	Blood	Feathers	GLS
Colonies	1	0	0
Individuals	21	0	0



Figure 15: Common gulls colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **red-legged kittiwake** (*Rissa brevirostris*) is a small gull (smaller than the black-legged kittiwake) measuring about 37 cm and weighting around 377 g. This species has a complete molt after the breeding period and a partial molt (nape and head feathers) at the end of the non-breeding period (Fleishman et al., 2019a). This species looks alike the black-legged kittiwake, with the main physical difference that their legs have different colors (as indicated in their common names).

Distribution: When black-legged kittiwakes are found in both Pacific and Atlantic Ocean, red-legged kittiwakes are only found in four colonies in the Bering Sea (Pacific Ocean): the Pribilof and Aleutian Islands.

Diet: The red-legged kittiwakes are surface feeder. During the breeding period, they mostly feeding on small fish (<10 cm, e.g. Northern lampfish (*Stenobranchius leucopsarus*), walleye pollock (*Theragra chalcogramma*)) and marine invertebrates (e.g. squids and amphipods). No information is available about their winter diet even though a recent study, based on nitrogen stable isotopes suggested that this species have a constant diet throughout the year (Fleishman et al., 2019).

Table 12: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the red-legged kittiwake, used in the present thesis.

	Blood	Feathers	GLS
Colonies	1	1	0
Individuals	4	61	0



Figure 16: Red-legged kittiwakes colonies sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



Glaucous gull (*Larus hyperboreus*) is a large pale gull measuring from 42 to 49 cm length and weighting from 1250 to 2700 g. Glaucous gulls have two molts per year. A partial molt occurs during the non-breeding period (some body and head feathers, upperparts and breast feathers), near or on breeding grounds. A complete molt occurs at the end of the breeding period, usually starting at the breeding area and completing at the wintering ground.

Distribution: glaucous gulls have a circumpolar distribution with colonies on both Atlantic and Pacific Oceans. During winter, they migrate south and mostly winter in coastal areas.

Diet: glaucous gulls are generalist predators throughout the year, feeding on fish, invertebrates (crustacea, mollusks), eggs, chicks of other birds, small birds and mammals, and vegetation. They can also be scavenger. Prey quantities might vary depending on areas and seasons. They also feed on human garbage.

Table 13: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the glaucous gull, used in the present thesis.

	Blood	Feathers	GLS
Colonies	6	6	4
Individuals	164	138	52



Figure 3: Glaucous gull colonies (red dot) sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



Glaucous-winged gull (*Larus glaucescens*) is a large gull measuring about 60 cm for male and 57 cm for female and weighting around 1224 g for males and 957 g for females. They have two molts per year, including a partial molt at the end of the non-breeding period (only wings and tail retained) and a complete molt at the end of the breeding season. Their molting pattern is not well studied compared to other gulls.

Distribution: Glaucous-winged gull are specialized on the Pacific Ocean and found from the north-central Bering sea to south of West American coast. During winter, they are found or along the coast or pelagically, from the sea-ice front zone in the Bering sea to southern regions, even South America.

Diet: Glaucous-winged gulls are surface feeders. This omnivorous species feeds on large varieties of fish, marine invertebrates and carrions and even human garbage.

Table 14: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the glaucous-winged gull, used in the present thesis.

	Blood	Feathers	GLS
Colonies	0	1	0
Individuals	0	12	0

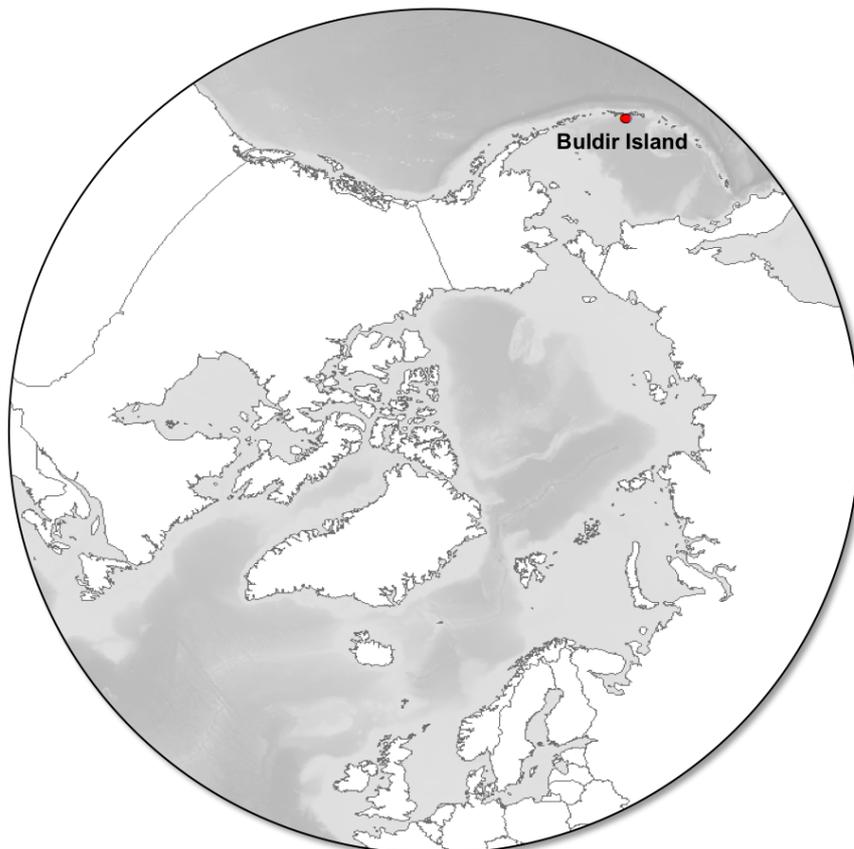


Figure 18: Glaucous winged gull colonies (red dot) sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **herring gull** (*Larus argentatus*) is a widespread species, largely found in northern hemisphere, about 56–66 cm length for a weight from 800 to 1250 g. They have two molts per year: a partial molt at the end of the non-breeding period and or near the wintering ground (some to most body feathers) and a complete molt at the end of the breeding period.

Distribution: During breeding periods, they are mostly found in northern areas. They migrate and winter south during non-breeding periods.

Diet: During the breeding period, herring gulls are generalists' predators, in addition to be scavengers. They rely on a large diet varying from fish, mollusks, crustaceans, insects, mammals, berries, eggs, young and even adults of other seabird species. This diet might vary over areas and seasons.

Table 19: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the herring gull, used in the present thesis.

	Blood	Feathers	GLS
Colonies	1	5	4
Individuals	13	98	22



Figure 19: Herring gull colonies (red dot) sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **ivory gull** (*Pagophila eburnea*) measures about 40 to 43 cm long and weight from 448 to 687 g. The ivory gull is one of the only gull in the present thesis to only have one molt per year. They undergo a complete molt from march to September. Breeding individuals usually suspend their feather molt during the chick rearing.

Distribution: This species is specialized in the high Arctic during the breeding period. They migrate south during winter but stay relatively close to the ice edge.

Diet: Ivory gulls are opportunistic feeders throughout the year, relying on fish and invertebrates, but also on carcasses and feces. They have a high trophic level close to murrens and fulmars' diet.

Table 20: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the ivory gull, used in the present thesis.

	Blood	Feathers	GLS
Colonies	1	1	0
Individuals	23	26	0



Figure 20: Ivory gull colonies (red dot) sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis



The **lesser-black backed gull** (*Larus fuscus*) measures about 51 to 61 cm long and weight from 550 to 1200 g.

Distribution: This species is found from the High Arctic to south Europe. During winter, individuals are found wintering in the North Africa or all over Europe (East and West).

Diet: They are opportunistic throughout the year and feed on a wide range of preys including small fish, aquatic invertebrates, birds 'eggs and chicks in addition to rodents or berries.

Table 21: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the lesser-black backed gull, used in the present thesis.

	Blood	Feathers	GLS
Colonies	2	4	0
Individuals	21	70	0



Figure 21: Lesser Black-backed gull colonies (red dot) sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis

Procellarids

Procellarid comprises more than 90 species widely distributed (Arctic to Antarctica, Pacific to Atlantic) with many different ecologies. In the present doctoral work, we only focused on the northern fulmars.

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The **northern fulmar** (*fulmarus glacialis*) is a medium-sized petrel measuring about 45-50 cm and weighting from 450 to 1000 g. This species only has one molt per year during which most of feathers (body, wings and tail) are replaced, starting during the breeding period and finishing during the winter (January) on their wintering ground, before heading back to their breeding colonies.

Distribution: Northern fulmar is a petrel specialized in North Hemisphere, from south to Arctic areas, in both Atlantic and Pacific regions. They migrate south of their breeding colonies during the non-breeding period.

Diet: This species is omnivorous, feeding on fish, cephalopods, zooplankton, offal and carrion during the breeding period. No information is available about their non-breeding period diet.

Table 22: Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the northern fulmars, used in the present thesis.

	Blood	Feathers	GLS
Colonies	13	9	6
Individuals	357	356	169



Figure 22: Northern fulmar colonies (red dot) sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis

Phalacrocoracid (commonly called cormorants or shags).

The Phalacrocoracid comprises 42 species, widely distributed. In the present thesis, we only focused on the European shag. This section is based on the website “The Birds of North America, birdsna.org” and more specifically on Orta et al. (2020).

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The **European shag** (*Phalacrocorax aristotelis*) measures about 65 to 80 cm and weighting around 1760-2154 g for males and 1407-1788 g for females. This species comprises three sub-species. In the present case, we focus on the European shag specialized in the Atlantic (*Phalacrocorax aristotelis aristotelis*).

Distribution: This species is mostly found from south to North Europe in addition to Iceland. During winter, excepted the northern populations who migrate south during winter, most of the populations do not disperse far from their breeding grounds. They are mostly costal species.

Diet: European shags dive (from 21 to 40 meters) to find their preys and rely on a wide range of fish (i.e. Gadidae, Gobiidae, Pleuronectidae, Cottidae, Pholidae, Clupeidae, Labridae, Callionamidae).

Table 23 Number of samples (blood, feathers) and geotracking devices (GLS), per colonies and individuals for the European shags, used in the present thesis.

	Blood	Feathers	GLS
Colonies	4	1	0
Individuals	77	13	0



Figure 23: European shag colonies (red dot) sampled from 2015 to 2018 for the ARCTOX network and used in the present thesis

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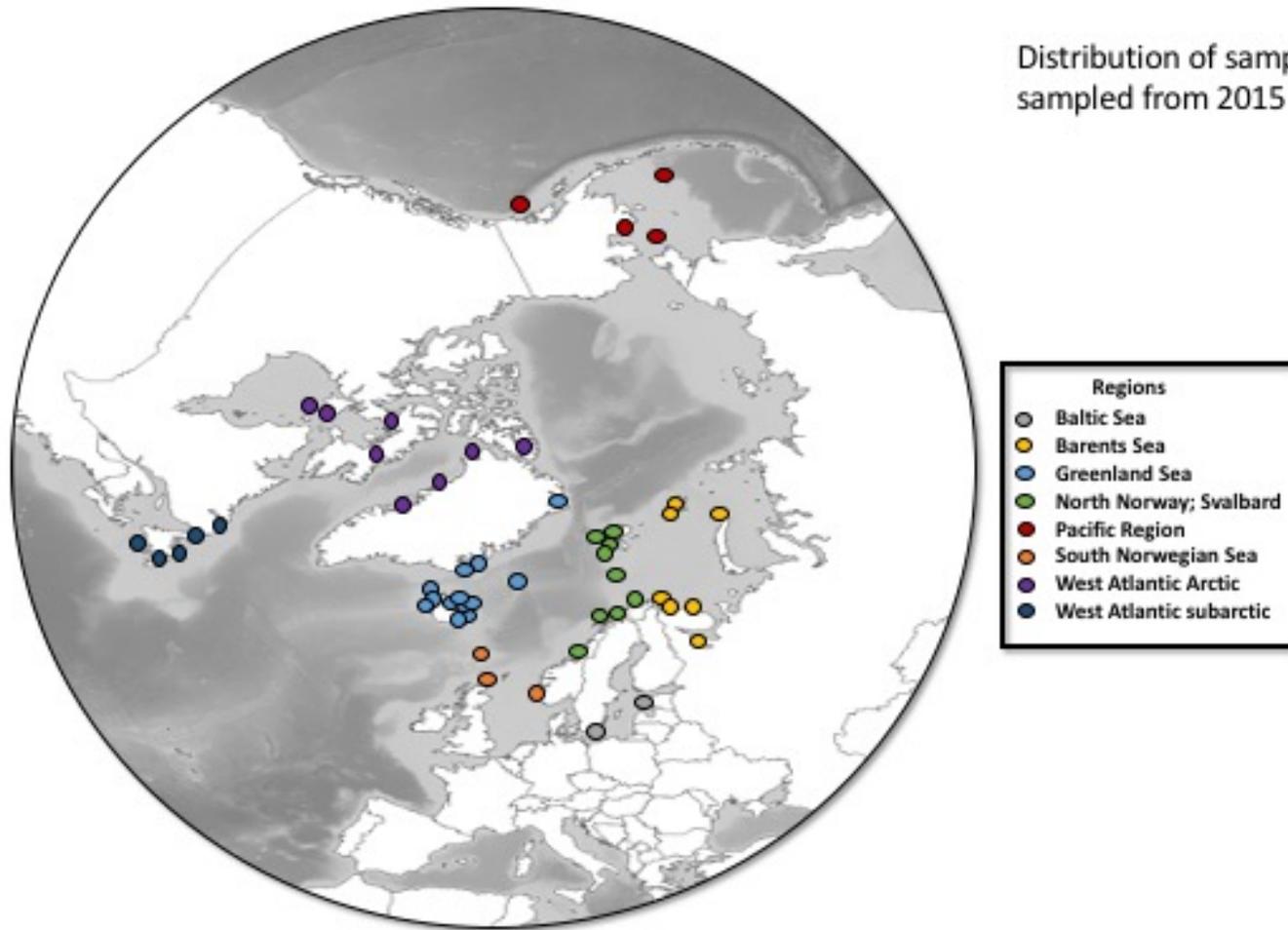
Fort et al., 2010 Geographic and seasonal variability in the isotopic niche of little auks. *Marine Ecology Progress Series*. 414: 293-302. doi: 10.3354/meps08721

BirdLife International 2018. *Alca torda*. *The IUCN Red List of Threatened Species* 2018: e.T22694852A131932615. Downloaded on 11 September 2019.

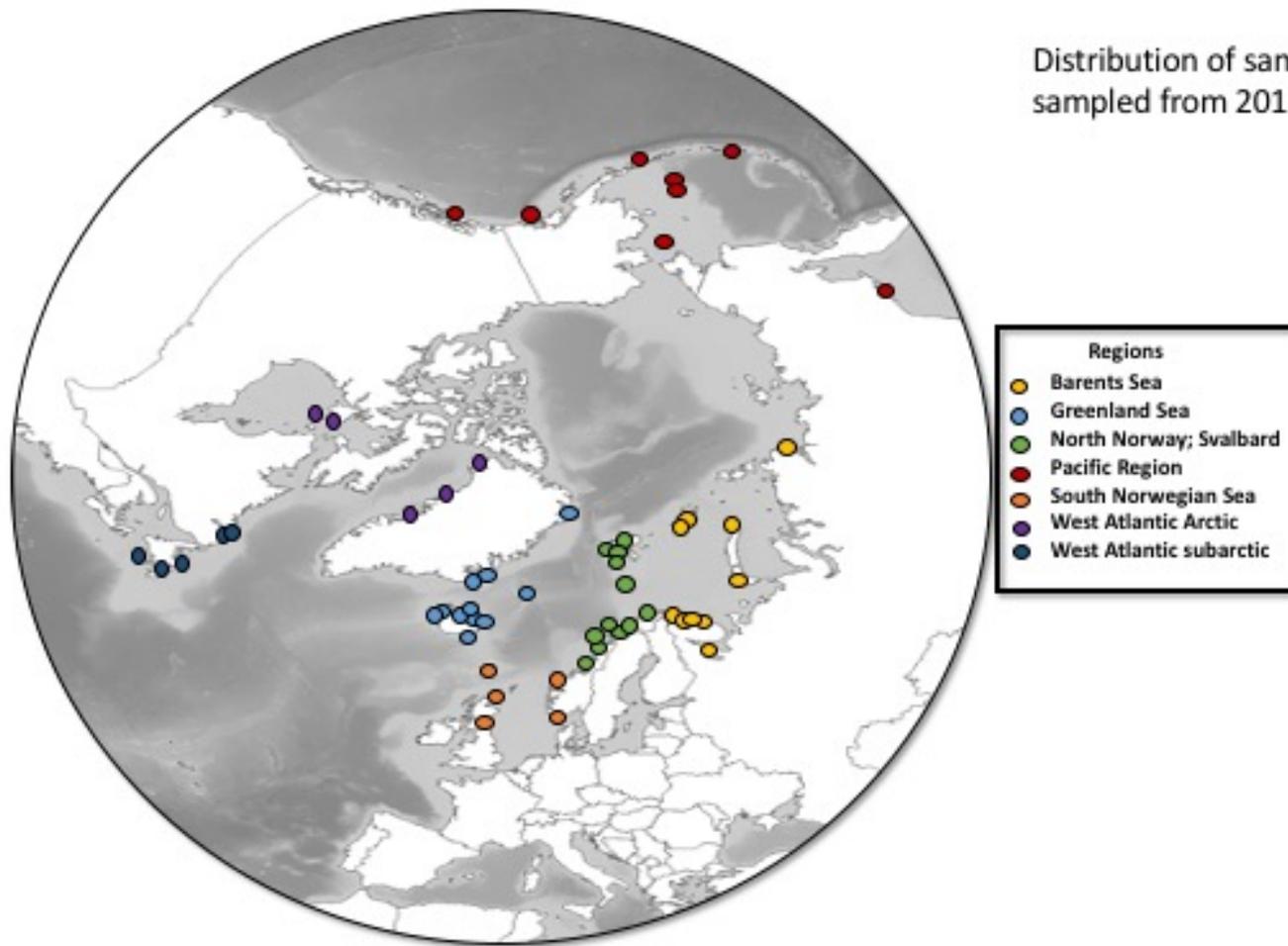
Piatt, J.F.; Nettleship, D.N. (1985). "Diving depths of four alcids". *The Auk*. **102** (2): 293–297. doi:[10.2307/4086771](https://doi.org/10.2307/4086771).

Annex 2

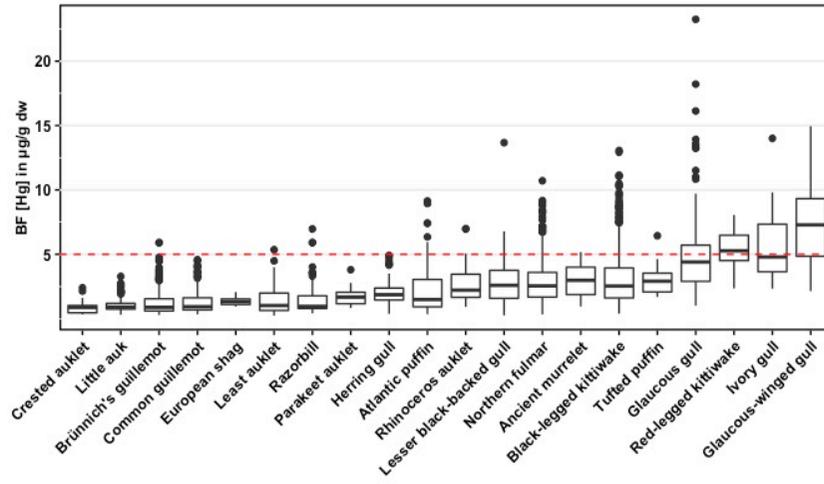
Distribution of sampling sites where **blood** has been sampled from 2015 to 2018



Distribution of sampling sites where **feathers** has been sampled from 2015 to 2018

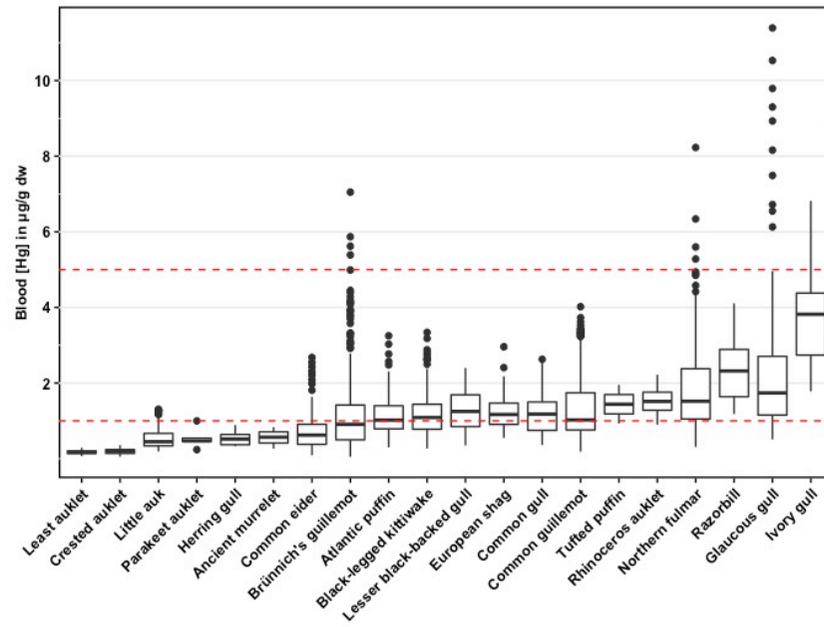


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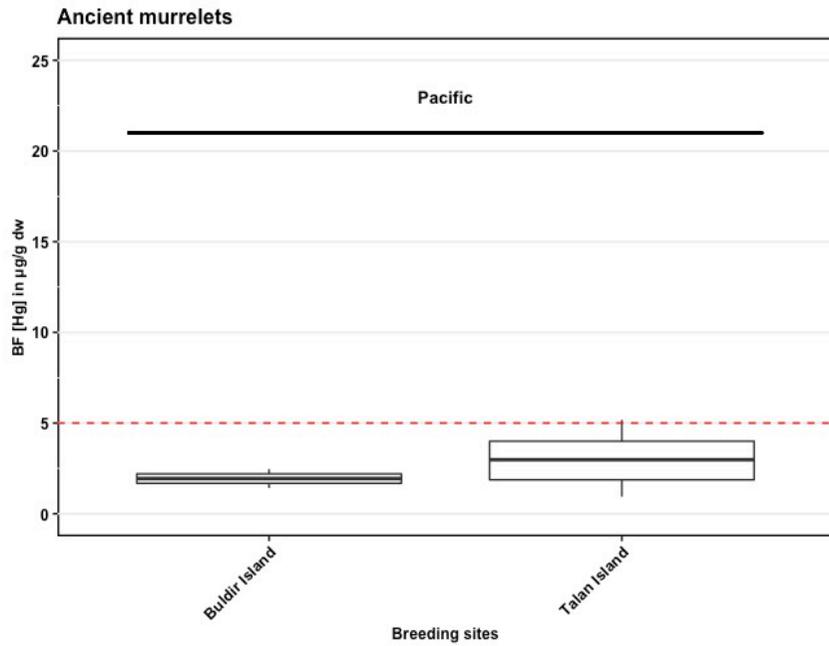


All species

Raw blood



Body feathers

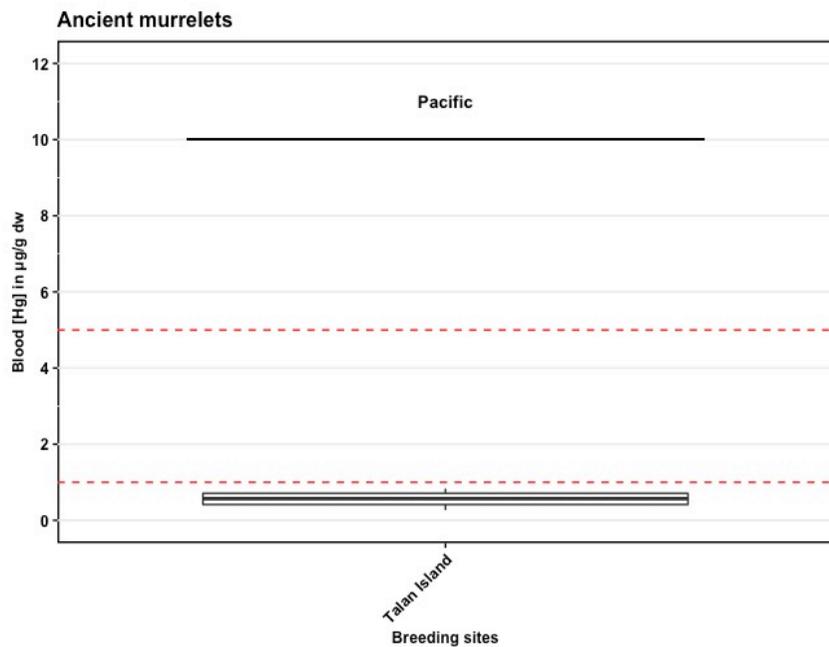


Ancient murrelets

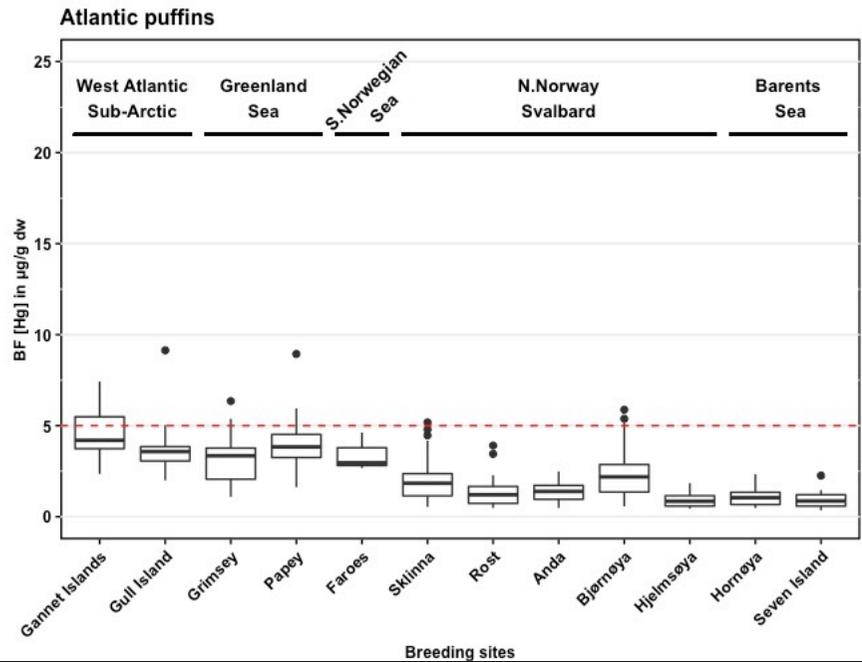


© A. Andreev

Raw blood



Body feathers

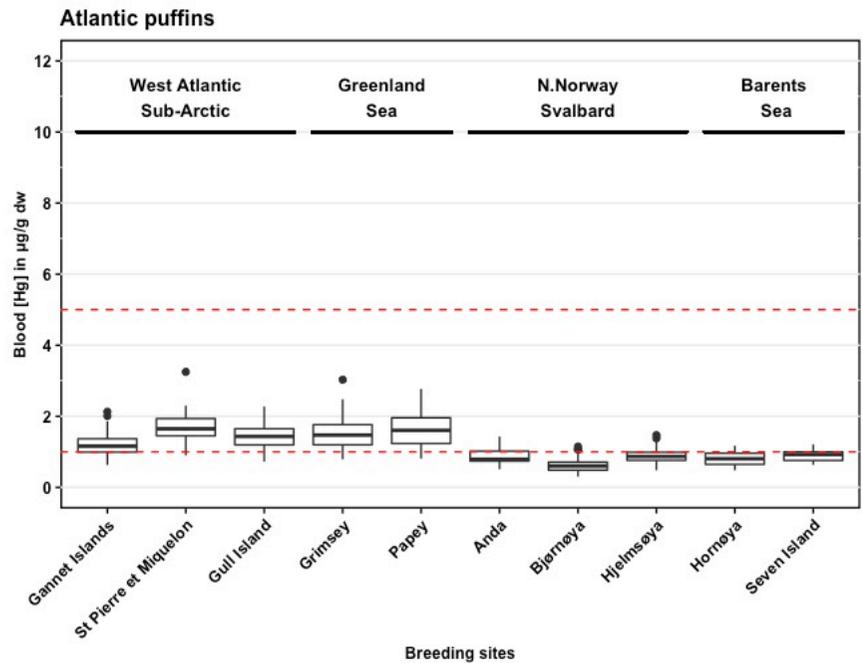


Atlantic puffins

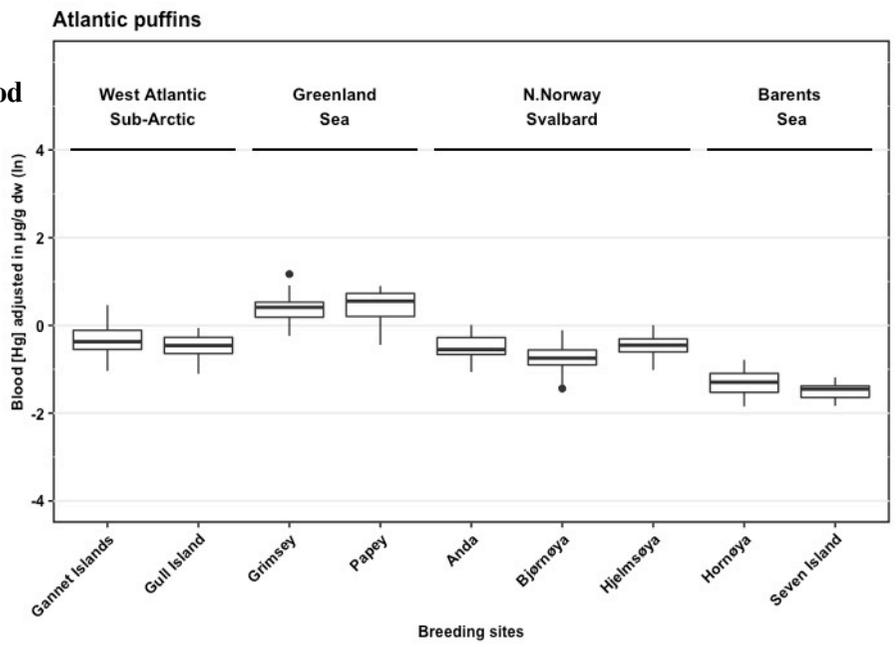


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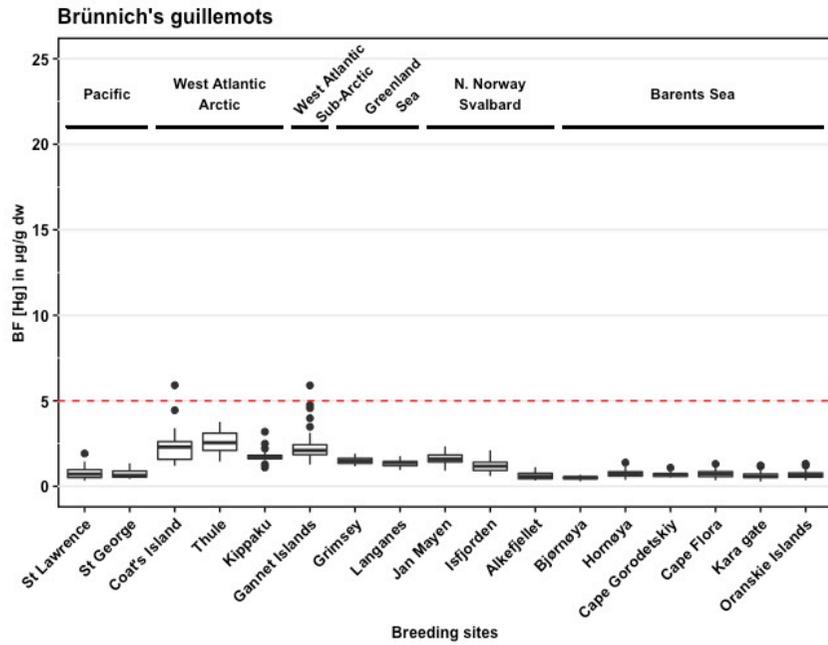
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Adjusted blood



Body feathers

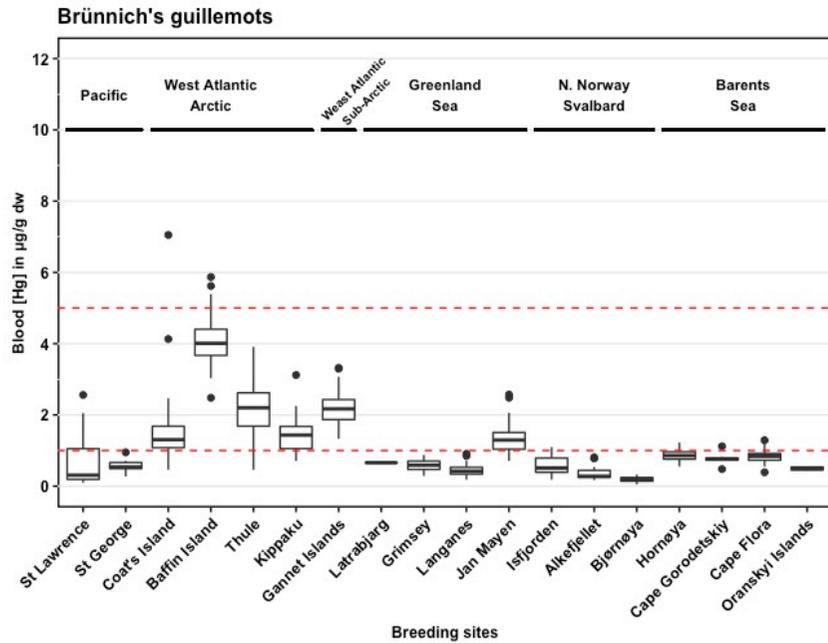


Brünnich's guillemots

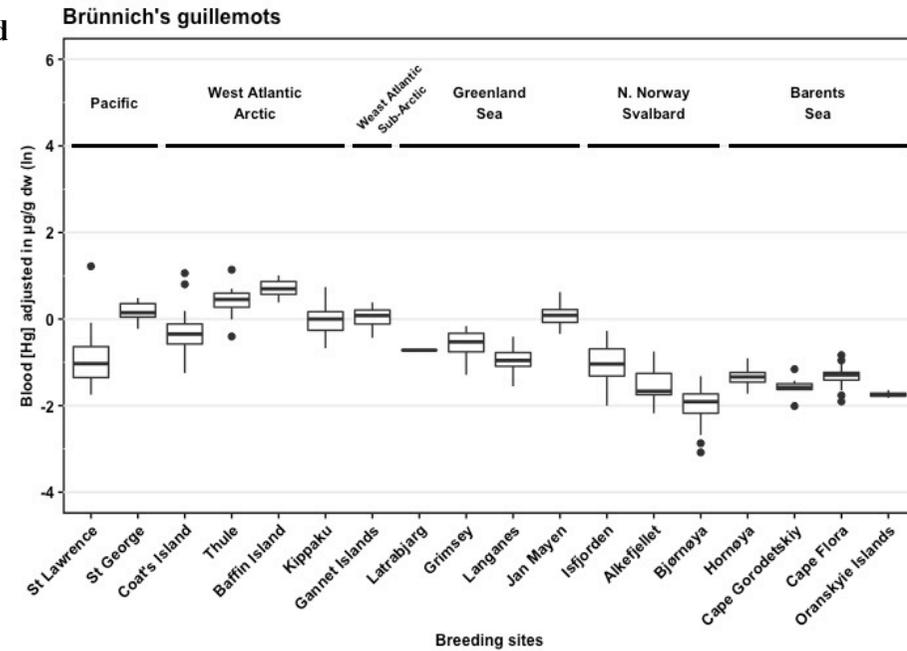
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Raw blood

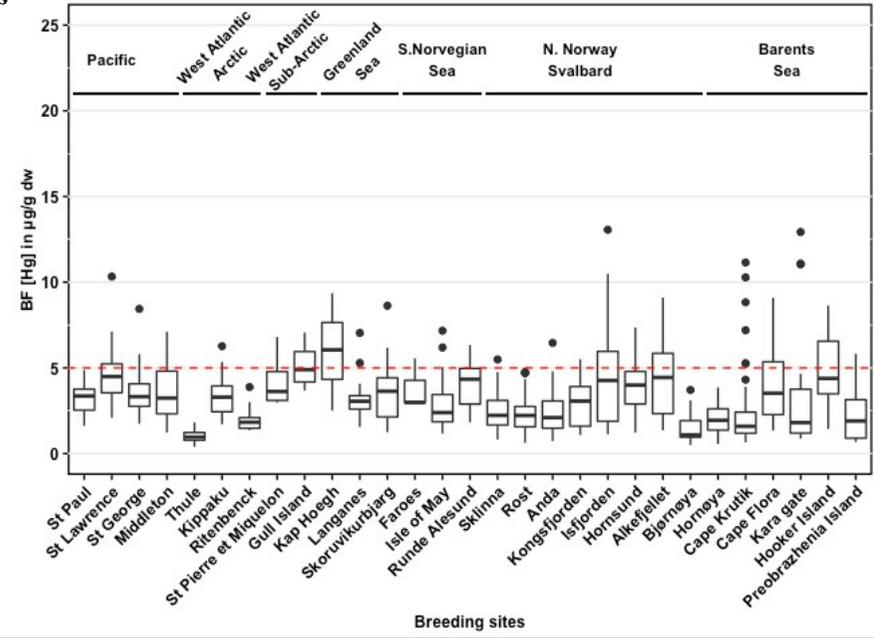


Adjusted blood



Body feathers

Black-legged kittiwakes

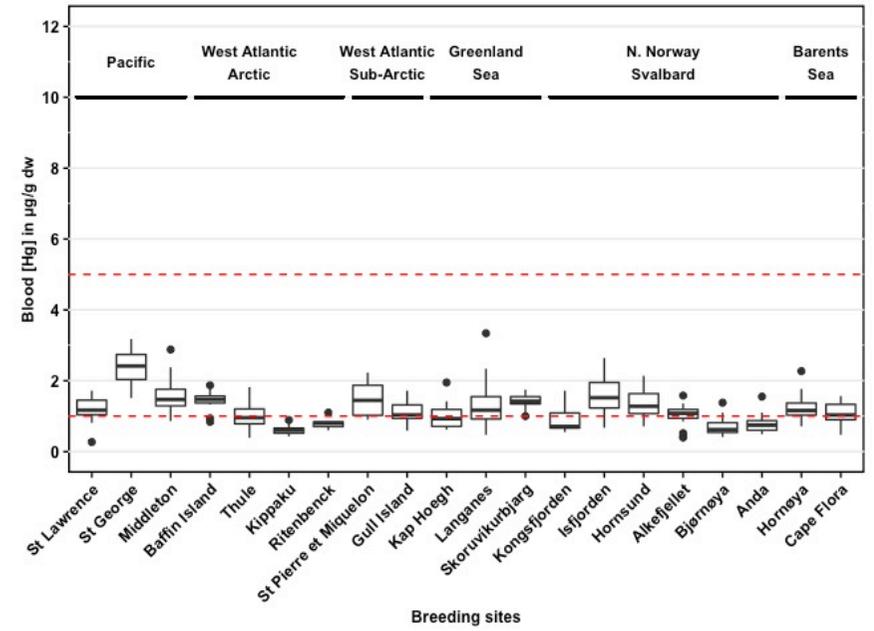


Black-legged kittiwakes



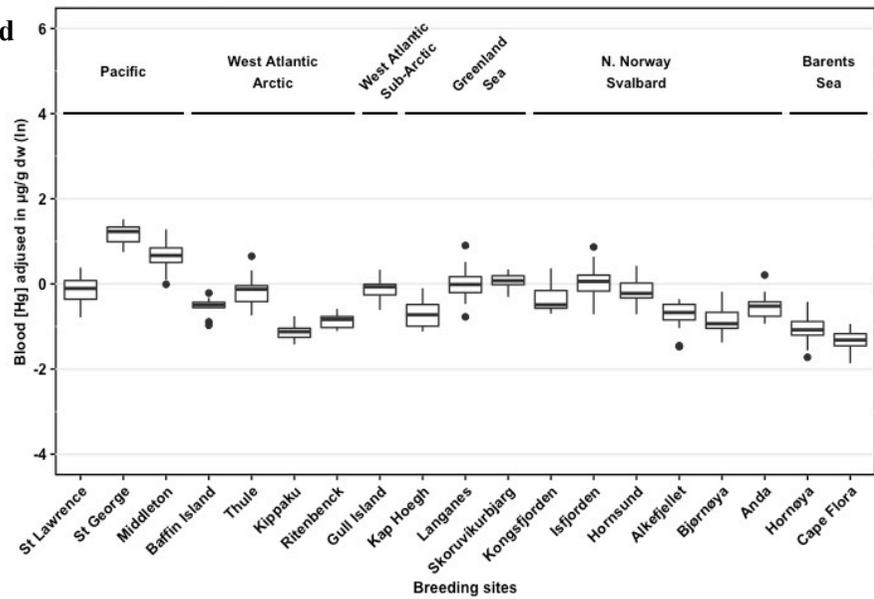
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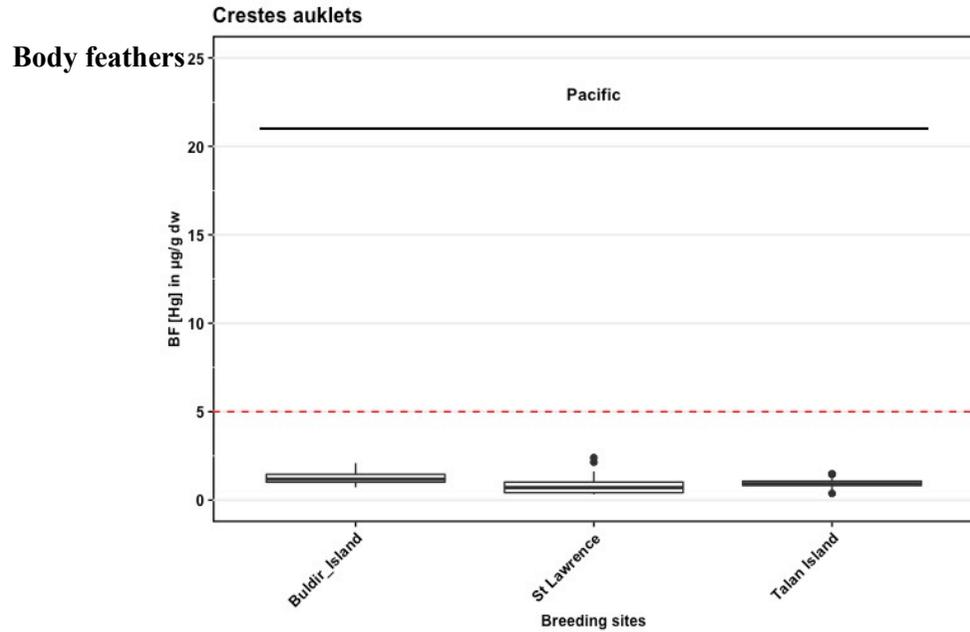
Black-legged kittiwakes



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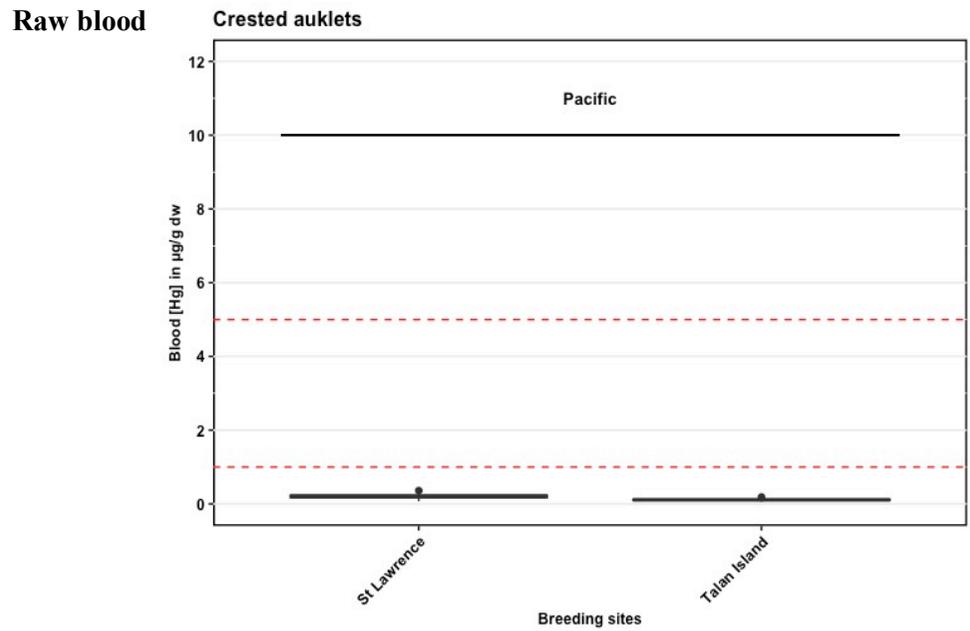
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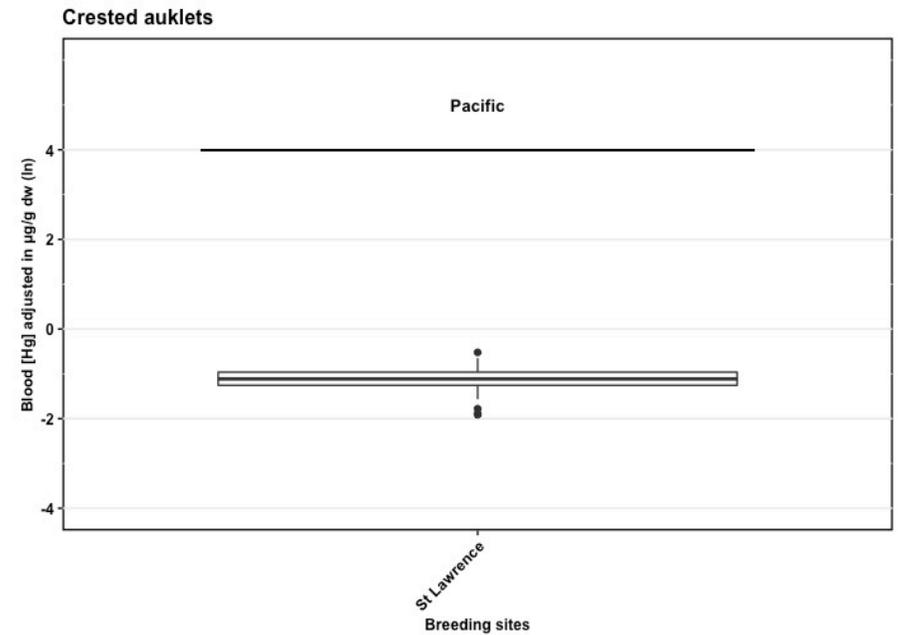


Crested auklets

© A. Will



Adjusted blood

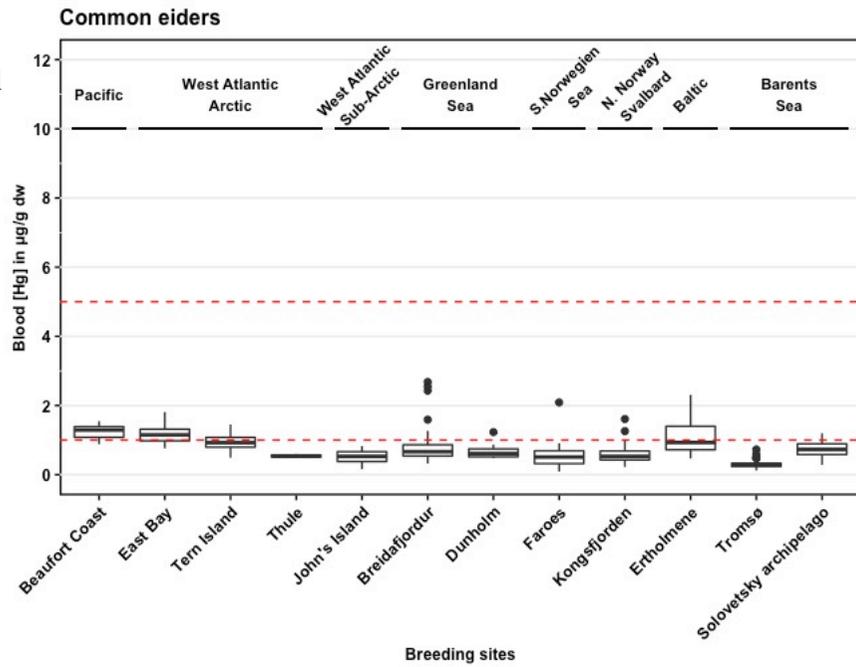


Common eiders

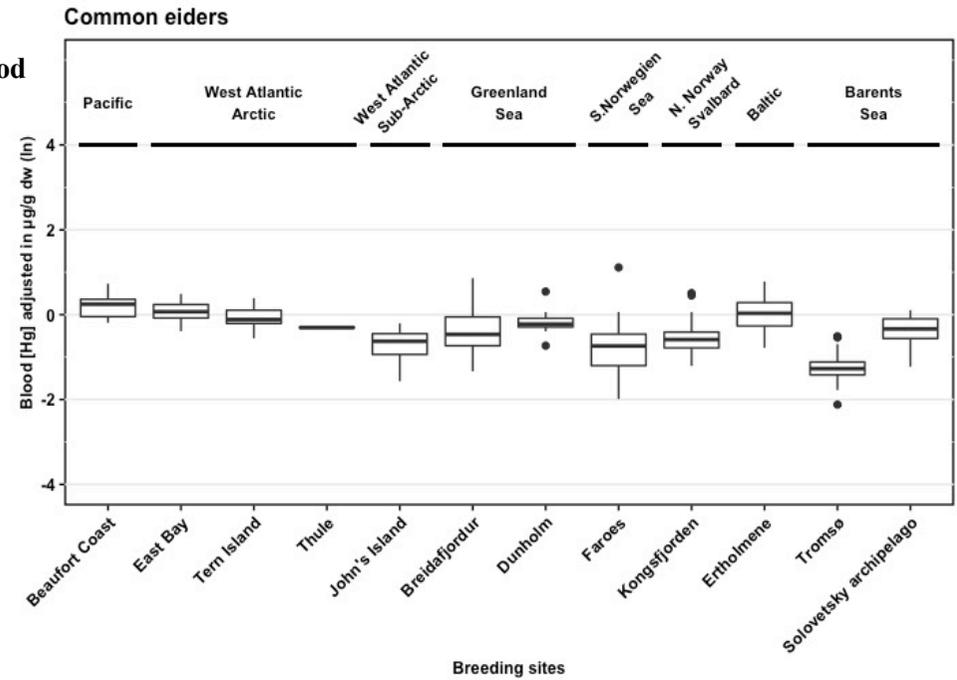
© C. Albert



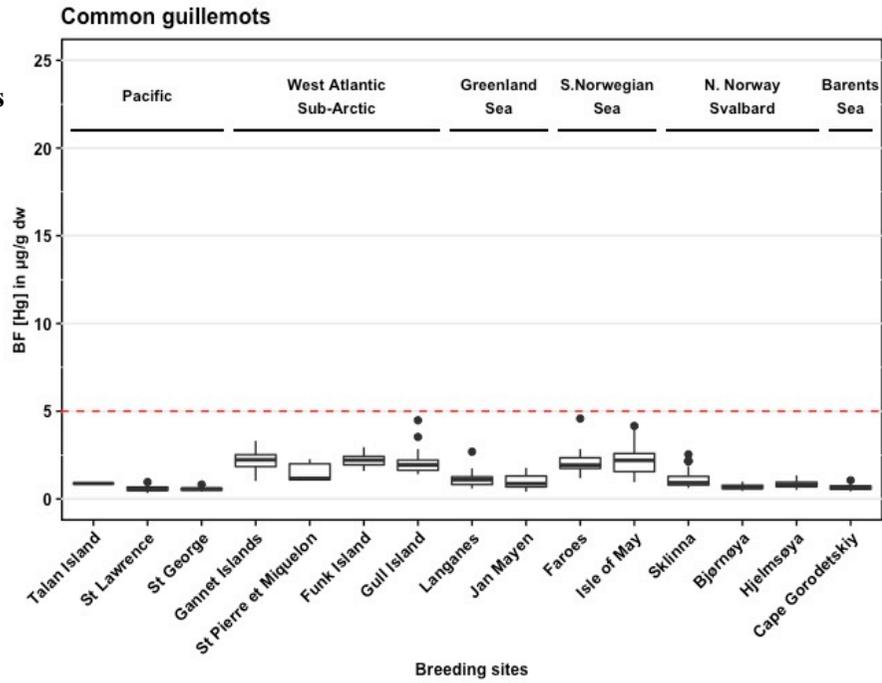
Raw blood



Adjusted blood



Body feathers

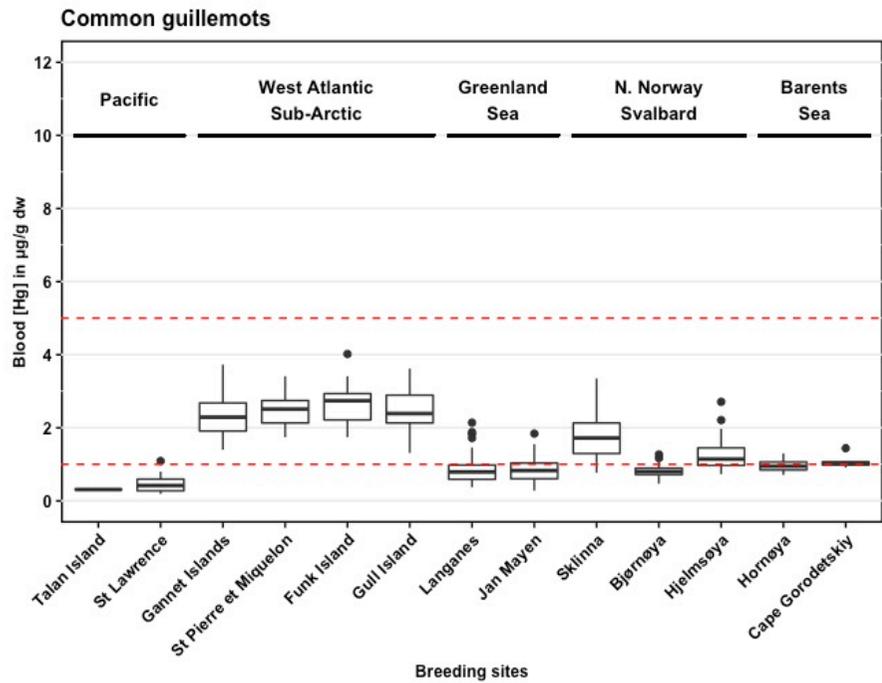


Common guillemots

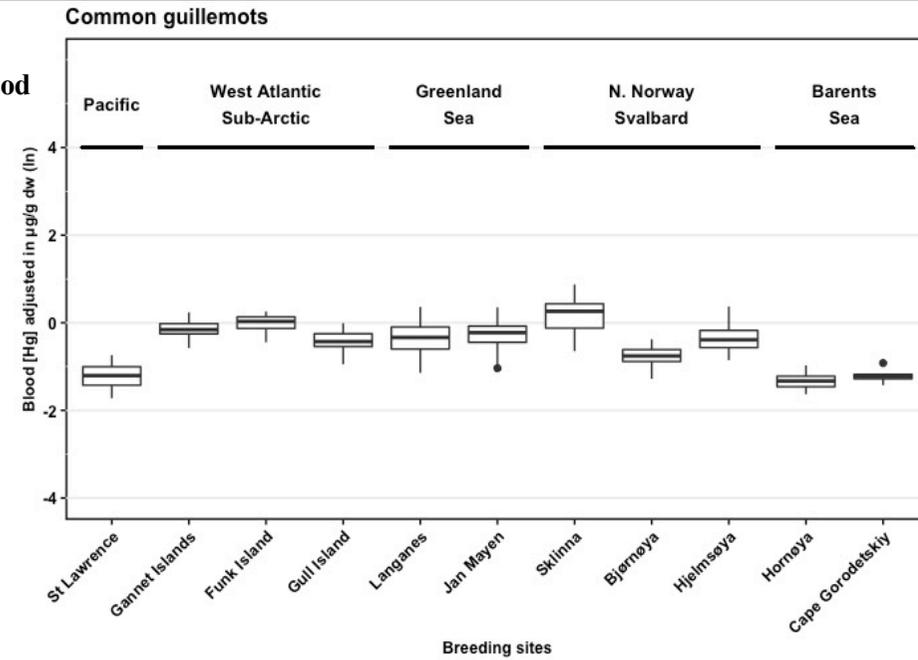


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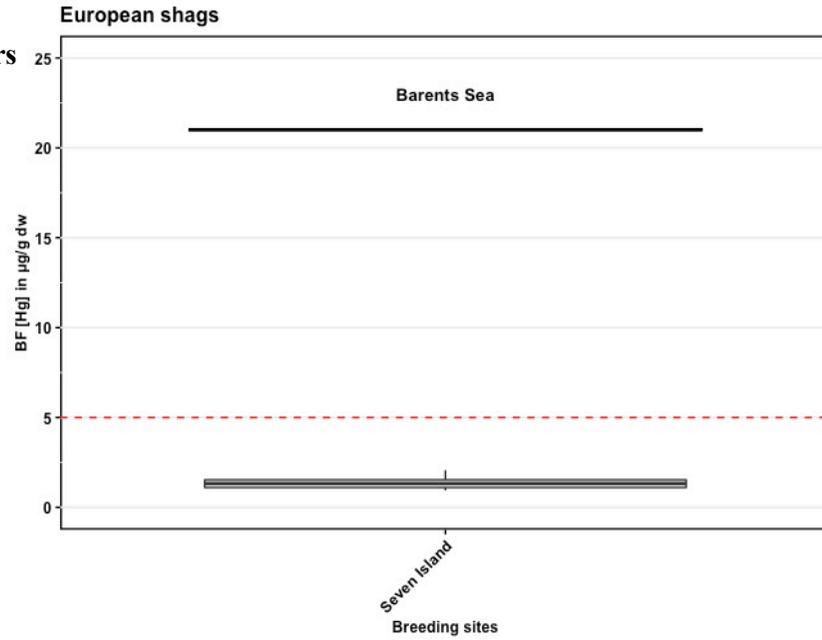
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Adjusted blood



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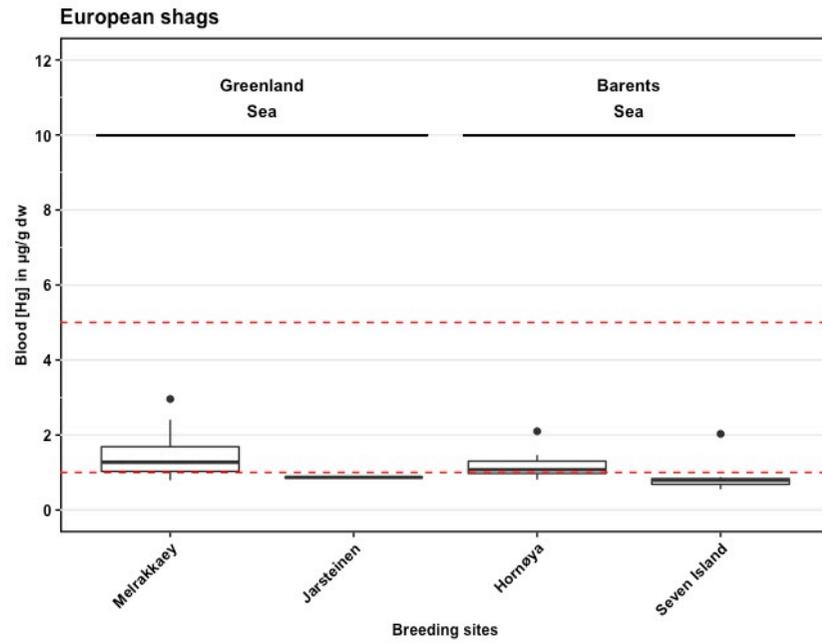


European shags

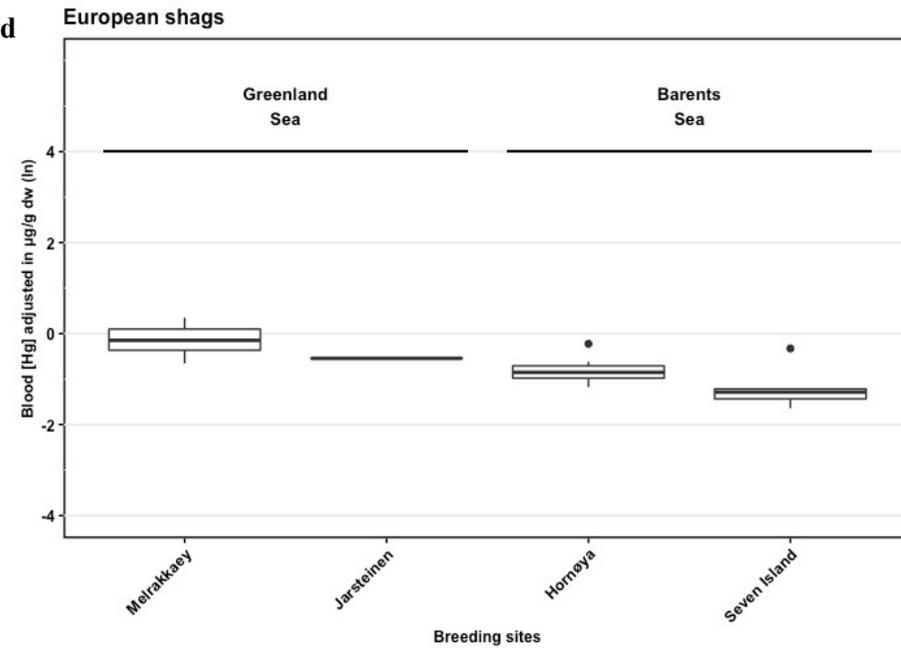


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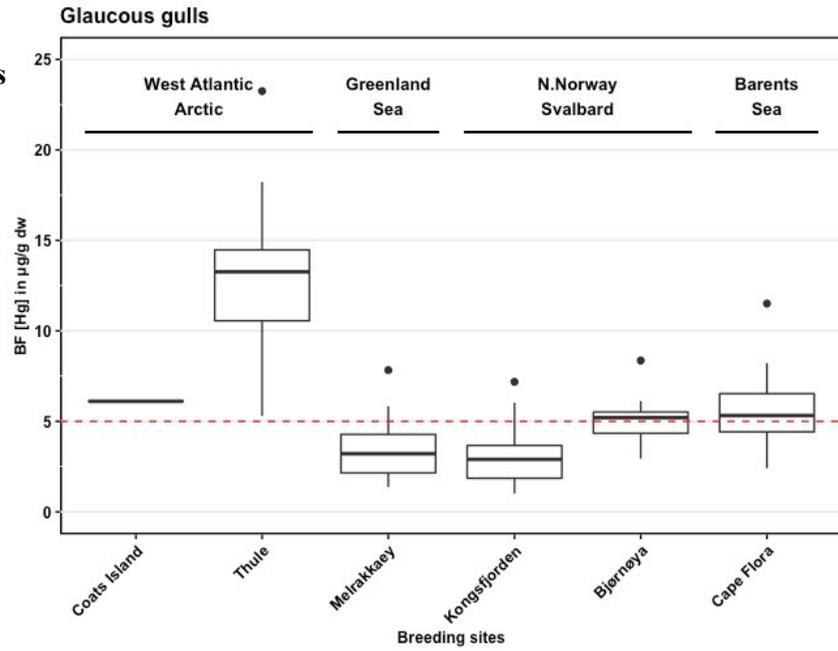
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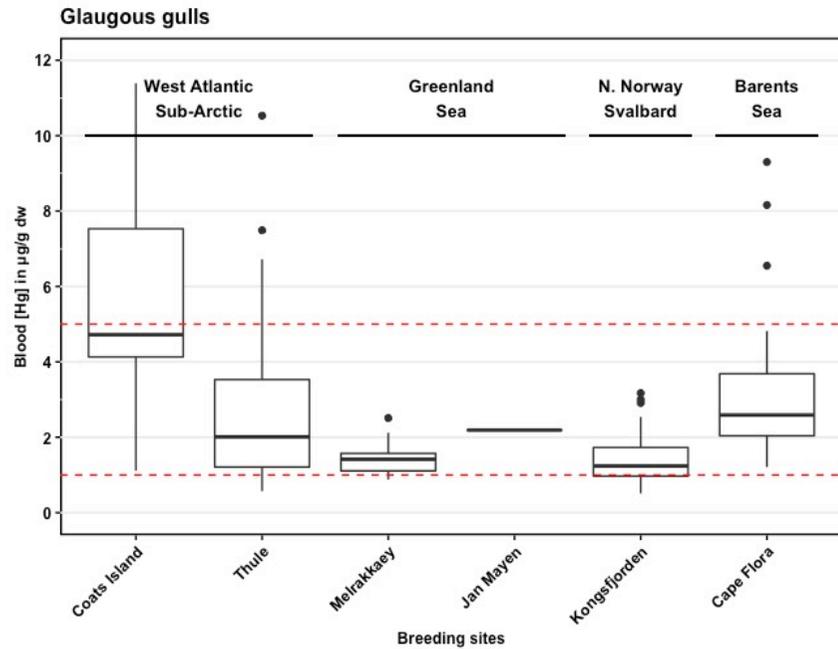
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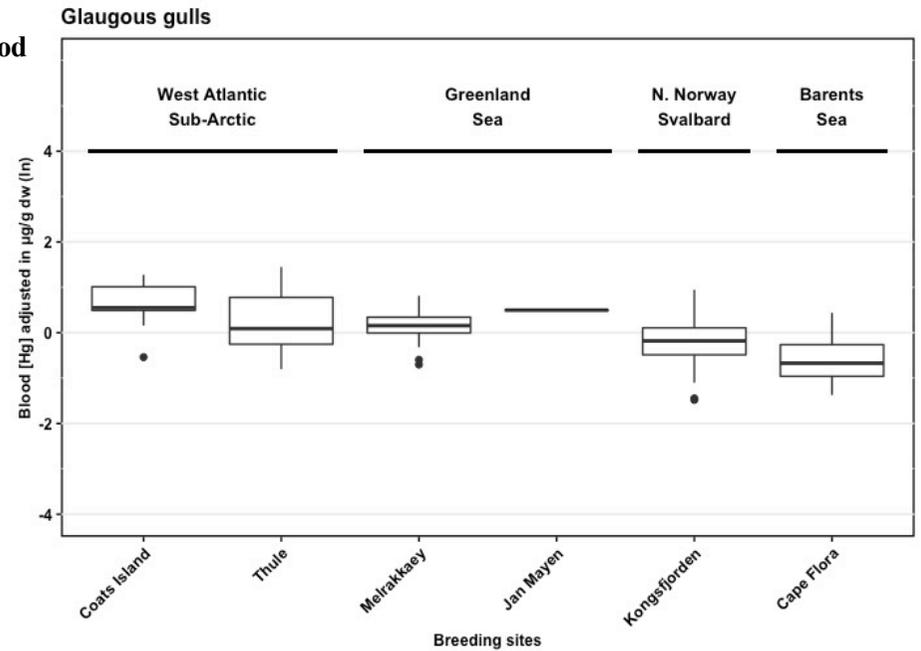
Glaucous gulls



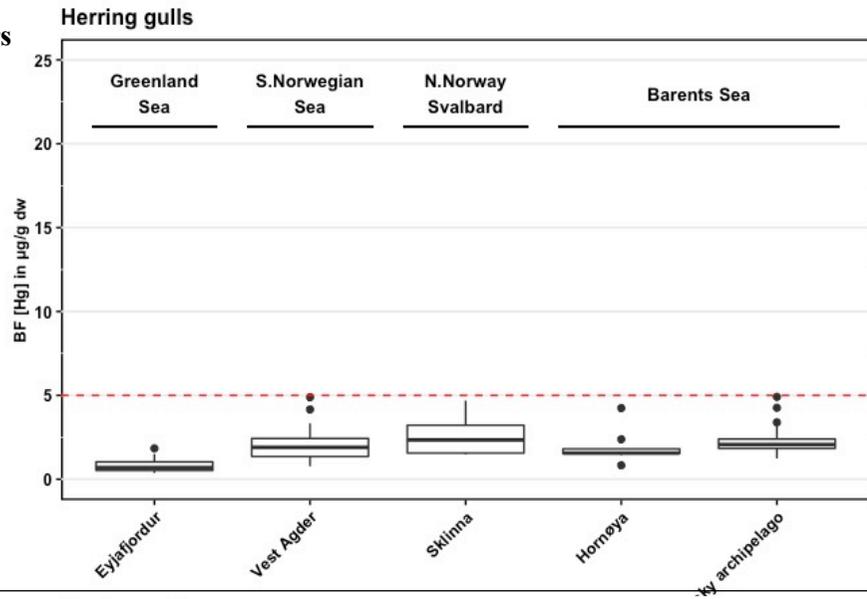
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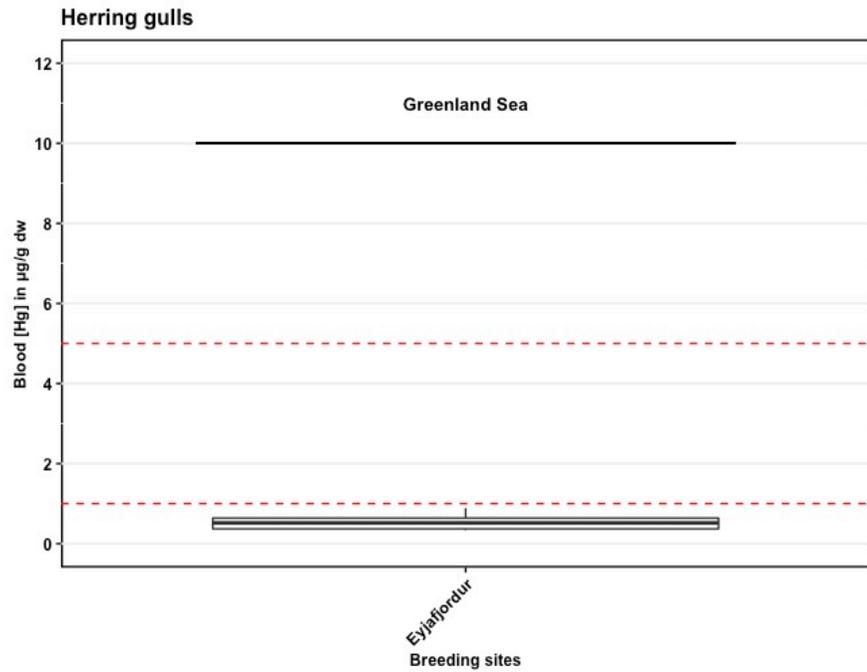
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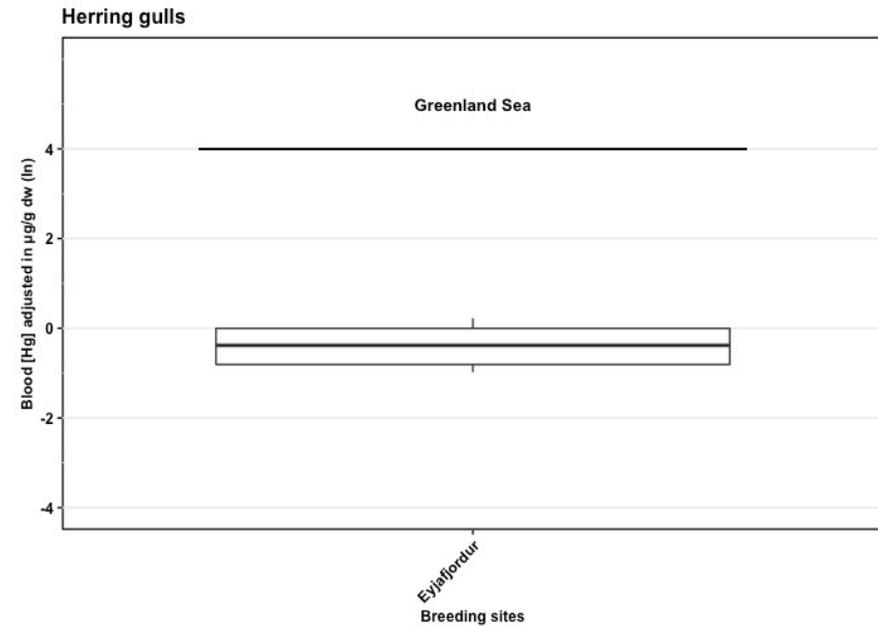
Herring gulls



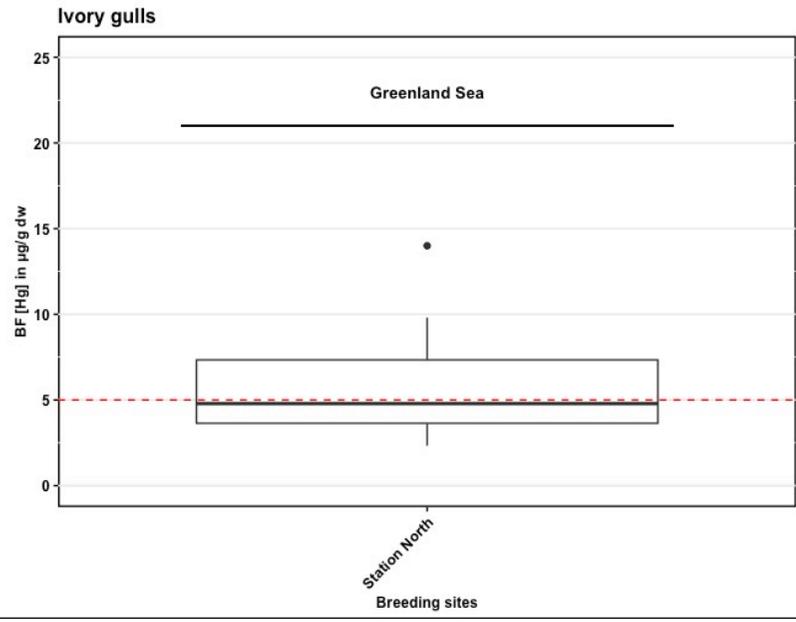
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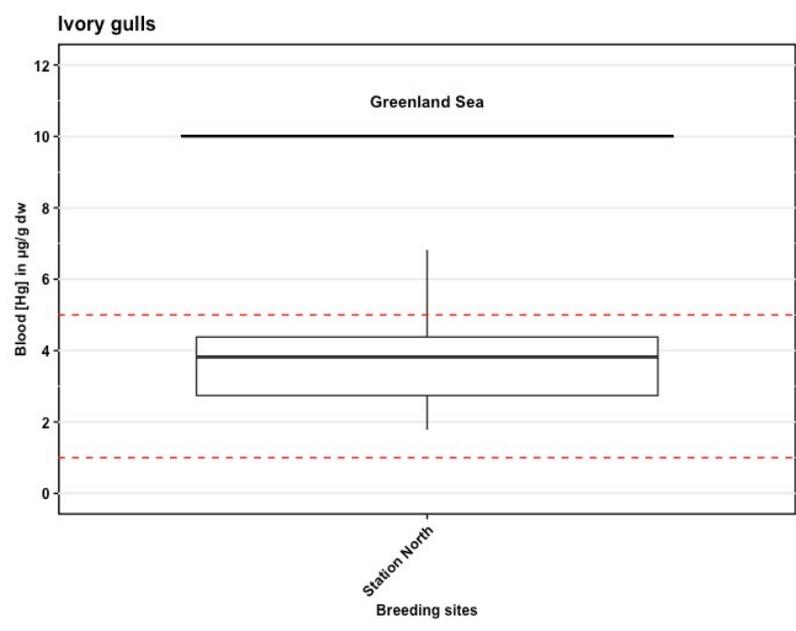


Ivory gulls

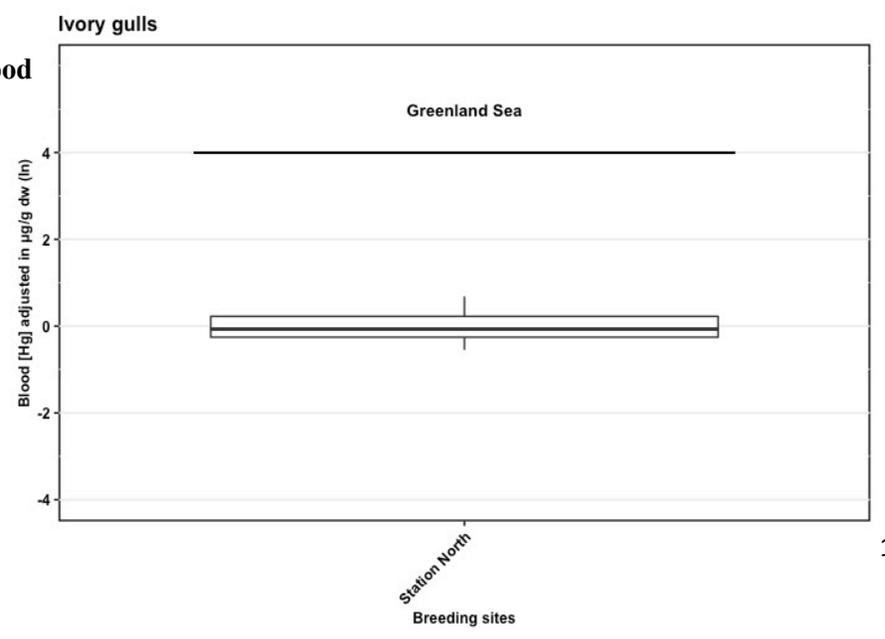
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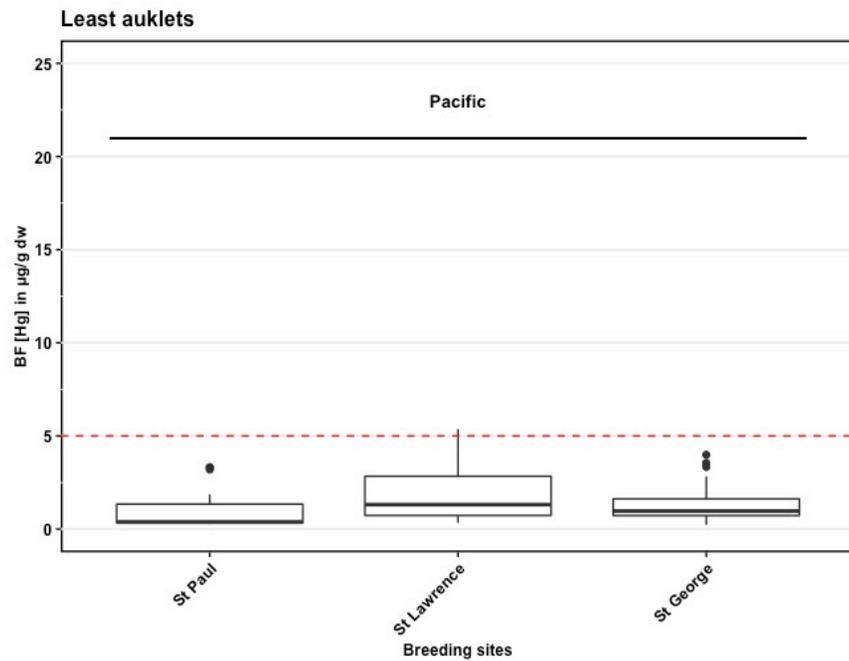
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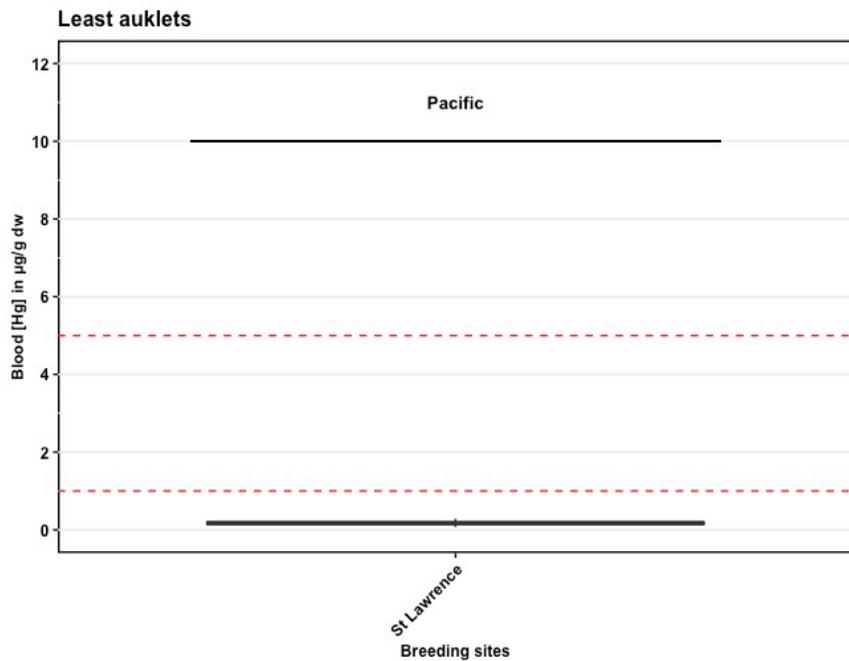
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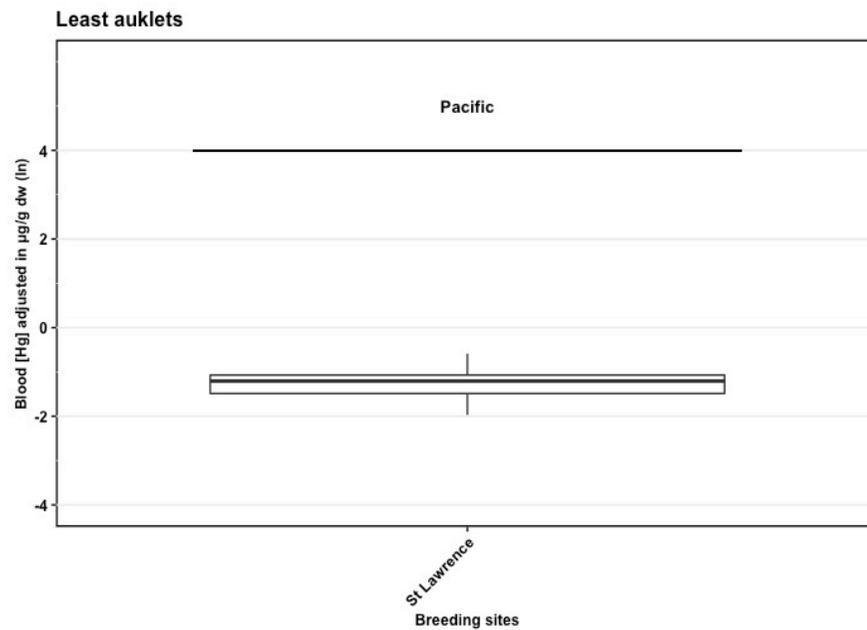
Least auklets



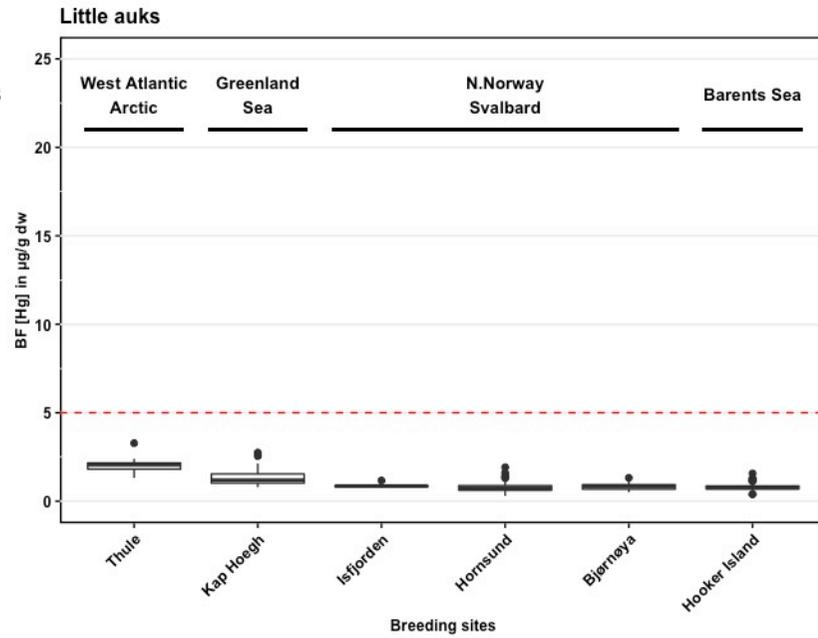
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Adjusted blood



Body feathers

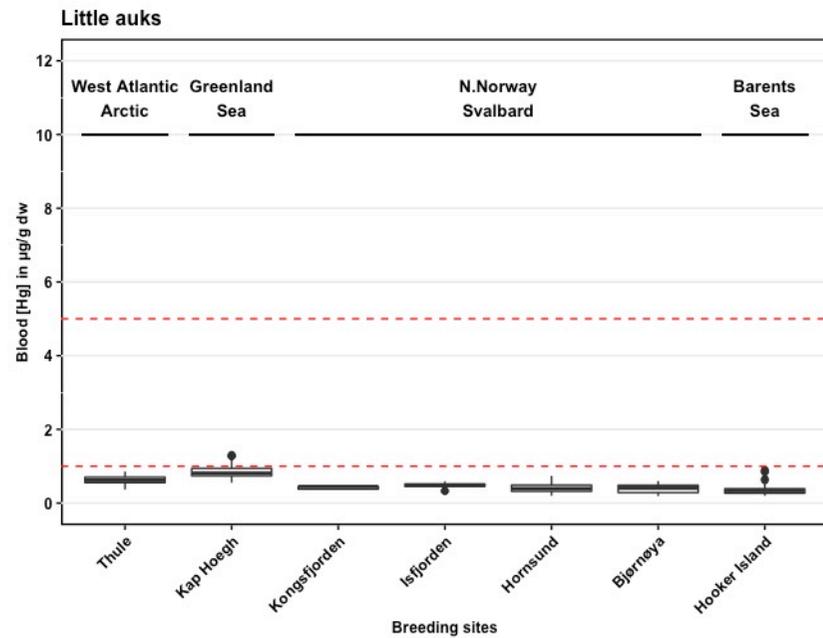


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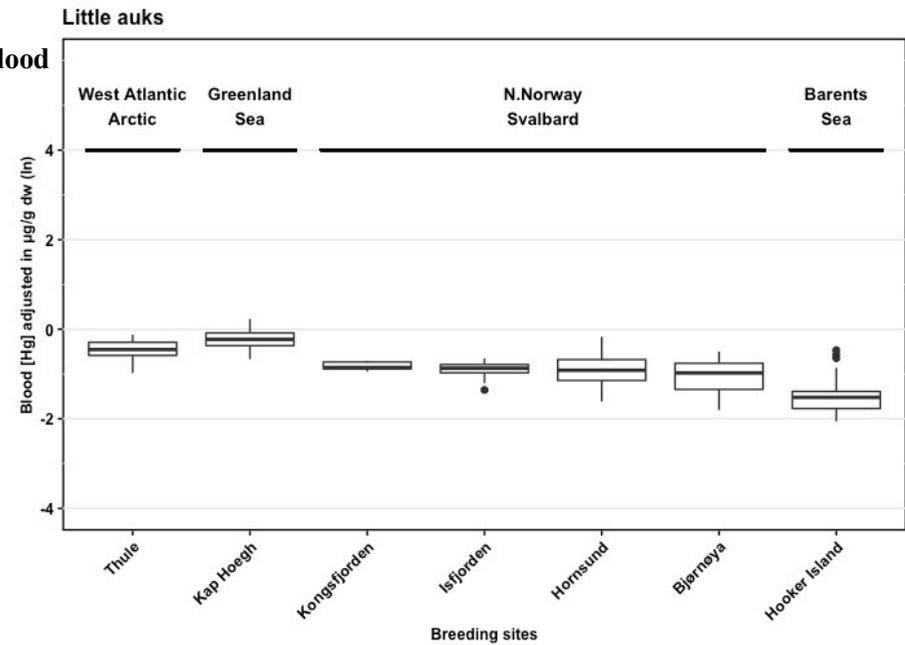
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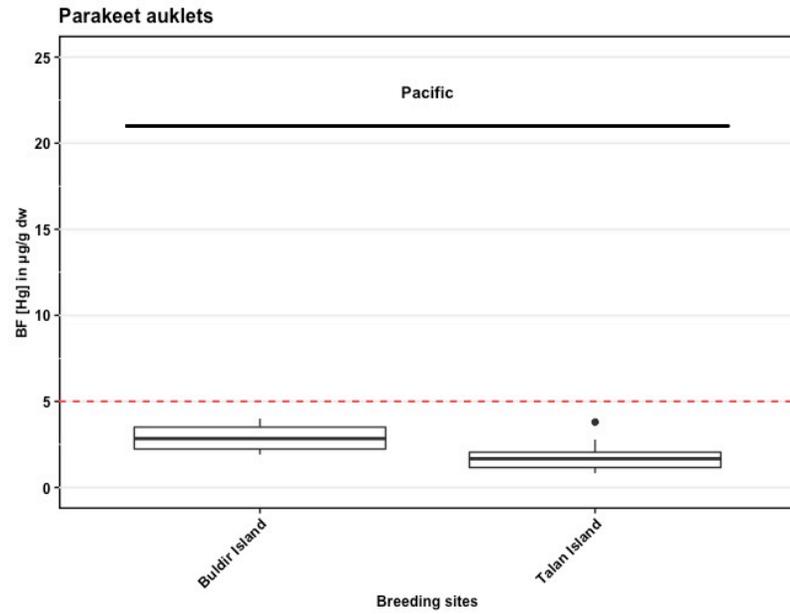
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Body feathers

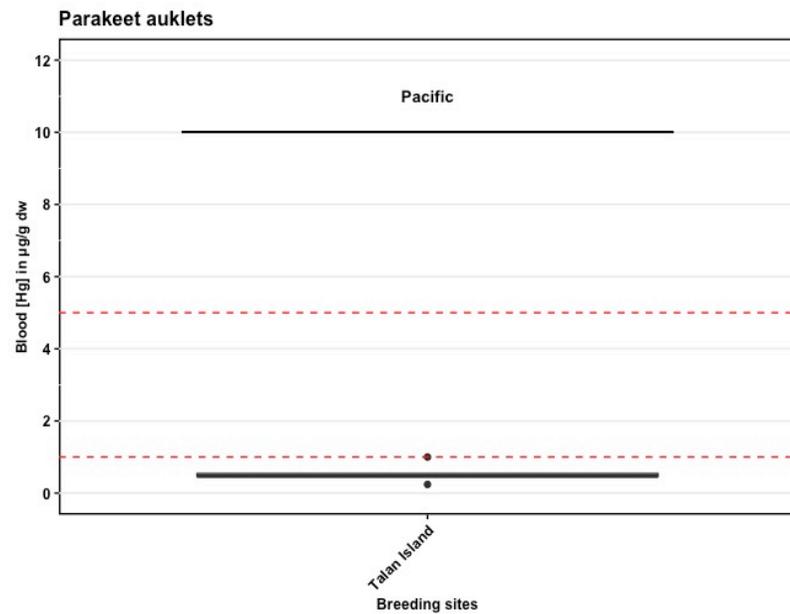


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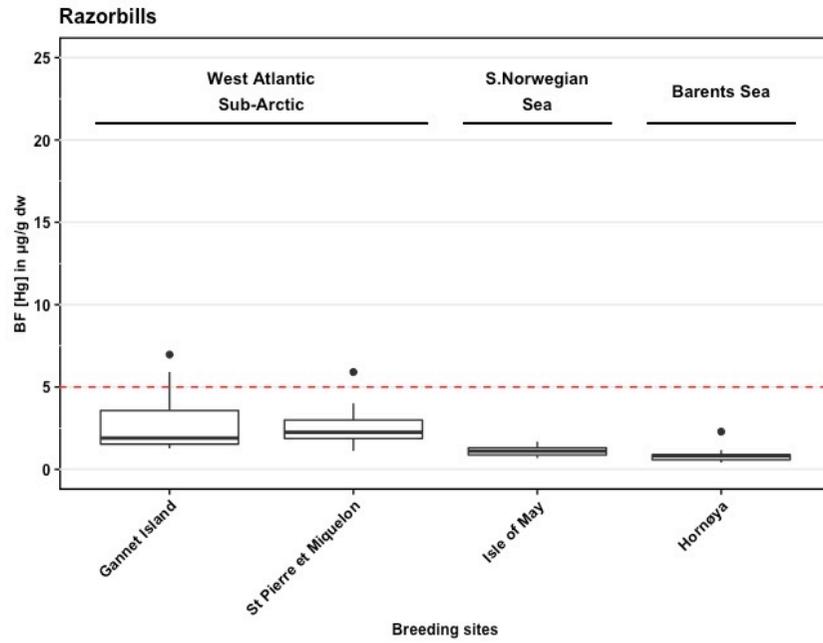
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Raw blood



Body feathers

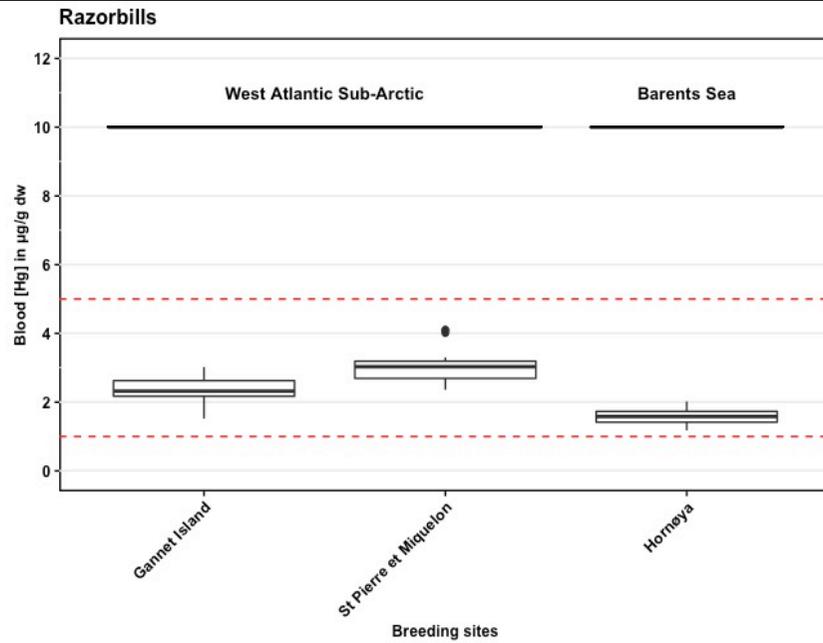


Razorbills

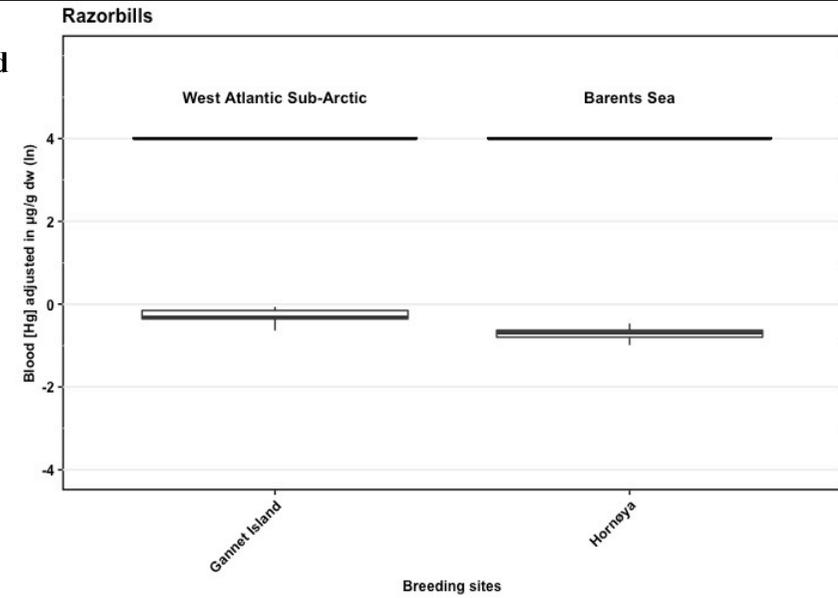


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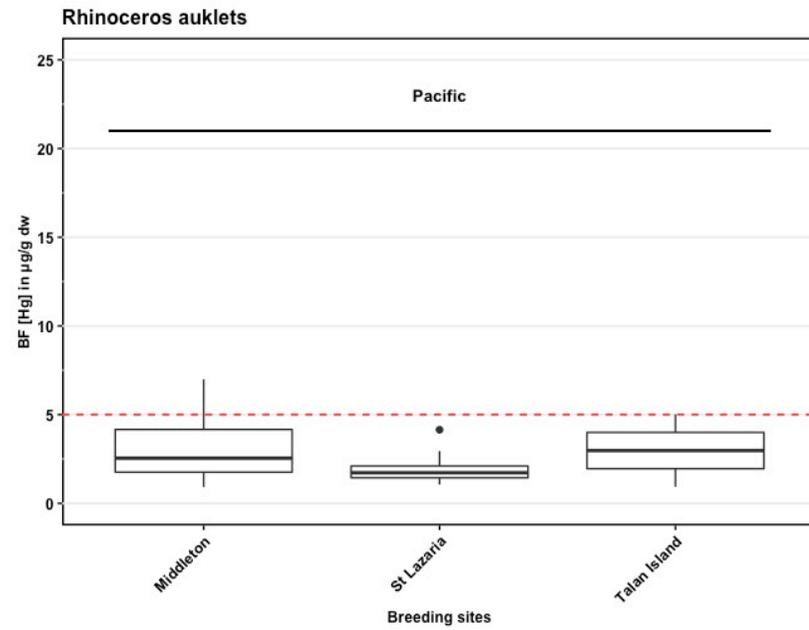
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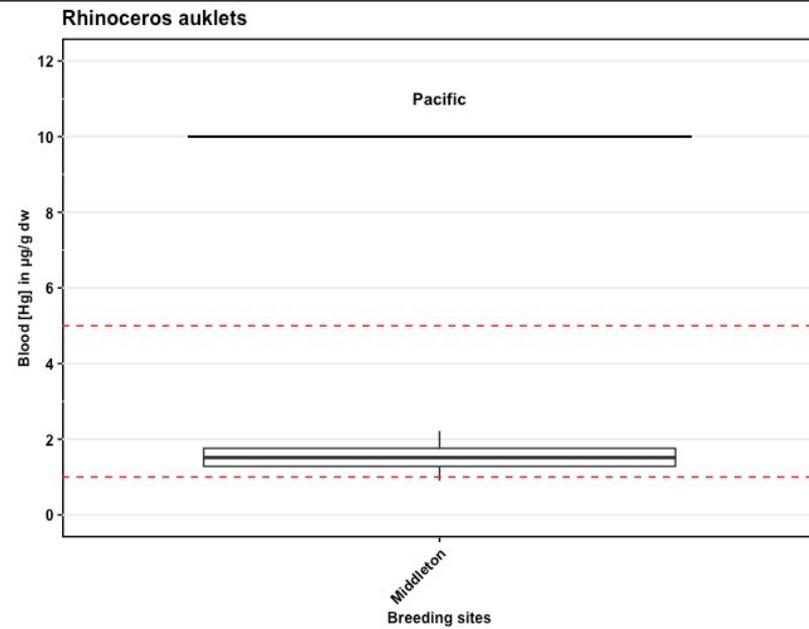
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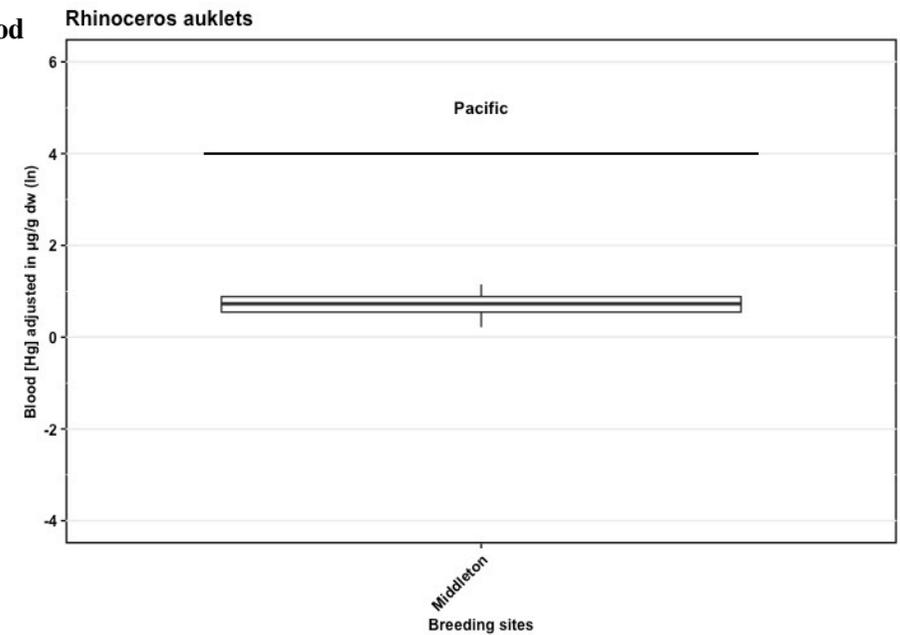
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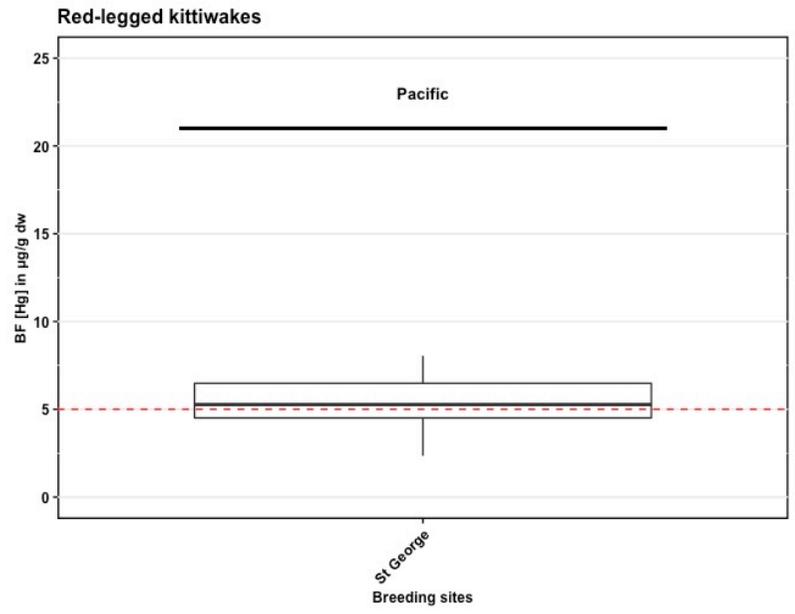
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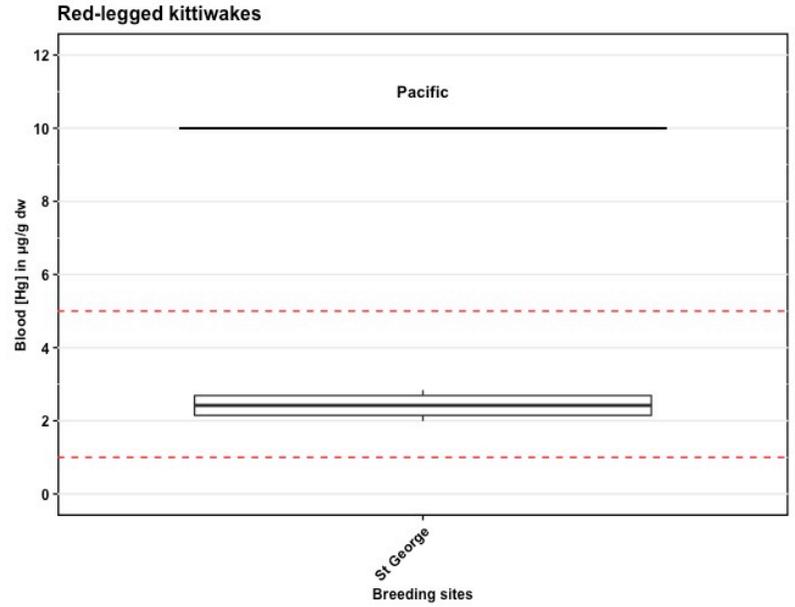
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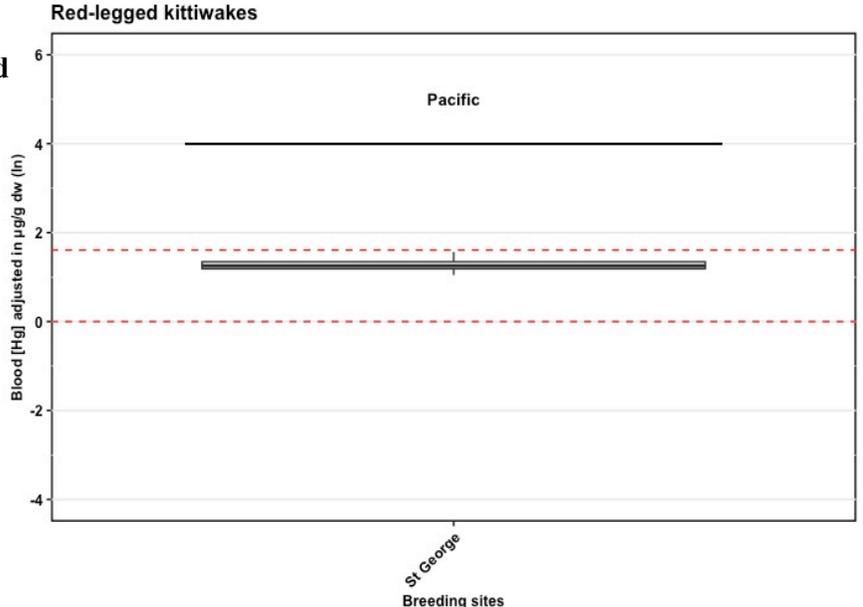
Red-legged kittiwakes



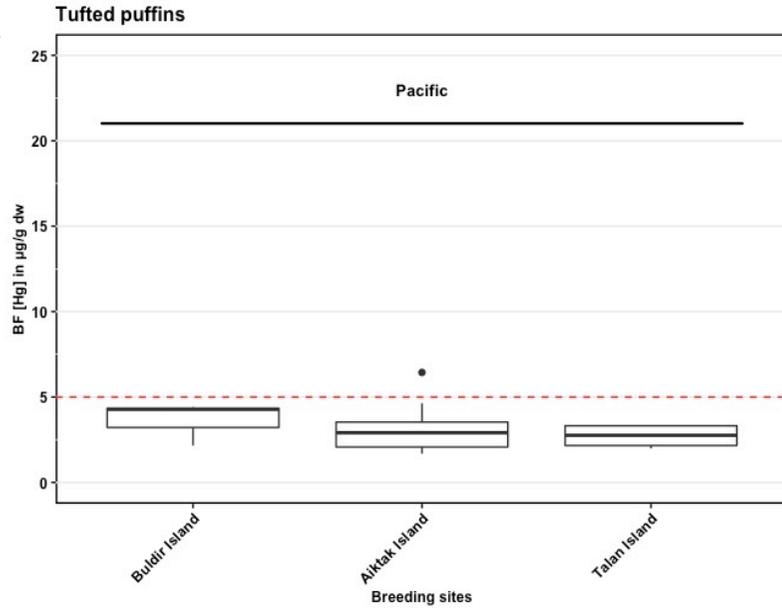
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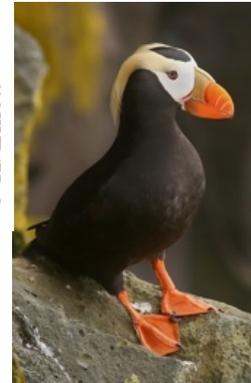
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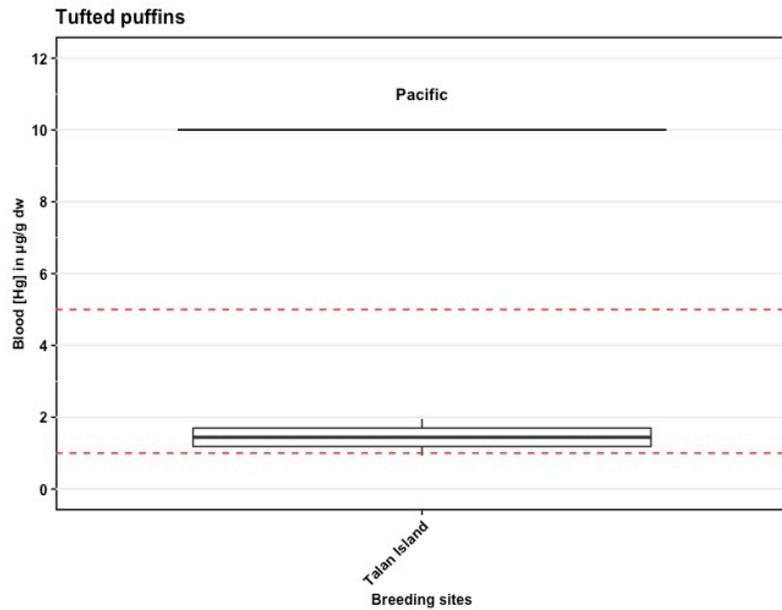
Body feathers



Tufted puffins



Raw blood



Annex 3

Mean Hg concentrations ($\mu\text{g/g dw}$) and $\delta^{15}\text{N}$ in blood, in addition to the blood adjusted per species, regions and sampling sites.

Species	Region	Sampling site	Blood mean \pm SD	n	d15N in Blood mean \pm SD	n	Blood adjusted mean \pm SD	n	BF mean \pm SD	n	
Ancient murrelet	Pacific	Buldir Island	NA	NA	NA	NA	NA	NA	1.94 \pm 0.74	2	
		Chowiet Island	NA	NA	NA	NA	NA	NA	NA	NA	
		Talan Island	0.56 \pm 0.19	12	NA	NA	NA	NA	2.91 \pm 1.30	29	
Atlantic puffin	Barent Sea	Hornoya	0.81 \pm 0.18	56	13.58 \pm 0.45	56	-1.29 \pm 0.26	56	1.08 \pm 0.43	40	
		Seven Island	0.88 \pm 0.17	20	14.43 \pm 0.23	50	-1.51 \pm 0.19	20	0.92 \pm 0.43	28	
	Greeland Sea	Grimsey	1.50 \pm 0.46	36	11.59 \pm 0.22	36	0.38 \pm 0.29	36	3.22 \pm 1.26	31	
		Papey	1.61 \pm 0.49	42	11.47 \pm 0.41	42	0.48 \pm 0.33	42	4.05 \pm 1.83	16	
	NorthNorway Svalbard	Anda	0.89 \pm 0.30	7	12.60 \pm 0.16	7	-0.50 \pm 0.36	7	1.35 \pm 0.60	15	
		Bjornoya	0.62 \pm 0.22	35	11.97 \pm 0.30	35	-0.73 \pm 0.34	35	2.36 \pm 1.31	32	
		Hjelmsøya	0.88 \pm 0.19	81	12.5 \pm 0.34	81	-0.46 \pm 0.22	81	0.93 \pm 0.41	42	
		Rost	NA	NA	NA	NA	NA	NA	1.40 \pm 0.89	22	
		Sklinna	NA	NA	NA	NA	NA	NA	1.96 \pm 1.09	58	
	NorthwestAtlantic SubArctic	Gannet Islands	1.22 \pm 0.32	58	13.23 \pm 0.37	58	-0.34 \pm 0.32	58	4.57 \pm 1.45	16	
		Gull Island	1.45 \pm 0.34	40	13.99 \pm 0.30	40	-0.49 \pm 0.26	40	3.78 \pm 1.42	20	
		St Pierre et Miquelon	1.69 \pm 0.51	22	NA	NA	NA	NA	NA	NA	
	SouthNorwegian Sea	Faroes	NA	NA	NA	NA	NA	NA	3.42 \pm 1.05	3	
	Black legged kittiwake	Barent Sea	Cape Flora	1.09 \pm 0.27	48	14.61 \pm 0.44	48	-1.32 \pm 0.21	48	3.96 \pm 1.94	72
			Cape Krutik	NA	NA	NA	NA	NA	NA	2.27 \pm 2.00	76
Hooker Island			NA	NA	NA	NA	NA	NA	4.83 \pm 2.53	8	
Hornoya			1.24 \pm 0.41	15	14.2 \pm 0.20	15	-1.08 \pm 0.33	15	1.97 \pm 0.81	73	
Kara gate			NA	NA	NA	NA	NA	NA	3.37 \pm 3.51	23	
Greeland Sea		Kap Hoegh	1.00 \pm 0.32	25	14.30 \pm 0.17	25	-0.73 \pm 0.27	25	6.03 \pm 2.05	20	
		Langanes	1.32 \pm 0.55	37	12.41 \pm 0.52	37	0.00 \pm 0.31	37	3.15 \pm 1.19	23	
		Skoruvíkurbjarg	1.42 \pm 0.20	15	12.72 \pm 0.24	15	0.06 \pm 0.17	15	3.68 \pm 2.04	15	
NorthNorway Svalbard		Alkefjellet	1.03 \pm 0.31	19	14.19 \pm 0.77	19	-0.72 \pm 0.32	19	4.55 \pm 2.15	33	
		Anda	0.78 \pm 0.25	20	12.26 \pm 0.23	20	-0.55 \pm 0.29	20	2.30 \pm 1.12	75	
		Bjornoya	0.68 \pm 0.20	55	13.11 \pm 0.41	67	-0.87 \pm 0.27	54	1.45 \pm 0.73	68	
		Hjelmsøya	NA	NA	NA	NA	NA	NA	NA	NA	

		Hornsund	1.34±0.39	16	12.84±0.17	15	-0.15±0.31	15	3.98±1.81	16
		Isfjorden	1.58±0.48	37	12.73±0.52	37	0.05±0.33	37	4.62±3.01	48
		Kongsfjorden	0.89±0.35	20	11.77±0.25	20	-0.33±0.35	20	2.96±1.38	34
		Rost	NA	NA	NA	NA	NA	NA	2.28±0.97	74
		Sklinna	NA	NA	NA	NA	NA	NA	2.43±1.02	74
NorthwestAtlantic Arctic		Baffin Island	1.45±0.29	14	15.81±0.19	14	-0.52±0.20	14	NA	NA
		Kippaku	0.61±0.11	25	14.83±0.11	25	-1.12±0.18	25	3.35±1.10	26
		Rittenbank	0.79±0.14	10	14.86±0.23	10	-0.87±0.17	10	2.04±0.80	10
		Thule	0.98±0.38	18	12.74±0.71	18	-0.16±0.33	18	0.99±0.39	17
NorthwestAtlantic SubArctic		Gull Island	1.12±0.30	20	12.46±0.48	20	-0.10±0.24	20	5.19±1.11	20
		St Pierre et Miquelon	1.49±0.54	6	NA	NA	NA	NA	4.26±1.77	4
Pacific		Buldir Island	NA	NA	NA	NA	NA	NA	NA	NA
		Middleton	1.57±0.41	61	14.66±0.34	60	0.67±0.29	60	3.55±1.63	41
		Preobrazhenia Island	NA	NA	NA	NA	NA	NA	2.36±1.85	7
		Saint George	2.37±0.56	8	14.45±0.23	8	1.16±0.27	8	3.63±1.37	30
		Saint Lawrence	1.20±0.38	16	15.91±0.69	16	-0.11±0.33	16	4.54±1.69	32
		Saint Paul	NA	NA	NA	NA	NA	NA	3.20±1.00	11
SouthNorwegian Sea		Faroes	NA	NA	NA	NA	NA	NA	3.83±1.50	3
		Isle of May	NA	NA	NA	NA	NA	NA	2.80±1.36	39
		Runde Alesund	NA	NA	NA	NA	NA	NA	4.17±1.38	17
Brunnich guillemot	Barent Sea	Cape Flora	0.84±0.19	37	13.77±0.31	37	-1.31±0.22	37	0.74±0.21	61
		Gorodetskiy Cape	0.77±0.16	10	14.09±0.30	29	-1.57±0.21	10	0.69±0.13	29
		Hornoya	0.87±0.15	64	13.99±0.23	64	-1.34±0.17	64	0.74±0.20	60
		Kara gate	NA	NA	NA	NA	NA	NA	0.64±0.20	40
		Oranskie islands	0.50±0.05	4	14.58±0.50	31	-1.74±0.08	4	0.70±0.20	34
Greeland Sea		Grimsey	0.62±0.16	35	11.79±0.13	35	-0.55±0.26	35	1.51±0.21	19
		Jan Mayen	1.31±0.37	86	12.23±0.32	86	0.08±0.23	86	1.63±0.32	65
		Langanes	0.39±0.11	29	11.53±0.29	29	-0.96±0.28	29	1.35±0.21	27
		Latrabjarg	0.66	1	12.83	1	-0.72	1	NA	NA

NorthNorway Svalbard	Alkefjellet	0.35±0.16	31	12.92±0.43	31	-1.55±0.34	31	0.63±0.20	32	
	Bjornoya	0.20±0.07	42	12.54±0.55	52	-1.97±0.37	42	0.50±0.10	50	
	Isfjorden	0.58±0.25	33	12.75±0.49	33	-1.01±0.39	33	1.17±0.37	32	
NorthwestAtlantic Arctic	Baffin Island	4.11±0.86	20	15.24±0.36	19	0.70±0.19	19	NA	NA	
	Coats Island	1.48±0.84	72	14.86±0.43	72	-0.32±0.37	72	2.27±0.84	50	
	Kippaku	1.43±0.50	30	13.81±0.27	30	-0.04±0.31	30	1.79±0.44	20	
	Thule	2.17±0.75	19	13.57±0.68	19	0.42±0.33	19	2.61±0.71	17	
NorthwestAtlantic SubArctic	Gannet Islands	2.18±0.50	41	13.74±0.29	41	0.03±0.21	41	2.39±0.95	41	
Pacific	Buldir Island	NA	NA	NA	NA	NA	NA	NA	NA	
	Saint George	0.58±0.21	7	13.25±0.72	7	0.17±0.25	7	0.72±0.25	42	
	Saint Lawrence	0.66±0.60	40	15.66±1.39	40	-0.96±0.56	40	0.77±0.33	40	
Common eider	Baltic	Ertholmene	1.12±0.51	25	10.01±1.34	25	0.02±0.38	25	NA	NA
	Barent Sea	Seven Island	NA	NA	NA	NA	NA	NA	0.48	1
		Solovetsky archipelago	0.72±0.22	24	10.15±0.44	24	-0.37±0.33	24	1.16±0.47	69
	Greeland Sea	Breidafjordur	0.79±0.50	54	11.83±1.00	54	-0.40±0.50	54	0.95±0.44	76
		Dunholm	0.68±0.23	10	10.68±0.74	10	-0.18±0.33	10	1.09±0.64	10
	NorthNorway Svalbard	Kongsfjorden	0.59±0.23	84	11.02±0.91	84	-0.58±0.33	84	1.07±0.36	70
		Selvaer	NA	NA	NA	NA	NA	NA	0.89±0.26	43
		Sklinna	NA	NA	NA	NA	NA	NA	NA	NA
		Tromso	0.30±0.10	81	11.19±0.44	81	-1.26±0.26	81	0.72±0.53	75
	NorthwestAtlantic Arctic	East Bay	1.17±0.28	23	12.70±0.67	23	0.07±0.23	23	1.23±0.68	23
		Tern Island	0.94±0.21	24	12.37±0.52	24	-0.06±0.23	24	NA	NA
		Thule	0.54±0.08	2	11.27±0.48	2	-0.30±0.04	2	NA	NA
	NorthwestAtlantic SubArctic	Johns Island	0.53±0.20	19	12.04±0.76	19	-0.73±0.38	19	NA	NA
	Pacific	Beaufort Coast	1.23±0.20	33	15.31±0.66	33	0.19±0.26	33	NA	NA
		Buldir Island	NA	NA	NA	NA	NA	NA	1.33	1
	SouthNorwegian Sea	Faroes	0.55±0.38	27	10.22±1.04	27	-0.77±0.62	27	1.39±2.06	78
Vest Agder		NA	NA	NA	NA	NA	NA	NA	NA	
Common guillemot	Barent Sea	Gorodetskiy Cape	1.07±0.19	6	14.60±0.23	27	-1.21±0.17	6	0.66±0.14	47

		Hornoya	0.97±0.16	35	14.34±0.18	35	-1.33±0.17	35	NA	NA
Greeland Sea		Jan Mayen	0.84±0.29	91	11.82±0.48	91	-0.28±0.29	91	0.98±0.37	63
		Langanes	0.85±0.36	85	12.01±0.47	85	-0.33±0.34	85	1.11±0.39	37
NorthNorway Svalbard		Bjornoya	0.81±0.15	51	13.56±0.41	75	-0.75±0.21	51	0.67±0.13	48
		Hjelmsøya	1.23±0.37	72	13.47±0.39	72	-0.36±0.27	72	0.85±0.20	58
		Sklinna	1.82±0.68	38	12.57±0.32	38	0.20±0.39	38	1.07±0.40	68
NorthwestAtlantic SubArctic		Funk Island	2.65±0.56	19	14.30±0.46	19	-0.01±0.17	19	2.21±0.39	23
		Gannet Islands	2.36±0.60	33	14.34±0.56	33	-0.15±0.18	33	2.17±0.50	33
		Gull Island	2.42±0.57	40	15.02±0.20	40	-0.42±0.21	40	2.04±0.60	40
		St Pierre et Miquelon	2.47±0.47	26	NA	NA	NA	NA	1.53±0.56	5
Pacific		Saint George	NA	NA	NA	NA	NA	NA	0.58±0.11	12
		Saint Lawrence	0.46±0.22	27	16.21±1.12	27	-1.22±0.25	27	0.59±0.17	36
		Talan Island	0.31	1	NA	NA	NA	NA	0.88	1
SouthNorwegian Sea		Faroes	NA	NA	NA	NA	NA	NA	2.26±1.05	8
		Isle of May	NA	NA	NA	NA	NA	NA	2.22±0.88	39
Common gull	Baltic	Estonia	1.29±0.66	21	11.10±1.40	21	-0.08±0.46	21	NA	NA
Crested Auklet	Pacific	Buldir Island	NA	NA	NA	NA	NA	NA	1.30±0.58	4
		Saint Lawrence	0.20±0.06	52	13.95±0.48	52	-1.14±0.28	52	0.84±0.51	33
		Talan Island	0.11±0.05	5	NA	NA	NA	NA	0.95±0.30	15
European shag	Barent Sea	Hornoya	1.18±0.35	12	13.27±0.11	12	-0.83±0.26	12	NA	NA
		Seven Island	0.87±0.42	10	13.44±0.15	40	-1.25±0.36	10	1.35±0.35	13
	Greeland Sea	Melrakkaey	1.38±0.47	54	13.29±0.30	54	-0.15±0.27	54	NA	NA
	SouthNorwegian Sea	Jarsteinen	0.87	1	11.42	1	-0.54	1	NA	NA
Glaucous gull	Barent Sea	Cape Flora	3.14±1.70	39	15.36±0.86	39	-0.56±0.47	39	5.42±1.82	41
	Greeland Sea	Jan Mayen	2.19	1	12.77	1	0.50	1	NA	NA
		Melrakkaey	1.42±0.39	28	12.32±1.30	28	0.14±0.35	28	3.45±1.73	17
	NorthNorway Svalbard	Bjornoya	NA	NA	NA	NA	NA	NA	5.01±1.15	20
		Kongsfjorden	1.43±0.64	53	13.35±1.02	53	-0.24±0.48	53	2.97±1.40	47
NorthwestAtlantic Arctic	Coats Island	5.76±3.05	11	16.21±0.62	11	0.63±0.51	11	6.11	1	

		Thule	2.66±2.21	33	14.45±0.89	33	0.19±0.60	33	13.03±4.75	12
Glaucous winged gull	Pacific	Buldir Island	NA	NA	NA	NA	NA	NA	7.61±3.88	12
Herring gull	Barent Sea	Hornoya	NA	NA	NA	NA	NA	NA	1.89±0.97	9
		Solovetsky archipelago	NA	NA	NA	NA	NA	NA	2.24±0.73	42
	Greeland Sea	Eyja fjordur	0.56±0.20	13	10.71±1.06	13	-0.40±0.41	13	0.82±0.42	16
	NorthNorway Svalbard	Sklinna	NA	NA	NA	NA	NA	NA	2.60±1.12	13
Vest Agder		NA	NA	NA	NA	NA	NA	2.11±1.10	18	
Horned puffin	Pacific	Buldir Island	NA	NA	NA	NA	NA	NA	4.61	1
		Chowiet Island	NA	NA	NA	NA	NA	NA	NA	NA
		Talan Island	NA	NA	NA	NA	NA	NA	4.42±1.3	4
Ivory gull	Greeland Sea	Station North	3.81±1.71	7	16.5±0.45	7	0.00±0.44	7	6.02±3.60	10
Least auklet	Pacific	Saint George	NA	NA	NA	NA	NA	NA	1.29±0.83	47
		Saint Lawrence	0.17±0.06	27	13.73±0.44	27	-1.25±0.36	27	1.82±1.28	34
		Saint Paul	NA	NA	NA	NA	NA	NA	1.05±1.11	18
Lesser black backed gull	Barent Sea	Solovetsky archipelago	NA	NA	NA	NA	NA	NA	2.97±1.17	36
	Greeland Sea	Eyja fjordur	0.94±0.26	7	11.99±1.07	7	-0.18±0.15	7	1.90±1.04	8
		Reykjanes	1.45±0.58	14	10.41±1.87	14	0.59±0.66	14	NA	NA
	NorthNorway Svalbard	Lemmingvaer	NA	NA	NA	NA	NA	NA	8.31±9.05	3
	SouthNorwegian Sea	Faroes	NA	NA	NA	NA	NA	NA	2.98±2.93	23
Little auk	Barent Sea	Hooker Island	0.36±0.13	87	11.64±0.17	86	-1.53±0.32	86	0.81±0.21	69
	Greeland Sea	Kap Hoegh	0.84±0.17	88	11.76±0.23	88	-0.22±0.19	88	1.34±0.44	64
	NorthNorway Svalbard	Bjornoya	0.39±0.12	27	11.35±0.47	26	-1.04±0.37	26	0.82±0.19	31
		Hornsund	0.41±0.12	126	11.08±0.45	126	-0.92±0.30	126	0.82±0.35	37
		Isfjorden	0.48±0.08	14	11.76±0.26	14	-0.90±0.19	14	0.90±0.16	5
		Kongsfjorden	0.43±0.05	5	10.97±0.23	5	-0.82±0.10	5	NA	NA
	NorthwestAtlantic Arctic	Thule	0.63±0.14	19	12.36±0.17	19	-0.45±0.24	19	2.08±0.52	10
Northern fulmar	Greeland Sea	Breidafjordur	3.77±0.74	11	14.08±0.39	11	0.68±0.27	11	3.72±1.22	20
		Grimsey	3.34	1	13.50	1	0.73	1	NA	NA
		Holmahals	1.04	1	14.35	1	-0.65	1	NA	NA

		Iceland	2.65	1	13.41	1	0.52	1	NA	NA
		Jan Mayen	1.56±0.59	83	14.32±0.86	83	-0.30±0.36	83	2.72±1.20	60
		Langanes	2.64±1.04	80	14.14±0.50	80	0.25±0.44	80	2.84±1.19	56
		Papey	2.16±0.82	12	14.12±0.47	12	0.08±0.36	12	NA	NA
		Reykjanes	3.93±1.49	7	13.95±0.31	7	0.71±0.41	7	NA	NA
	NorthNorway Svalbard	Alkefjället	0.39±0.01	2	15.18±0.03	2	-1.85±0.04	2	1.37±1.08	6
		Bjornoya	0.90±0.32	61	15.84±0.40	61	-1.22±0.31	61	1.47±0.91	69
	NorthwestAtlantic Arctic	Baffin Island	1.05±0.52	24	14.51±0.79	17	-0.41±0.38	17	NA	NA
		Labrador Sea	1.31±0.89	31	13.67±0.65	31	-0.23±0.65	31	4.92±2.62	31
		Thule	2.69±2.29	10	15.31±0.27	9	0.03±0.87	9	2.69±2.29	10
	Pacific	Buldir Island	NA	NA	NA	NA	NA	NA	NA	NA
	SouthNorwegian Sea	Eynhallow	2.27±0.55	34	14.45±0.54	34	-0.65±0.28	34	3.22±1.60	76
		Faroes	NA	NA	NA	NA	NA	NA	3.04±1.13	28
Parakeet auklet	Pacific	Buldir Island	NA	NA	NA	NA	NA	NA	2.90±0.95	4
		Chowiet Island	NA	NA	NA	NA	NA	NA	NA	NA
		Talan Island	0.54±0.28	5	NA	NA	NA	NA	1.70±0.74	21
Razorbill	Barent Sea	Hornoya	1.57±0.23	19	13.90±0.24	19	-0.71±0.13	19	0.79±0.31	40
	NorthwestAtlantic SubArctic	Gannet Islands	2.38±0.41	11	14.72±0.20	11	-0.29±0.17	11	2.89±1.84	13
		St Pierre et Miquelon	3.07±0.52	19	NA	NA	NA	NA	2.62±1.19	15
	SouthNorwegian Sea	Isle of May	NA	NA	NA	NA	NA	NA	1.12±0.33	10
Red legged kittiwake	Pacific	Saint George	2.42±0.39	4	14.22±0.26	4	1.28±0.21	4	5.32±1.33	61
Rhinoceros auklet	Pacific	Middleton	1.51±0.38	20	14.43±0.12	20	0.71±0.26	20	3.04±1.62	28
		Saint Lazaria	NA	NA	NA	NA	NA	NA	1.96±0.81	14
		Talan Island	NA	NA	NA	NA	NA	NA	2.97±2.89	2
Tufted puffin	Pacific	Aiktak Island	NA	NA	NA	NA	NA	NA	3.12±1.42	11
		Buldir Island	NA	NA	NA	NA	NA	NA	3.62±1.26	3
		Talan Island	1.44±0.72	2	NA	NA	NA	NA	2.72±0.72	4

Papers

Paper 1

Using blood and feathers to investigate large-scale Hg contamination in Arctic seabirds: a review.

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Review article

Using blood and feathers to investigate large-scale Hg contamination in Arctic seabirds: A review

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ABSTRACT

Mercury (Hg), because of its deleterious effects on wildlife and its high concentrations in polar regions, has been widely studied in the Arctic. This provided important information regarding food web contamination, spatial and temporal trends of Hg in ecosystems or risk assessments for wildlife and Humans. Among the Arctic biota, seabirds have been among the most studied species due to their sensitivity to this toxicant, their role as bioindicators of the contamination status of their environment, and their consumption by Arctic communities. However, most studies that investigated Hg in Arctic seabirds focused on measurements in internal organs or in eggs, while few investigations have been performed on blood and feathers, despite the relevant and complementary information they provide. Here, we first provide a detailed overview of the specific information blood and feathers can bring when investigating Hg contamination of Arctic seabirds, including new knowledge on the poorly studied non-breeding period. Second, we perform a comprehensive review of the use of blood and feathers as non-lethal tissues to study Hg in Arctic seabirds. This review demonstrates important interspecific variations in Hg blood concentrations according to seabird trophic status, with seaducks generally presenting the lowest Hg concentrations while auks have the highest ones. However, all the observed Hg concentrations are below the admitted toxicity thresholds. Hg concentrations in feathers follow similar trends and gulls appear to be the most contaminated species, likely as a consequence of contrasting migratory and overwintering strategies. This review also confirms strong spatial variations with higher concentrations found in the Canadian Arctic and Pacific waters than in Greenland and the European Arctic. It also identifies some major understudied areas such as West Greenland, Aleutian Islands and Russia. Finally, we provide a thorough review of the current knowledge regarding molting patterns in Arctic seabirds, which is an essential information to interpret Hg concentrations measured in feathers. Overall, our results point out the importance of blood and feathers in seabird ecotoxicological assessments and highlight the need for large scale international collaborations and research programs.

1. Introduction

Mercury (Hg) is a metallic trace element naturally found in the environment, released by volcanic eruptions or weathering. However, human activities, such as coal burning or gold mining, have been responsible for a large increase in Hg emissions, especially since the 19th century (UNEP, 2013). Because of its solubility, its ability to enter and biomagnify along the food chains (as methylmercury – MeHg (Morel et al., 1998)), and its toxicity for human and wildlife (e.g., Tan et al., 2009; Wolfe et al., 1998), this pollutant requires close monitoring. More specifically, a major emphasis has been put towards the understanding of Hg levels in marine organisms, underlying modulators and resulting impacts on nervous systems, reproduction or development (Eagles-Smith et al., 2018). Anthropogenic Hg is mainly emitted by mid-

latitude northern countries, with still increasing emissions in some regions such as Asia (UNEP, 2013). It is then transported over large distances to polar regions by atmospheric and oceanic currents and, in the case of the Arctic, by rivers (Sonke et al., 2018; UNEP, 2013). Therefore, polar top predators are among the most exposed organisms to Hg (Cherel et al., 2018) and have thus been largely studied both in toxicity and conservation contexts, but also as efficient indicators to monitor Hg contamination in polar marine ecosystems (e.g., Carravieri et al., 2013; Dietz et al., 2013; Mallory et al., 2017; Sun et al., 2006; Verreault et al., 2010). Among those polar top predators, Arctic seabirds, being long-lived species with a circumpolar distribution, have been widely used to study long-term and relatively large-scale Hg exposure (see Becker, 2003). However, these studies mainly focused on the period during which seabirds are at their breeding site (hereafter the breeding

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season), even though this period represents a part of their annual cycle only. Conversely, investigations for the non-breeding period are still scarce, mainly because most of seabirds are offshore during this period, making their sampling logistically difficult. Previous studies suggested that Arctic seabirds could be exposed to higher Hg levels during the non-breeding period with subsequent impacts on their reproduction (e.g. Fort et al., 2014). Hence, improving our knowledge of bird contamination during the non-breeding period – which mostly occurs outside the Arctic – is crucial, first to determine its role on seabird exposure to Hg, but also to better understand the origin of this contamination, in space and time.

Several tissues were targeted within the various studies which investigated Hg in Arctic seabirds, providing information about seabird Hg contamination at different temporal scales. Indeed, once ingested through the diet (the main source of Hg intake), Hg enters the blood stream where it stays a few weeks/months (e.g. half-life of 30 days in great skua (*Stercorarius skua*) (Bearhop et al., 2000) and of 40–65 days in Cory's shearwaters (*Calonectric borealis*) (Monteiro and Furness, 2001)). Hg in blood thus informs about recent intakes (e.g. from late migration to chick-rearing when blood is sampled during the breeding season). Once Hg is assimilated, it is transported and distributed to the different organs, where it can be demethylated (i.e. protective mechanism against Hg toxicity) and/or stored (Dietz et al., 2013). While internal organs such as liver, brain, kidney and muscle give access to longer term Hg contamination compared to blood (Mallory et al., 2018), the liver also informs, depending on the species, about Hg demethylation abilities (Dehn et al., 2005; Kim et al., 1996b; Thompson and Furness, 1989). Finally, egg production and feather synthesis (molt) are two major excretion pathways for Hg. The study of eggs therefore informs about seabird Hg contamination during the laying time (and/or during the pre-breeding time in case of “capital” breeders (i.e. allocation of stored nutrients to egg formation) (Becker, 2003; Bond and Diamond, 2010; Mallory et al., 2017)). Cover feathers provide information about Hg contamination during a relatively longer period (over several weeks/months according to species and molting patterns (Furness et al., 1986, see below, Annex 1)). Because of toxicological risks for seabirds and consumers (seabirds and their eggs are consumed in several Arctic regions (Bard, 1999; Chan, 1998; Mallory and Braune, 2012)), internal tissues and eggs have received most of the attention (Figs. 1 and 2). Conversely, non-consumed tissues such as blood and feathers received much less attention, despite the relevant and complementary spatio-temporal information they could provide, and the fact that they can be non-lethally sampled. In the present study, we thus review and discuss the current knowledge of Hg contamination in

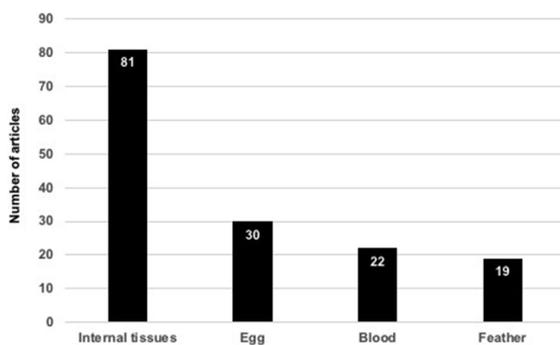


Fig. 1. Occurrence of tissues analysed for Hg in Arctic seabirds in the 101 reviewed articles. Some publications studied several tissues. Internal tissues comprise liver (n = 38), kidneys (n = 10), muscle (n = 25), heart (n = 1), bones (n = 1), brain (n = 4), gonads (n = 1) and lungs (n = 1). Feathers comprise body feathers (n = 14), head feathers (n = 3) and flight feathers (n = 4).

Arctic seabirds, with a specific focus on the information provided by blood and feather tissues. We will also highlight how these tissues and their combination with new approaches could help us improving knowledge of seabird yearly (breeding and non-breeding) Hg contamination.

2. Methods

2.1. Reviewing process

A review of the literature was done on Scopus searching for three keywords: “seabirds”, “Arctic” and “mercury” on December 2018. The Arctic boundaries used were as defined by the Conservation of Arctic Flora and Fauna (CAFF) working group (CAFF, 1996), and only the species breeding within these boundaries and spending most of the year feeding in oceanic food webs were included in the analysis (Annex 1). Blood and feather Hg concentrations were extracted and, when necessary, blood values were converted to dry weight using a moisture of 79.13% (Eagles-Smith et al., 2008) to allow comparison between published values. We considered feather concentrations provided in wet weight directly comparable to those in dry weight as moisture content is negligible in feathers (e.g. Stettenheim, 2000). A review of molting patterns for the retained species was also performed and is summarized in Annex 1.

2.2. Statistical analyses

The dataset does not provide sufficient information to test any temporal or spatial trend in Hg concentrations (underrepresentation of some families, species, countries or years). However, for both blood and feathers, we used ANOVA to compare Hg concentrations between families (see Figs. S1 and S2). More specifically, for blood, we used mean values provided by each study for each species and then compared Hg concentrations between families being adults anatids (n = 29) and larids (n = 19) and removed the alcids (n = 4) and the procellariids (n = 1) because of limited data (see Fig. S1). For feathers, we used mean values provided by each study for each species and then compared Hg concentrations between families being adults alcids (n = 31), anatids (n = 8) and larids (n = 19) and removed Hg concentrations for hydrobatids (n = 1) and stercorariids (n = 2) because of limited data (see Fig. S2). Statistical significance was assumed at $p < 0.05$ and analyses were run with R studio (version 1.2.1335).

3. Results and discussion

3.1. Hg assessment in blood and feathers of Arctic seabirds

The present review shows that 101 published articles (see Annex 2) reported Hg concentrations in Arctic seabirds (i.e. species/populations breeding in the Arctic) since 1984. In total, 42 species (list provided in Annex 1) from all around the Arctic were assessed, yet with an emphasis on the Canadian Arctic (n = 38 publications), Svalbard (n = 23), Aleutian Islands (n = 16), Alaska (n = 14) and West Greenland (n = 7). In addition, internal tissues and eggs were more investigated than blood and feathers (81, 30, 22 and 19 studies, respectively), also integrating a wider spatial scale (Figs. 1 and 2). Among studied species, three families received most of the attention: gulls (hereafter larids) (n = 56 publications), auks (hereafter alcids) (n = 49) and seabirds (hereafter anatids) (n = 44), covering all foraging strategies (benthic, pelagic, coastal, oceanic, divers, surface feeders, scavengers) and diets (insectivorous, molluscivorous, planktivorous, piscivorous, omnivorous).

Blood Hg concentrations were investigated in a total of 12 seabird species but anatids and larids were the most studied (see Table 1). For the Anatid family (n = 12 studies), the common eider (*Somateria mollissima*), the king eider (*Somateria spectabilis*), the Steller's eider (*Polyicta stelleri*), the spectacled eider (*Somateria fischeri*) and the long-

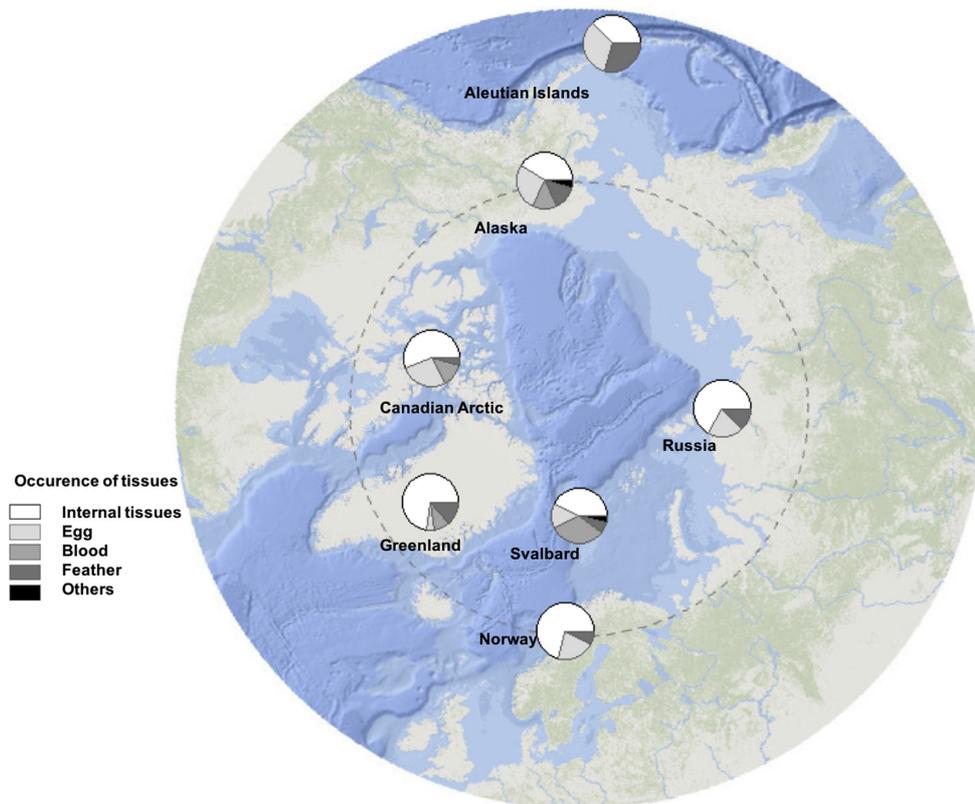


Fig. 2. Occurrence of tissues analysed for Hg in Arctic seabirds in the 101 reviewed articles, by Arctic region. Some publications studied several tissues. Internal tissues comprise liver (n = 38), kidneys (n = 10), muscle (n = 25), heart (n = 1), bones (n = 1), brain (n = 4), gonads (n = 1) and lungs (n = 1). Others (excrements, n = 2). Feathers comprise body feathers (n = 14), head feathers (n = 3) and flight feathers (n = 4).

tailed duck (*Clangula hyemalis*) were studied, covering Alaska, Svalbard and the Canadian Arctic. For the Larid family, investigations focused on the black-legged kittiwake (*Rissa tridactyla*) (n = 8 studies) covering both the Canadian Arctic and Svalbard, and the ivory gull (*Pagophila eburnea*) (n = 1 study) in Svalbard. Finally, four studies investigated Hg contamination of alcids breeding in the Canadian Arctic (Brünnich's guillemots (*Uria lomvia*) and black guillemots (*Cepphus grylle*)), East Greenland (little auks (*Alle alle*)) and Svalbard (Mandt's black guillemot (*Cepphus grylle mandtii*)), and one study investigated Hg contamination in procellariids (northern fulmars (*Fulmarus glacialis*)) from the Canadian Arctic.

Most studies which measured Hg concentrations in feathers focused on body (n = 18 publications) and flight (n = 4) feathers. They covered more areas than blood investigations and were mainly performed in the Aleutian Islands (n = 7 publications) and in Alaska (n = 3), followed by East Greenland (n = 3), Svalbard (n = 2), Russia (n = 2), the Canadian Arctic (n = 2) and Norway (n = 1). They encompassed alcids (n = 10), larids (n = 9), anatids (n = 8), procellariids (n = 2) and stercorariids (n = 1) (see Tables 2 and 3). There was a large heterogeneity between the different regions. In the Pacific Arctic, the Aleutian Islands are the only area where Hg contamination has been measured in feathers, targeting alcids (Kittlitz's murrelet (*Brachyramphus brevirostris*), pigeon guillemot (*Cepphus columba*), tufted puffin (*Fratercula cirrhata*)), anatids (common eider (*Somateria mollissima*)), hydrobatids (fork-tailed storm-petrel (*Oceanodroma furcata*)) and larids (black-legged kittiwakes (*Rissa tridactyla*), glaucous-winged gull (*Larus glaucescens*)). In the Atlantic Arctic, each country was studied, providing

information for alcids (Brünnich's guillemot, common guillemot (*Uria aalge*), little auk), anatids (king eider, common eider, long-tailed duck), larids (black-legged kittiwake, Arctic tern (*Sterna paradisae*), Sabine's gull (*Xema sabini*), ivory gull, glaucous gull (*Larus hyperboreus*), herring gull (*Larus argentatus*)), procellariids (northern fulmar) and stercorariids (parasitic jaeger (*Stercorarius parasiticus*), long-tailed jaeger (*Stercorarius longicaudus*)). The common eider and the black-legged kittiwake are the only two species studied in both the Atlantic and Pacific Arctic. In Russia, only one site (Chaun, Siberia) was studied with feather Hg measurements performed in anatids (long-tailed duck and king eider), larids (herring gull, Arctic tern, glaucous gull, Sabine's gull) and stercorariids (long-tailed and parasitic jaeger). Flight feathers were processed in three studies that only focused on alcids (Brünnich's guillemot, Kittlitz's murrelet), anatids (long-tailed-duck, common eider), larids (herring gull, Arctic tern, black-legged kittiwakes, ivory gull and glaucous gull) and procellariids (northern fulmar) (Table 3) (Kenney et al., 2018; Kim et al., 1996b; Mallory et al., 2015). Finally, three studies focused on the non-breeding period only, and all were performed on little auks breeding in East Greenland (Fort et al., 2014, 2016; Amélineau et al., 2019).

3.2. Blood as a tool to monitor Hg contamination during the breeding season

During the breeding season, seabirds aggregate in colonies where blood samples can be more easily collected. Sampling periods extend from the pre-laying to the late chick-rearing stages, allowing investigations on adult seabird short-term Hg contamination (i.e. from

Table 1
Total Hg concentrations measured in blood samples (whole blood $\mu\text{g/g}$ dw mean \pm SD or SE; red blood cell $\mu\text{g/g}$ dw mean \pm SD; sample size is given into bracket) for seabirds breeding in the Arctic. If sexes were differentiated, F for female or M for male is added. * wet weight converted to dry weight following a moisture content of 79.13% (Eagles-Smith et al., 2008), a mmol/g to $\mu\text{g/g}$, b overall sample size = 105.

Family	Species	Sampling year	Colony location	Country	Whole blood	Red blood cell	References	
Alcid	Mand's black guillemot (<i>Cepphus grylle mandtii</i>)	2015	Kongsfjorden	Svalbard		0.32 \pm 0.80 (10)	Echbo et al. (2019)	
	Black guillemot (<i>Cepphus grylle</i>)	NA	Prince Leopold Island	Canada	4.58 (1)		Mallory et al. (2018)	
	Brünnich guillemot (<i>Uria lomvia</i>)	NA	Prince Leopold Island	Canada	2.88 \pm 0.50 (SD) (6)		Mallory et al. (2018)	
	Little auk (<i>Alte alle</i>)	2010	Kap Hoegh	Greenland	0.84 \pm 0.20 (SD) (35)		Fort et al. (2014)	
	Common eider (<i>Somateria mollissima</i>)	2014	Kaktovik	Alaska	1.13 \pm 0.04 (SD) F (20)		Miller et al. (2019)	
	Common eider (<i>Somateria mollissima</i>)	NA	Cape Dorset	Canada	1.22 \pm 0.33 (SD) (9)		Mallory et al. (2018)	
	Common eider (<i>Somateria mollissima</i>)	2011	Storholmen, Kongsfjorden	Svalbard	0.57 \pm 0.14 (SD) (26) F a; 0.30–0.94 (range) F (29) *		Fenstad et al. (2017), (2016)	
	Common eider (<i>Somateria mollissima</i>)	2013	East bay	Canada	2.06 \pm 0.29 (SE) (112) F *		Provancher et al. (2017)	
	Common eider (<i>Somateria mollissima</i>)	2014	East bay	Canada	2.25 \pm 0.29 (SE) (109) F *		Provancher et al. (2017)	
	Common eider (<i>Somateria mollissima</i>)	2013	East bay	Canada	0.91 \pm 0.24 (SD) (98) F *		Provancher et al. (2016)	
	Common eider (<i>Somateria mollissima</i>)	2014	East bay	Canada	1.05 \pm 0.29 (SD) (92) F *		Provancher et al. (2016)	
	Common eider (<i>Somateria mollissima</i>)	2000	Western Prudhoe Bay Oil Field	Alaska	0.87 \pm 0.04 (SE) (20) F		Franson et al. (2004)	
	Common eider (<i>Somateria mollissima</i>)	2000	Eastern Prudhoe Bay Oil Field	Alaska	0.73 \pm 0.03 (SE) (20) F		Franson et al. (2004)	
	Common eider (<i>Somateria mollissima</i>)	1997	East bay	Canada	1.10 \pm 0.05 (SE) (11) F *		Wayland et al. (2001)	
	Common eider (<i>Somateria mollissima</i>)	1998	East bay	Canada	1.10 \pm 0.10 (SE) (15) F *		Wayland et al. (2001)	
	King eider (<i>Somateria spectabilis</i>)	2010–2014	Utqiagvik	Alaska	0.64 \pm 0.03 (SD) F (28)		Miller et al. (2019)	
	King eider (<i>Somateria spectabilis</i>)	NA	Cape Dorset	Canada	2.04 \pm 0.76 (SD) (6)		Mallory et al. (2018)	
	Larid	King eider (<i>Somateria spectabilis</i>)	2001–2003	Karrak lake	Canada	0.81 \pm 0.24 (SD) (147) F *		Wayland et al. (2008a, 2008b)
		King eider (<i>Somateria spectabilis</i>)	2001	Karrak lake & Adventure lake	Canada	0.65 (0.60–0.70) (SE) (63) F *		Wayland et al. (2008a, 2008b, 2007)
King eider (<i>Somateria spectabilis</i>)		2002	Karrak lake & Adventure lake	Canada	0.90 (0.85–0.95) (SE) (69) F *		Wayland et al. (2008a, 2008b, 2007)	
King eider (<i>Somateria spectabilis</i>)		2003	Karrak lake & Adventure lake	Canada	0.85 (0.80–0.90) (SE) (74) F *		Wayland et al. (2008a, 2008b, 2007)	
King eider (<i>Somateria spectabilis</i>)		1996 (prebreeding)	Prudhoe Bay Oil Field	Alaska	1.44 \pm 0.19 (SE) (7) M *		Wilson et al. (2004)	
King eider (<i>Somateria spectabilis</i>)		1996 (prebreeding)	Prudhoe Bay Oil Field	Alaska	1.05 \pm 0.09 (SE) (4) F *		Wilson et al. (2004)	
King eider (<i>Somateria spectabilis</i>)		1996 (nesting)	Prudhoe Bay Oil Field	Alaska	1.48 \pm 0.09 (SE) (4) F *		Wilson et al. (2004)	
Long-tailed duck (<i>Clangula hyemalis</i>)		2010–2012, 2014	Utqiagvik	Alaska	0.95 \pm 0.18 (SD) F (15)		Miller et al. (2019)	
Long-tailed duck (<i>Clangula hyemalis</i>)		2000	Western Prudhoe Bay Oil Field	Alaska	0.67 \pm 0.06 (SE) (20) M		Franson et al. (2004)	
Long-tailed duck (<i>Clangula hyemalis</i>)		2000	Eastern Prudhoe Bay Oil Field	Alaska	0.57 \pm 0.04 (SE) (20) M		Franson et al. (2004)	
Spectacled eiders (<i>Somateria fischeri</i>)		2010–2014	Utqiagvik	Alaska	0.49 \pm 0.08 (SD) (42) F		Miller et al. (2019)	
Spectacled eiders (<i>Somateria fischeri</i>)		1995 (prebreeding)	Prudhoe Bay Oil Field	Alaska	0.96 \pm 0.05 (SE) (14) M *		Wilson et al. (2004)	
Spectacled eiders (<i>Somateria fischeri</i>)		1995 (prebreeding)	Prudhoe Bay Oil Field	Alaska	0.57 \pm 0.09 (SE) (6) F *		Wilson et al. (2004)	
Spectacled eiders (<i>Somateria fischeri</i>)		1995 (nesting)	Prudhoe Bay Oil Field	Alaska	0.72 \pm 0.09 (SE) (9) F *		Wilson et al. (2004)	
Spectacled eiders (<i>Somateria fischeri</i>)		2008; 2010–2012; 2014	Utqiagvik	Alaska	1.05 \pm 0.14 (SE) (10) F *		Wilson et al. (2019)	
Steller's eider (<i>Polysticta stelleri</i>)		NA	Prince Leopold Island	Canada	1.36 \pm 0.19 (SD) (36) F		Miller et al. (2019)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)		2015	Kongsfjorden	Svalbard	3.04 \pm 1.41 (SD) (9)		Mallory et al. (2018)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)		2015	Kongsfjorden	Svalbard	2.00 \pm 0.59 (SD) (20) M		Blévin et al. (2018)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)		2015	Kongsfjorden	Svalbard	1.43 \pm 0.38 (SD) (20) F		Blévin et al. (2018)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)		2012	Kongsfjorden	Svalbard	1.14 \pm 0.07 (SE) (22) M	0.89 \pm 0.05 (SE) (22) F		Blévin et al. (2017)
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2012	Kongsfjorden	Svalbard	0.90 \pm 0.25 (SD) (48) F	1.14 \pm 0.07 (SE) (22) M		Blévin et al. (2017)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2012 (chick rearing)	Kongsfjorden	Svalbard	1.25 \pm 0.33 (SD) (44) M	0.90 \pm 0.25 (SD) (48) F		Tartu et al. (2016)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>) ADULT	2012 (incubation)	Kongsfjorden	Svalbard	1.13 \pm 0.32 (SD) (44) M	1.25 \pm 0.33 (SD) (48) F		Tartu et al. (2016)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>) ADULT	2012 (chick rearing)	Kongsfjorden	Svalbard	1.82 \pm 0.46 (SD) (44) M	1.13 \pm 0.32 (SD) (44) M		Tartu et al. (2016)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>) ADULT	2012	Kongsfjorden	Svalbard	NA	1.82 \pm 0.46 (SD) (44) M		Tartu et al. (2016)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>) ADULT	2013	Kongsfjorden	Svalbard	NA	NA		Tartu et al. (2016)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2012	Kongsfjorden	Svalbard	1.80 \pm 0.45 (SD) (34) M	NA		Tartu et al. (2016)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2008	Kongsfjorden	Svalbard	2.06 \pm 0.44 (SD) M b	1.80 \pm 0.45 (SD) (34) M		Tartu et al. (2015)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2009	Kongsfjorden	Svalbard	2.33 \pm 0.55 (SD) M b	NA		Goutte et al. (2015)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2008	Kongsfjorden	Svalbard	1.97 \pm 0.44 (SD) F b	NA		Goutte et al. (2015)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2009	Kongsfjorden	Svalbard	2.01 \pm 0.41 (SD) F b	NA		Goutte et al. (2015)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2008	Kongsfjorden	Svalbard	NA	NA		Tartu et al. (2013)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2011	Kongsfjorden	Svalbard	1.68 \pm 0.27 (SD) (6)	0.60–3.30		Tartu et al. (2013)	
Ivory gull (<i>Pegophila eburnea</i>)	April 2011	Barentsøya	Svalbard				Lucia et al. (2016)	

(Continued on next page)

Table 1 (continued)

Family	Species	Sampling year	Colony location	Country	Whole blood	Red blood cell	References
Procellariid	Ivory gull (<i>Pegophila eburnea</i>)	April 2012	Barentsøya	Svalbard	0.81 ± 0.05 (SD) (4)		Lucia et al. (2016)
	Ivory gull (<i>Pegophila eburnea</i>)	April 2013	Barentsøya	Svalbard	1.06 ± 0.16 (SD) (5)	1.39 ± 0.16 (SD) (40)	Lucia et al. (2016)
	Ivory gull (<i>Pegophila eburnea</i>)	April 2014	Barentsøya	Svalbard	3.62 ± 2.06 (SD) (7)	1.20 ± 0.07 (SD) (42)	Lucia et al. (2016)
	Northern fulmar (<i>Fulmarus glacialis</i>)	NA	Prince Leopold Island	Canada			Mallory et al. (2018)

late migration to chick-rearing (Bearhop et al., 2000; Evers et al., 2008; Monteiro and Furness, 1995)). Hg analyses are done on whole blood or on blood cells (concentrations in these two matrices being highly correlated (Sire et al. unpublished)). Blood is also a good proxy for Hg concentrations in other internal tissues. Indeed, Hg concentrations in blood have been found to be strongly correlated with Hg concentrations in brain and muscle (e.g., Fort et al., 2015; Mallory et al., 2018). In addition, as a non-lethally sampled tissue, blood has several advantages. First, individuals can be sampled every year for long-term Hg monitoring at the individual and population scales or several times along the breeding season. This provides specific information about Hg dynamics along the pre-laying, incubation and chick-rearing periods (see Lavoie et al., 2014). Second, a large number of individuals and species can be sampled at a broad scale, allowing spatial comparisons of Hg concentrations and thus large-scale monitoring. For instance, Ackerman et al. (2016) reviewed bird Hg contamination across the Northwestern America to map the distribution of Hg concentrations and highlighted hotspots of Hg contamination.

Our results show that anatids had lower Hg concentrations in blood than larids ($F_{1,46} = 14.36, p < 0.001$). For anatids, Hg concentrations ranged from $0.57 \pm 0.04 \mu\text{g/g dw}$ (long-tailed duck, Franson et al., 2004) to $2.25 \pm 0.29 \mu\text{g/g dw}$ (common eider, Provencher et al., 2017) (Table 1, Figs. 3 and 4) with an average value of $1.04 \pm 0.33 \mu\text{g/g dw}$. For larids, concentrations ranged from $0.81 \pm 0.05 \mu\text{g/g dw}$ (ivory gull, Lucia et al., 2016) to $3.04 \pm 1.41 \mu\text{g/g dw}$ (black-legged kittiwake, Mallory et al., 2018) (Table 1, Figs. 3 and 4) with an average value of $1.57 \pm 0.48 \mu\text{g/g dw}$. Finally, Hg concentrations in alcid blood samples ranged from $0.32 \pm 0.80 \mu\text{g/g dw}$ to $4.58 \mu\text{g/g dw}$, both measured in black guillemots (Eckbo et al., 2019 and Mallory et al., 2018, respectively), with an average value for this family of $2.16 \pm 1.58 \mu\text{g/g dw}$. Overall, these data show a high inter-species variability (see Table 1, Figs. 3 and 4). Even if alcids have the highest mean blood Hg concentration, too few values ($n = 4$) were reported to confirm that they are significantly more contaminated than the two other families. In addition, such variations in blood Hg concentrations among taxa are in accordance with the feeding ecology of these species. Indeed, anatids feed at low trophic levels (i.e. mostly on molluscs and crustaceans) during the breeding season. Guillemots feed at intermediate levels (mostly on planktivorous fish species) (Gaston and Jones, 1998) while larids occupy the highest trophic levels (see Braune et al., 2016; Burgess et al., 2013; Hobson et al., 2002, 1994 for a review) (Fig. 4). It should also be noted that all families reviewed in this study showed blood Hg concentrations below toxicity thresholds associated with substantial impairments to health and reproduction ($2 \mu\text{g/g ww}$, equivalent to approximately $9.6 \mu\text{g/g dw}$; Ackerman et al., 2016). These results also highlight spatial variations through the Arctic, with higher levels found in the Canadian Arctic and Alaska than in East Greenland or Svalbard, and thus confirms previous investigations using eggs which found higher values in the Canadian Arctic (Arctic Monitoring and Assessment Programme (AMAP), 2018; Provencher et al., 2014). They are also in agreement with previous investigations performed on internal tissues where higher Hg concentrations were found in the liver of Canadian Arctic seabirds (Northern Baffin Bay) compared to those from the European Arctic (Central Barents Sea) (Borgå et al., 2006). Although more analyses are needed to confirm this spatial trend by including various missing Arctic regions (e.g. Russia, Northwest Greenland, Aleutian Islands, Alaska), these observations provide additional elements towards the understanding of Hg spatial distribution in the Arctic.

Finally, Braune et al. (2007) adjusted Hg concentrations by trophic status in seabird eggs ($\delta^{15}\text{N}$) to take into account any change in their trophic position and, hence, to study temporal changes in Hg contamination. This method was subsequently used on different species and sites (Braune et al., 2016; Burgess et al., 2013). Blood, as eggs, represents relatively local (in the vicinity of the colony) and short-term dietary exposure. Blood Hg concentrations strongly depend on seabird

Table 2
Total Hg concentrations measured in body feathers (BF) (mean ± SD or SE, µg/g dw; sample size is given into bracket) for seabirds breeding in the Arctic. If sexes were differentiated, F for female or M for male is added.
a: ppb to µg/g; b: fw.

Family	Species	Sampling year	Colony location	Country	Body feathers	References
Alcid	Brünnich guillemot (<i>Uria lomvia</i>)	1992 & 1993	Hornøya	Norway	0.78 ± 0.18 (SD) (14)	Wenzel and Gabrielsen (1995)
	Common guillemot (<i>Uria adae</i>)	1992 & 1993	Hornøya	Norway	0.88 ± 0.19 (SD) (10)	Wenzel and Gabrielsen (1995)
	Kittlitz's murrelet (<i>Brachyramphus brevirostris</i>) ADULT	August 2008–2011	Agattu	Aleutian Islands	1.66 ± 1.28 (SE) (29) a, b	Kaler et al. (2014)
	Kittlitz's murrelet (<i>Brachyramphus brevirostris</i>) CHICK	August 2008–2011	Agattu	Aleutian Islands	1.47 ± 0.67 (SE) (8) a, b	Kaler et al. (2014)
	Kittlitz's murrelet (<i>Brachyramphus brevirostris</i>)	2008–2011	Agattu	Aleutian Islands	2.06 ± 1.28 (SD) (37)	Kenney et al. (2018)
	Kittlitz's murrelet (<i>Brachyramphus brevirostris</i>)	2011	Adak Island	Aleutian Islands	5.15 ± 2.51 (SD) (7)	Kenney et al. (2018)
	Little auk (<i>Alle alle</i>)	2007	Kap Högeh	Greenland	1.00 ± 0.22 (SD) (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2008	Kap Högeh	Greenland	1.13 ± 0.24 (SD) (19)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2009	Kap Högeh	Greenland	1.04 ± 0.26 (SD) (19)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2010	Kap Högeh	Greenland	1.39 ± 0.73 (SD) (49)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2011	Kap Högeh	Greenland	1.84 ± 0.85 (SD) (18)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2012	Kap Högeh	Greenland	1.25 ± 0.51 (SD) (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2013	Kap Högeh	Greenland	1.69 ± 0.80 (SD) (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2014	Kap Högeh	Greenland	1.15 ± 0.33 (SD) (19)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2015	Kap Högeh	Greenland	1.25 ± 0.53 (SD) (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2016	Kap Högeh	Greenland	1.60 ± 0.62 (SD) (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2007	Kap Högeh	Greenland	1.00 ± 0.22 (SD) (20)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2008	Kap Högeh	Greenland	1.13 ± 0.24 (SD) (19)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2009	Kap Högeh	Greenland	1.04 ± 0.26 (SD) (19)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2010	Kap Högeh	Greenland	1.37 ± 0.70 (SD) (45)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2011	Kap Högeh	Greenland	1.70 ± 0.66 (SD) (17)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2012	Kap Högeh	Greenland	1.25 ± 0.51 (SD) (20)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2013	Kap Högeh	Greenland	1.69 ± 0.80 (SD) (20)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2014	Kap Högeh	Greenland	2.11 ± 1.49 (SD) (20)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2010–2011	Kap Högeh	Greenland	1.53 ± 0.84 (SD) (78)	Fort et al. (2014)
	Pigeon guillemot (<i>Cephus columba</i>)	July 2004	Aleutian Islands	Aleutian Islands	7.11 ± 0.66 (SE) (38) a	Burger et al. (2009)
Pigeon guillemot (<i>Cephus columba</i>)	June 2004	Anchitka Island	Aleutian Islands	7.72 ± 0.84 (SE) (21) a	Burger et al. (2007)	
Pigeon guillemot (<i>Cephus columba</i>)	June 2004	Kiska Island	Aleutian Islands	6.36 ± 1.03 (SE) (17) a	Burger et al. (2007)	
Pigeon guillemot (<i>Cephus columba</i>)	2004	Prince William Sound	Alaska	2.81 ± 0.25 (SE) (40) a	Burger et al. (2007)	
Tufted puffin (<i>Fratercula cirrhata</i>)	July 2004	Aleutian Islands	Aleutian Islands	2.54 ± 0.19 (SE) (39) a	Burger et al. (2009)	
Tufted puffin (<i>Fratercula cirrhata</i>)	July 2004	Anchitka Island	Aleutian Islands	2.59 ± 0.29 (SE) (22)	Burger et al. (2009)	
Tufted puffin (<i>Fratercula cirrhata</i>)	July 2004	Kiska Island	Aleutian Islands	2.48 ± 0.25 (SE) (17)	Burger et al. (2009)	
Common eider (<i>Somateria mollissima</i>)	July 2007	Anchitka Island	Aleutian Islands	0.89 ± 0.16 (SE) (22) F a	Burger et al. (2008b)	
Common eider (<i>Somateria mollissima</i>)	July 2007	Kiska Island	Aleutian Islands	1.24 ± 0.32 (SE) (8) F a	Burger et al. (2008a)	
Common eider (<i>Somateria mollissima</i>)	July 2004	Aleutian Islands	Aleutian Islands	0.84 ± 0.08 (SE) (26) a	Burger et al. (2009)	
Common eider (<i>Somateria mollissima</i>)	July 2004	Anchitka Island	Aleutian Islands	0.85 ± 0.14 (SE) (13)	Burger et al. (2009)	
Common eider (<i>Somateria mollissima</i>)	July 2004	Kiska Island	Aleutian Islands	0.83 ± 0.08 (SE) (13)	Burger et al. (2009)	
Common eider (<i>Somateria spectabilis</i>)	2011	Storholmen, Kongsfjorden	Svalbard	0.50–1.90 µg/g ww (29) F	Fenstad et al. (2017)	
King eider (<i>Somateria spectabilis</i>)	2014	Urqiagyik	Alaska	0.80 ± 0.37 (SD) (28) F	Miller et al. (2019)	
King eider (<i>Somateria spectabilis</i>)	Jun, July 1993	Chaun	Russia	0.57 (1)	Kim et al. (1996a, 1996b)	
Long-tailed duck (<i>Clangula hyemalis</i>)	Jun, July 1993	Chaun	Russia	1.96 ± 3.31 (SD) (8)	Kim et al. (1996a, 1996b)	
Spectacled eider (<i>Somateria fischeri</i>)	2010–2014	Urqiagyik	Alaska	1.35 ± 1.00 (SD) (42) F	Miller et al. (2019)	
Fork-tailed storm-petrel (<i>Oceanodroma furcata</i>)	August 2008–2011	Agattu	Aleutian Islands	6.70 ± 1.63 (SE) (12) a, b	Kaler et al. (2014)	
Arctic tern (<i>Sterna paradisica</i>)	Jun, July 1993	Chaun	Russia	0.89 ± 0.15 (SD) (10)	Kim et al. (1996a, 1996b)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>)	2004	Prince William Sound	Alaska	2.91 ± 0.19 (SE) (61) a	Burger et al. (2008b)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>) ADULT	1992 & 1993	Hornøya	Norway	2.03 ± 0.40 (SD) µg g ⁻¹ ww (27)	Wenzel and Gabrielsen (1995)	
Black-legged kittiwakes (<i>Rissa tridactyla</i>) FLEDGLING	1992 & 1993	Hornøya	Norway	0.55 ± 0.10 (SD) µg g ⁻¹ ww (10)	Wenzel and Gabrielsen (1995)	
Glaucous gull (<i>Larus hyperboreus</i>)	Jun, July 1993	Chaun	Russia	5.96 (1)	Kim et al. (1996a, 1996b)	
Glaucous-winged gulls (<i>Larus glaucescens</i>)	July 2004	Aleutian Islands	Aleutian Islands	3.68 ± 0.37 (SE) (63) a	Burger et al. (2009)	
Glaucous-winged gulls (<i>Larus glaucescens</i>) ADULT	NA	Adak Island	Aleutian Islands	2.97 ± 0.57 (SE) (16) a	Burger et al. (2009)	
Glaucous-winged gulls (<i>Larus glaucescens</i>) ADULT	NA	Anchitka Island	Aleutian Islands	4.01 ± 0.54 (SE) (30) a	Burger et al. (2009)	
Glaucous-winged gulls (<i>Larus glaucescens</i>) ADULT	NA	Kiska Island	Aleutian Islands	3.76 ± 0.86 (SE) (17) a	Burger et al. (2009)	
Glaucous-winged gulls (<i>Larus glaucescens</i>) CHICKS	NA	Anchitka Island	Aleutian Islands	2.19 ± 0.26 (SE) (21) a	Burger et al. (2009)	

(continued on next page)

Table 2 (continued)

Family	Species	Sampling year	Colony location	Country	Body feathers	References
Stercorariid	Glaucous-winged gulls (<i>Larus glaucescens</i>) CHICKS	NA	Kiska Island	Aleutian Islands	1.68 ± 0.23 (SE) (15) a	Burger et al. (2009)
	Herring gull (<i>Larus argentatus</i>)	June, 1993	Chaun	Russia	6.10 ± 4.60 (SD) (5)	Kim et al. (1996a, 1996b)
	Herring gull (<i>Larus argentatus</i>)	Jun, July 1993	Chaun	Russia	6.06 ± 4.60 (SD) (6)	Kim et al. (1996a, 1996b)
	Ivory gull (<i>Pagophila eburnea</i>)	April 2011	Barentsøya	Svalbard	2.65 ± 0.42 (SD) (19)	Lucia et al. (2016)
	Ivory gull (<i>Pagophila eburnea</i>)	April 2012	Barentsøya	Svalbard	3.03 ± 0.41 (SD) (40)	Lucia et al. (2016)
	Ivory gull (<i>Pagophila eburnea</i>)	April 2013	Barentsøya	Svalbard	2.79 ± 0.24 (SD) (47)	Lucia et al. (2016)
	Ivory gull (<i>Pagophila eburnea</i>)	April 2014	Barentsøya	Svalbard	3.39 ± 0.30 (SD) (39)	Lucia et al. (2016)
	Ivory gull (<i>Pagophila eburnea</i>)	2010	Seymour Island	Canada	11.66 ± 6.52 (SD) (8)	Mallory et al. (2015)
	Sabine's gull (<i>Xema sabini</i>)	Jun, July 1993	Chaun	Russia	1.70 ± 0.47 (SD) (2)	Kim et al. (1996a, 1996b)
	Long-tailed jaeger (<i>Stercorarius longicaudus</i>)	Jun, July 1993	Chaun	Russia	1.95 ± 0.54 (SD) (5)	Kim et al. (1996a, 1996b)
Parasitic jaeger (<i>Stercorarius parasiticus</i>)	Jun, July 1993	Chaun	Russia	1.84 (1)	Kim et al. (1996a, 1996b)	

diet and trophic position (i.e. biomagnification process), which can vary between individuals, populations and species. Hence, and similarly to Braune and colleagues' method, we propose that the adjusted Hg concentrations in blood could be used to monitor Hg temporal and spatial trends not only in seabirds but also in the Arctic marine food chains.

3.3. Feathers as a tool to monitor Hg contamination at different seasons

3.3.1. Hg excretion to feathers, molting patterns and feathers to monitor Hg contamination through the Arctic

Approximately 70–90% of the Hg available in the bloodstream and internal tissues is excreted into the plumage during molt (Agusa et al., 2005; Braune, 1987; Honda et al., 1986). Hg binds to disulphide bridges of feather keratins (Crewther et al., 1965) and becomes stable once the feather is grown (Appelquist et al., 1984). Hence, Hg concentrations in feathers represent the Hg accumulated between two molts and inform about the Hg body burden at the molting time (Agusa et al., 2005; Furness et al., 1986). As a consequence, Hg concentrations in feathers are not affected by the age of the individual (Honda et al., 1986; Bustamante et al., 2016). As they can be non-lethally sampled, feathers have also been proposed as a good proxy to study seabird exposure to Hg (Monteiro and Furness, 1995). The Hg excreted into feathers is mainly under its organic and toxic MeHg form (> 80%) (Bond and Diamond, 2009; Renedo et al., 2017; Thompson and Furness, 1989). This means that measuring total Hg concentrations in seabird feather samples provides a good information about their contamination and exposure to MeHg. Similar to blood, feathers can be collected every year on the same individuals (Bearhop et al., 2000), hence allowing long-term assessment of individual Hg exposure. In addition, depending on the species, molting patterns and the type of feather considered, feathers can inform about Hg exposure during different periods of the year (Lavoie et al., 2014; Fort et al., 2014). Some Arctic seabirds molt once a year only, i.e. post or pre-breeding molt, as is the case for female anatids or ivory gulls, respectively. Some other species (most of the alcids, larids, and male anatids) molt twice a year (see the complete review of molting patterns in Annex 1). As an example, alcids have a first partial molt occurring before the breeding period and leading to a nuptial plumage, when only head and neck feathers are replaced. A second complete molt occurs after the breeding period during which all feathers, including body, head and flight feathers, are renewed. Conversely, the northern gannet (*Morus bassanus*) is the only species breeding in the Arctic that presents a continuous molt of body feathers (see Annex 1), which leads to a more complex interpretation of the feather Hg concentrations (Cherel et al., 2018).

Three different types of feather are commonly used in Hg assessments: flight (primary and secondary) feathers, body feathers and head feathers. Head feathers of most alcid and larid species (Annex 1) provide information about Hg contamination of individual seabirds specifically during the non-breeding period (Fort et al., 2014, Fig. 5). However, their use has been largely neglected to date. Primary and body feathers inform about seabird Hg contamination either over the breeding season for species molting twice a year or over a full year for those molting once a year (Fig. 5). Nevertheless, primary feathers show a much higher inter-feather variability of Hg concentrations compared to body feathers. Feathers are sequentially molted from primary 1 (P1) to primary 10 (P10), with higher Hg quantities excreted into P1 and then decreasing from P2 to P10 (Furness et al., 1986) (Fig. 5). Conversely, body feathers are molted at the same period and show lower variability (Brasso et al., 2013; Carravieri et al., 2014; Furness et al., 1986). Therefore, if primary feathers are chosen to investigate seabird exposure to Hg, they must be carefully selected in order to make Hg measurements comparable between the different individuals. In addition, some species also have a sequential molt of their body and flight feathers over several months or an unknown molting pattern. The use of these species for feather Hg assessment is thus more complicated. This

Table 3

Total Hg concentrations (mean \pm SD $\mu\text{g/g}$ dw; sample size is given into bracket) measured in flight feathers (a: primary or b: secondary) for seabirds breeding in the Arctic.

Family	Species	Sampling year	Colony location	Country	Flight feathers	References
Alcid	Brünnich's guillemot (<i>Uria lomvia</i>)	NA	Prince Leopold Island	Canada	1.94 \pm 0.63 (10) a	Mallory et al. (2015)
	Kittlitz's murrelet (<i>Brachyramphus brevirostris</i>)	2011	Adak Island	Aleutian Islands	37.18 (1) b	Kenney et al. (2018)
Anatid	Long-tailed duck (<i>Clangula hyemalis</i>)	June 1993	Chaun	Russia	0.70 \pm 0.20 (5) a	Kim et al., 1996a, 1996b
	Common eider (<i>Somateria mollissima</i>)	NA	Cape Dorset, Nunavut	Canada	0.59 \pm 0.21 (10) a	Mallory et al. (2015)
Larid	Herring gull (<i>Larus argentatus</i>)	June 1993	Chaun	Russia	6.10 \pm 4.60 (5) a	Kim et al., 1996a, 1996b
	Arctic tern (<i>Sterna paradisae</i>)	June 1993	Chaun	Russia	0.90 \pm 0.10 (5) a	Kim et al., 1996a, 1996b
	Black-legged kittiwakes (<i>Rissa tridactyla</i>)	NA	Prince Leopold Island	Canada	3.58 \pm 0.92 (2) a	Mallory et al. (2015)
	Ivory gull (<i>Pagophila eburnea</i>)	NA	Seymour Island	Canada	15.79 \pm 14.13 (8) a	Mallory et al. (2015)
	Glaucous gull (<i>Larus hyperboreus</i>)	NA	Nasaruvaalik Island	Canada	2.31 \pm 1.68 (4) a	Mallory et al. (2015)
Procellariid	Northern fulmar (<i>Fulmarus glacialis</i>)	NA	Prince Leopold Island	Canada	2.71 \pm 0.72 (10) a	Mallory et al. (2015)

is the case of the Atlantic puffin (*Fratercula arctica*), the whiskered auklet (*Aethia pygmaea*), the Leach's storm-petrel (*Oceanodroma leucorhoa*) and the Fork-tailed storm-petrel (*Oceanodroma furcata*), the pelagic cormorant (*Stercorarius longicaudus*) and the red-faced cormorant (*Stercorarius pomarinus*) (Gaston and Jones, 1998; Harris, 1974; Pyle, 2016; Tootchin and Fenneman, 2014; Van Tets, 1959). For instance, the Atlantic puffin, as most of the alcids, undergoes two molts per year but this species presents unsynchronized post-breeding molts between individuals (Gaston and Jones, 1998; Harris and Yule, 1977).

3.3.2. Hg contamination of Arctic seabirds during the breeding season or through the entire year

At the Arctic scale, Hg concentrations measured in body feathers significantly differed between bird families (ANOVA, $F_{2,55} = 10.19$, $p < 0.001$), with the lowest mean Hg concentrations measured in anatids ($0.92 \pm 0.19 \mu\text{g/g}$ dw), followed by alcids ($2.20 \pm 1.23 \mu\text{g/g}$ dw) and larids ($3.53 \pm 1.74 \mu\text{g/g}$ dw).

More specifically, while Hg concentrations measured in female anatids (for which body feathers represent yearly local exposure) did not exceed $2 \mu\text{g/g}$ dw and showed little intra- and interspecific variations, alcids (for which body feathers represent contamination during breeding period only), reached concentrations of $7 \mu\text{g/g}$ dw and showed

higher interspecific variations (See Table 2, Figs. 6 and 7). As observed for blood, these results are consistent with the knowledge of species' diet and trophic position. For instance, little auks (1.00 ± 0.22 to $2.11 \pm 1.49 \mu\text{g/g}$ dw; Fort et al., 2016; Amélineau et al., 2019) feed mostly on zooplankton, while tufted puffins (2.48 ± 0.25 to $2.59 \pm 0.29 \mu\text{g/g}$ dw; Burger and Gochfeld, 2009) and Kittlitz's murrelets (from 1.66 ± 1.28 (Kaler et al., 2014) to $5.15 \pm 2.51 \mu\text{g/g}$ dw (Kenney et al., 2018)) feed on a mixed diet of fish and invertebrates (Gaston and Jones, 1998; Hobson et al., 1994). However, these differences might also be explained by the spatial variations of Hg contamination of their prey. For instance, common ($0.88 \pm 0.19 \mu\text{g/g}$ dw; Wenzel and Gabrielsen, 1995), Brünnich's ($0.78 \pm 0.18 \mu\text{g/g}$ dw; Wenzel and Gabrielsen, 1995), and pigeon guillemots ($7.72 \pm 0.84 \mu\text{g/g}$ dw; Burger and Gochfeld, 2009) are all mostly piscivorous (Gaston, 2000; Hobson et al., 1994) species during the breeding season. However, Hg concentrations measured in pigeon guillemots and Kittlitz's murrelets were about six and ten times higher than in common and Brünnich's guillemots, reflecting a hotspot of Hg contamination in the Aleutian Islands (Ackerman et al., 2016), where tufted puffins were also sampled. Those Hg concentrations in feathers are in accordance with previous works showing that pigeon guillemots breeding in the Pacific Ocean (Western North America) are facing potential toxicological risk

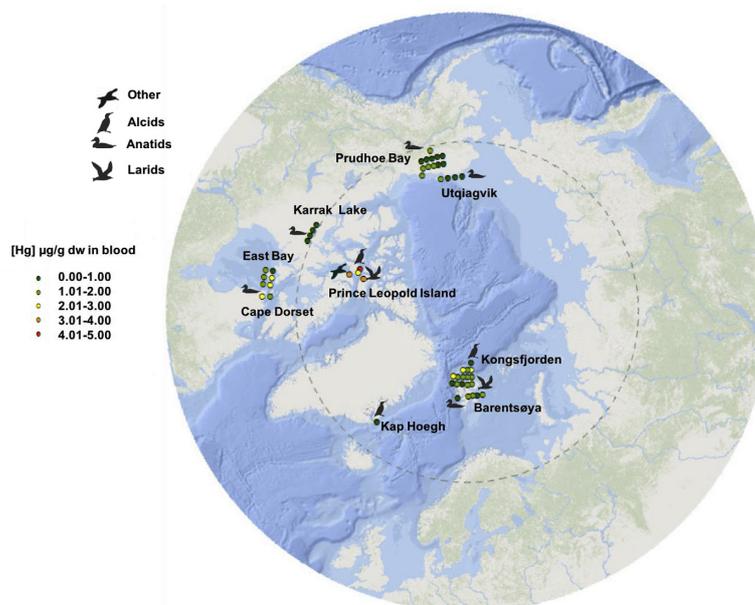


Fig. 3. Spatial distribution of Hg concentrations measured in blood.

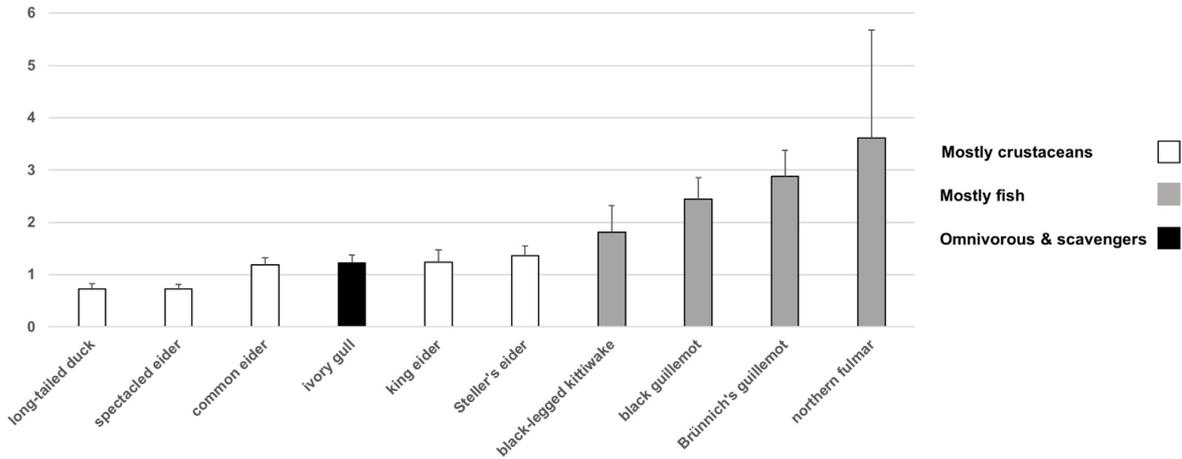


Fig. 4. Mean Hg concentrations (mean ± SD µg/g dw) measured in blood by species, in relation to bird diet during the breeding season.

linked to higher Hg concentrations in this region (Ackerman et al., 2016; Arctic Monitoring and Assessment Programme (AMAP), 2018).

The larid family showed the highest Hg concentrations in body feathers as well as the highest intra- and interspecific variations, with values ranging from $0.89 \pm 0.15 \mu\text{g/g dw}$ (Arctic tern, Kim et al., 1996a, 1996b) to $11.66 \pm 6.52 \mu\text{g/g dw}$ (ivory gull, Mallory et al., 2015) (See Table 2, Figs. 6 and 7). Interestingly, the lowest values were

measured in Arctic terns breeding in Russia, while this species is reaching toxicological concerns in the Canadian Arctic, where it faces both high Hg concentrations (in both eggs and liver) and decreasing population trends (Provencher et al., 2014, Akearok et al., 2010). In addition, the highest Hg concentrations were measured in ivory gulls breeding in the Canadian Arctic, while the lowest values for this species were measured in birds from Svalbard. Again, these results are

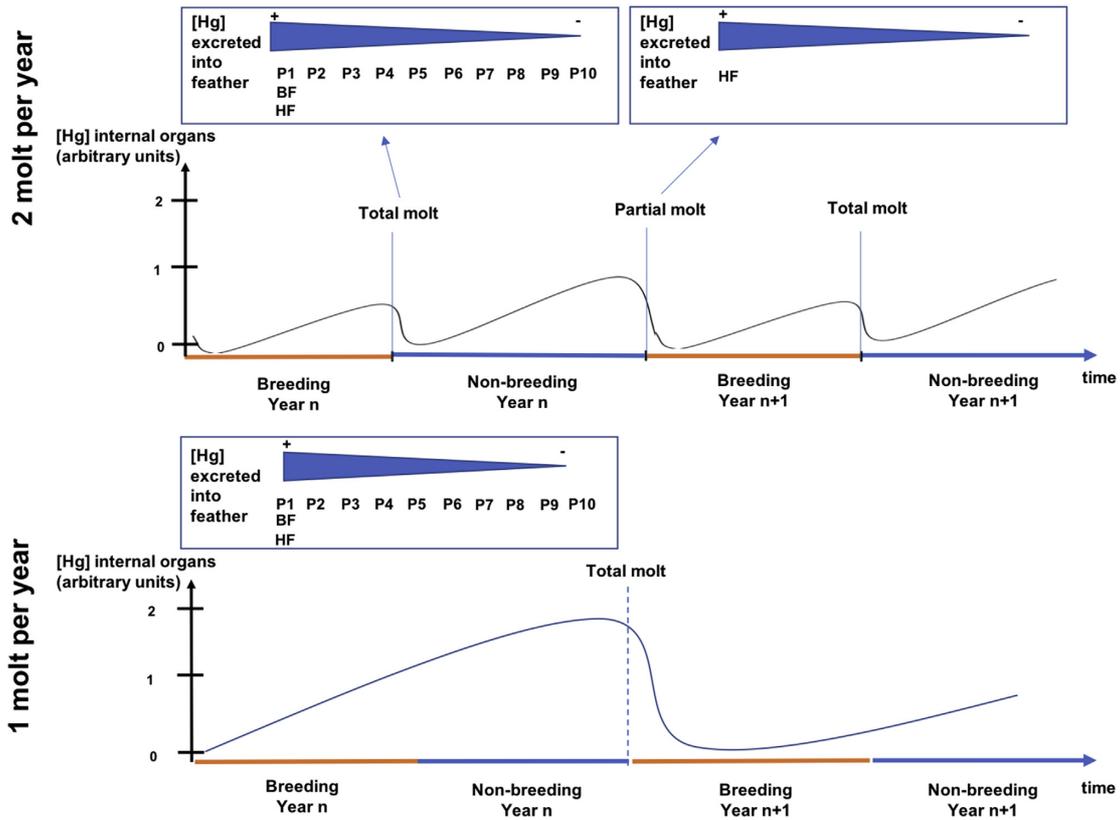


Fig. 5. Schematic representation of Hg excretion from internal tissues and organs to body, head and primary feathers for seabirds with one and two molts per year.

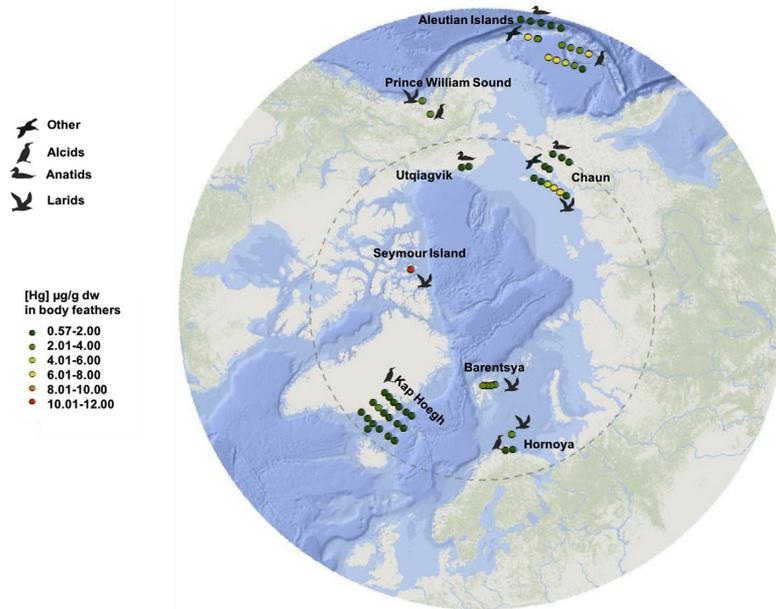


Fig. 6. Spatial distribution of Hg concentrations in body feathers.

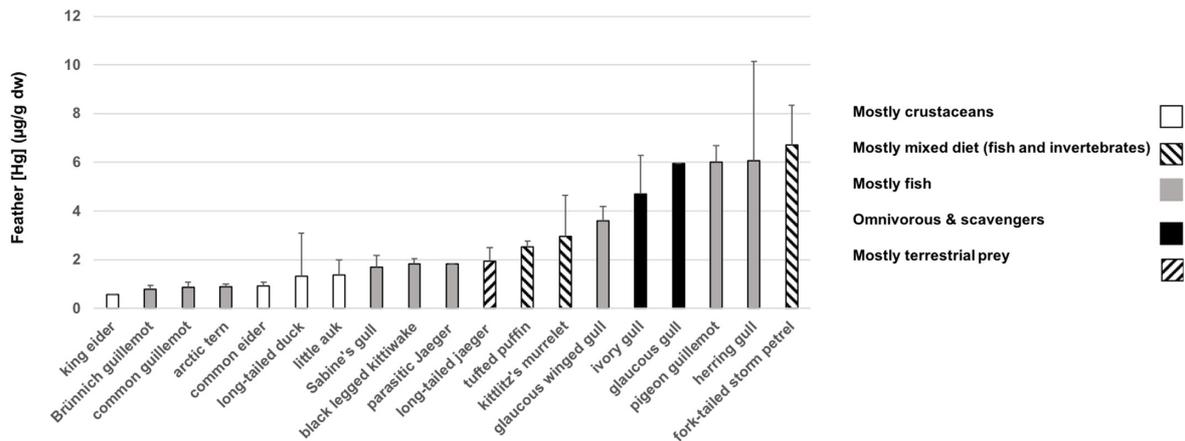


Fig. 7. Mean Hg concentrations (mean \pm SD $\mu\text{g/g dw}$) measured in body feathers, by species, in relation to bird diet during the breeding season.

consistent with Hg spatial trends highlighted from the study of other seabird tissues (Provencher et al., 2014) or other species (Arctic Monitoring and Assessment Programme (AMAP), 2018), showing that the Canadian Arctic marine biota presents higher Hg contamination compared to other Arctic regions. High Hg concentrations measured in black-legged kittiwakes, ivory gulls, glaucous, glaucous-winged and herring gulls are in accordance with their mostly piscivorous diet (opportunistic diet, from young to older fish, see Braune et al., 2016; Burgess et al., 2013; Hobson et al., 2002, 1994 for a review) and with previous studies focused on bird eggs and liver (Provencher et al., 2014).

For the other seabird families, data are much more limited. Hg concentrations in body feathers were investigated only once in hydrobatids, including the study of fork-tailed storm-petrels breeding in the Aleutian Islands, for which the molting pattern is unclear (see Annex 1, but see Harris, 1974) and exhibited the highest Hg concentrations

($6.70 \pm 1.63 \mu\text{g/g dw}$; Kaler et al., 2014), close to concentrations observed in some larids. This species mostly feeds on relatively low trophic level prey (amphipods, small fish or cephalopods - (Hobson et al., 1994)), and like pigeon guillemots and Kittlitz's murrelet, might be exposed to higher Hg concentrations in the Aleutian Islands. Similarly, stercorariids were barely investigated, with feather Hg measurements in long-tailed and parasitic jaegers breeding in Russia only. As these species have two molts per year, we suggest that body feathers represent Hg exposure during the breeding period, as for most of alcid and larid species. Long-tailed and parasitic jaegers showed Hg concentrations of 1.95 ± 0.54 and $1.84 \mu\text{g/g dw}$, respectively (Kim et al., 1996a, 1996b). The former species preferentially feeds on lemmings during the breeding season (see Gilg et al., 2013 for a review) while the latter mostly consumes robbed fishes caught by other seabirds (Davis et al., 2005). Their Hg concentrations, close to the ones of Sabine's gulls (also having two molts per year – see Annex 1) from the same region

Table 4
Total Hg concentrations measured in head feathers (HF) (mean \pm SD $\mu\text{g/g}$ dw, sample size is given in brackets) for seabirds breeding in the Arctic.

Family	Species	Sampling year	Location	Country	Head feather	References
Alcid	Little auk (<i>Alle alle</i>)	2007	Kap Hoegh (East Greenland)	Greenland	3.73 \pm 1.33 (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2008	Kap Hoegh (East Greenland)	Greenland	2.86 \pm 0.71 (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2009	Kap Hoegh (East Greenland)	Greenland	3.06 \pm 0.99 (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2010	Kap Hoegh (East Greenland)	Greenland	3.17 \pm 0.82 (40)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2011	Kap Hoegh (East Greenland)	Greenland	3.21 \pm 0.99 (25)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2012	Kap Hoegh (East Greenland)	Greenland	2.27 \pm 0.41 (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2013	Kap Hoegh (East Greenland)	Greenland	2.75 \pm 0.87 (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2014	Kap Hoegh (East Greenland)	Greenland	3.99 \pm 1.96 (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2015	Kap Hoegh (East Greenland)	Greenland	3.06 \pm 0.86 (20)	Amélineau et al. (2019)
	Little auk (<i>Alle alle</i>)	2007	Kap Hoegh (East Greenland)	Greenland	3.73 \pm 1.33 (20)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2008	Kap Hoegh (East Greenland)	Greenland	2.86 \pm 0.71 (20)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2009	Kap Hoegh (East Greenland)	Greenland	3.06 \pm 0.99 (20)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2010	Kap Hoegh (East Greenland)	Greenland	3.17 \pm 0.82 (40)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2011	Kap Hoegh (East Greenland)	Greenland	3.21 \pm 0.99 (25)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2012	Kap Hoegh (East Greenland)	Greenland	2.27 \pm 0.41 (20)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2013	Kap Hoegh (East Greenland)	Greenland	2.60 \pm 0.56 (19)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2014	Kap Hoegh (East Greenland)	Greenland	3.02 \pm 1.14 (16)	Fort et al. (2016)
	Little auk (<i>Alle alle</i>)	2010–2011	Kap Hoegh (East Greenland)	Greenland	3.17 \pm 0.83 (81)	Fort et al. (2014)

and also feeding on small fish, suggests that these jaeger populations also feed on intermediate trophic levels (see Figs. 6 and 7).

Eisler (1987) (see also Burger and Gochfeld, 1997) proposed a toxicity threshold in feathers of 5 $\mu\text{g/g}$ dw. Here, all of the anatids were below this limit. In the case of alcids and larids however, bird vulnerability depended on their breeding regions. Brünnich's and common guillemots from Norway, little auks from East Greenland and tufted puffin from Aleutian Islands were below the toxicity threshold. In Kittlitz's murrelets, most of the measured Hg concentrations were below the threshold, except for the population breeding in the Adak Island. For the pigeon guillemot, the Alaska population was below the threshold, while all the colonies from the Aleutian Islands were above. For larids, Hg concentrations of black-legged kittiwakes, Arctic tern, glaucous-winged and Sabine's gull were not of concern, while Hg concentrations of herring and glaucous gulls from Russia, and ivory gull from the Canadian Arctic, were exceeding the threshold. Finally, the fork-tailed storm-petrel from the Aleutian Islands showed Hg concentrations above this toxicity threshold. Overall, this suggests that some species and/or areas need to be carefully monitored to follow any trend in Hg contaminations and therefore permanent toxicity risks for these populations.

Flight feathers provide complementary information to other feathers, but have only been used in three studies within the Arctic (Kenney et al., 2018; Kim et al., 1996b; Mallory et al., 2015). While one of these studies sampled primary 1 or primary 3 only (Mallory et al., 2015), another focused on primary 5 (Kim et al., 1996b), and secondary feathers (Kenney et al., 2018). This makes comparisons difficult as those feathers do not molt at the same time. Nevertheless, a similar trophic/spatial pattern than in body feathers can be observed in primary feathers. The lowest Hg concentrations were found in anatids (0.59 \pm 0.21 $\mu\text{g/g}$ dw and 0.70 \pm 0.20 $\mu\text{g/g}$ dw respectively for common eiders of the Canadian Arctic; Mallory et al., 2015; and for long-tailed ducks in Russia; Kim et al., 1996a, 1996b) and the highest concentrations were observed in larids, with ivory gulls being the most contaminated species (15.79 \pm 14.13 $\mu\text{g/g}$ dw; Mallory et al., 2015). Brünnich's guillemots and northern fulmars had intermediate Hg concentrations (1.94 \pm 0.63 and 2.71 \pm 0.72 $\mu\text{g/g}$ dw, respectively; Mallory et al., 2015). Secondary feathers have only been used for the Kittlitz's murrelet breeding on the Aleutian Islands, with the highest Hg concentration being 37.18 $\mu\text{g/g}$ dw (Kenney et al., 2018). This unique value is in agreement with the Hg concentrations measured in Kittlitz's murrelet body feathers from the Aleutian Islands. However, as this type of feather has not been used in any other study, the comparison of its Hg concentration with other feathers remains uncertain.

4. Hg contamination of Arctic seabirds during the non-breeding period

As illustrated above through blood and body feather measurements, the breeding time is the most studied period in Arctic seabird Hg monitoring. However, investigations covering their entire annual cycle, and thus both the breeding and non-breeding periods, are important in order to have a complete understanding of seabird exposure to Hg and of the associated risks, both in the Arctic and outside of the Arctic. Indeed, most seabirds breeding in the Arctic migrate southwards to spend the non-breeding time outside the Arctic (Egevang et al., 2010; Gilg et al., 2013). Previous studies used bird carcasses legally shot or beached along the coasts (Bond et al., 2015; Fort et al., 2015) to get information for this specific period. However, for obvious ethical reasons or because stranded dead birds could show a bias in their Hg concentrations (e.g. starvation might affect Hg concentrations of internal tissues, Fort et al., 2015), an alternative solution is required. Seabird bycatch could provide opportunities to collect unbiased bird carcasses but would restrict samples to diving and piscivorous species and has never been used or validated until now. In that context, analysis of feathers appears as a useful and non-lethal approach to obtain information about bird non-breeding contamination, by the use of 1) head feathers for species molting twice a year, or 2) body feather for species molting once a year only. In this latter case, body feathers integrate information about Hg accumulated over the year and extraction of the non-breeding information should be performed. As proposed for the clapper rails (*Rallus longirostris obsoletus*) by Ackerman et al. (2012), it could be relevant to use published equations to predict Hg concentrations in different seabird tissues (i.e. head and body feathers, blood) and thus obtain information for specific seasons.

Head feathers have been used in only three studies focused on little auks from East Greenland to assess Hg contamination during the non-breeding period (Table 4), with values ranging from 2.27 \pm 0.41 to 3.99 \pm 1.96 $\mu\text{g/g}$ dw (Amélineau et al., 2019; Fort et al., 2016, 2014). These studies showed that non-breeding (head feathers) concentrations were significantly higher than breeding (body feathers) ones and suggested a higher exposure to Hg during the non-breeding period (Fort et al., 2014). These values are below the 5 $\mu\text{g/g}$ toxicity threshold proposed by Eisler (1987). A long-term study of the same population did not find a link between Hg concentrations and adult survival (Amélineau et al., 2019). Even though this method has been scarcely used in the Arctic, other studies combining carbon and nitrogen stable isotope analyses together with Hg measurements (both in feathers and/or in blood), demonstrated important seasonal variations in Hg contamination in Double-crested Cormorants (*Phalacrocorax auratus*)

(Lavoie et al., 2014; Ofukany et al., 2012), Caspian terns (*Hydroprogne caspia*) (Lavoie et al., 2014) and common terns (*Sterna hirundo*) (Nisbet et al., 2002). This highlights the importance to further study such seasonal variations in Hg exposure, which could be performed by the analysis of feathers.

5. Chicks as relevant stage for assessing Hg local contamination

Chicks, through measurements in blood, down and fully-grown feathers, can provide valuable information about pre-breeding and local Hg contamination (see Bond and Diamond, 2010 for the eggs). In the case of capital breeders (i.e. allocation of stored nutrients to egg formation) (Akearok et al., 2010), Hg accumulated by females during the late migration and egg formation is transferred to the eggs (Burger et al., 2008b). In the case of income breeders (i.e. allocation of local nutrient to egg formation), eggs represent female dietary Hg intakes during the egg formation only (Bond and Diamond, 2010). Several species (e.g. Atlantic puffin or razorbill) have an intermediate strategy (Bond and Diamond, 2010). As the down is grown into the egg before its hatch, Hg concentrations in eggs and chick down are correlated (Furness, 1997; Stewart et al., 1997). Hence, down was proposed as an alternative to egg collection (Monteiro and Furness, 1995), as it can be less destructive to assess Hg contamination in declining species (such as the ivory gull). For conservation purpose, eggshells could also be a useful non-lethal tissue to monitor Hg concentrations (Peterson et al., 2017).

During chick growth, the down is replaced by feathers, which thus integrate Hg from the chick diet. Therefore, chick feathers are useful indicators of the local Hg contamination during the chick rearing period (Blévin et al., 2013; Furness, 1997). Indeed, most adults feed their chicks with relatively local food (except in a few species such as for instance northern fulmars which can travel several hundreds of kilometers to find their prey (Weimerskirch et al., 2001)). Unlike eggs, chick feathers and blood have been scarcely studied in the Arctic (Kaler et al., 2014; Tartu et al., 2016; Wenzel and Gabrielsen, 1995). Chick feathers constitute a good alternative to adult feathers to assess Hg local contamination (Blévin et al., 2013; Monteiro and Furness, 1995), with the advantage to integrate, in most cases, the local Hg contamination only (see above) while adult body feathers can integrate Hg exposure over much larger geographical scales, especially in species molting once a year. Finally, chick blood informs about recent intakes and local contamination only and presents the same advantages than fully-grown feathers. However, collecting blood in growing chicks could be more challenging and stressful for the bird than collecting feathers or down.

6. Future direction: spatial ecotoxicology to link seabird movements to their contamination

Blood and feathers thus appear as important tissues for the monitoring of seabird Hg contamination and its temporal variations along their whole biological cycle. However, to further comprehend the vulnerability of these species to Hg marine contamination, it is essential to understand where individuals, populations or species get contaminated. Indeed, seabirds can use contrasting feeding areas, both at small spatial scale over a season and large scale when migrating thousands of kilometers between their breeding, molting and wintering grounds. Therefore, they potentially experience various levels of Hg environmental contamination (e.g. Leat et al., 2013). Different tools exist to track movements in Arctic seabirds. For instance, miniaturized electronic devices (e.g. GLS, GPS, Argos) are used to track large scale movements of birds over days, months or years and can be combined to Hg measurements in blood and feathers to highlight the role played by specific regions on their contamination (Fort et al., 2014).

Furthermore, Hg isotopes have been widely used in the last decades as a new and powerful perspective to explore the cycle of this element (Blum et al., 2014) and could thus be used to track the trophic and

spatial origins of Hg in polar seabirds (Renedo et al., 2018). Hg has seven naturally occurring stable isotopes that undergo both mass-dependent fractionation (MDF) and mass-independent fractionation (MIF), both related to distinct Hg isotopic ratios. Hg MDF (normally reported as $\delta^{202}\text{Hg}$) is induced during physicochemical reactions including environmental processes such as volatilization, redox transformations, methylation and demethylation, photochemical reactions and biological processes (Kritee et al., 2009, 2007; Perrot et al., 2015; Rodríguez-González et al., 2009; Zheng et al., 2007). Conversely, Hg MIF is essentially attributed to the photochemical reduction of inorganic Hg and photodemethylation of MeHg in aquatic systems before its incorporation into the food web (Bergquist and Blum, 2007; Zheng and Hintelmann, 2010). These two processes cause a significant “anomaly” in the fractionation of odd Hg isotopes (reported as $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$) which represents the deviation from the theoretical MDF. Further, as Hg MIF is not produced during biological processes (Kritee et al., 2009, 2007), its signature is conserved along the food webs. Therefore, the measurement of Hg MIF magnitudes and ratios in the tissues of top predators permits to trace Hg photochemical conditions in their foraging habitats (Masbou et al., 2018; Perrot et al., 2016; Renedo et al., 2018). Since seabirds forage in diverse locations at sea, both in the horizontal and vertical scales, their use for Hg isotopic investigations present important advantages for the investigation of Hg sources and its associated geochemical processes in the ocean (Renedo et al., 2018). In the Arctic, Point et al. (2011) were pioneers on the use of avian samples (eggs) as bioindicators for Hg isotopic analyses. These authors revealed the influence of ice cover on Hg marine photochemistry by the differences of Hg odd-MIF signatures between ice-covered and non-ice-covered oceanic areas of the Alaskan Arctic Ocean, using Brünnich's and common guillemot as bioindicators. Likewise, another study on seabird eggs from these species identified variations of Hg isotopic signatures between coastal and open ocean environments attributed to both different sources and processes (photochemistry) between these two reservoirs in the Alaskan region (Day et al., 2012). In the Southern Ocean, Hg isotopic signatures in blood samples of subantarctic adult penguins also permitted to illustrate different MeHg biogeochemical characteristics across inshore-offshore marine compartments depending on specific foraging behaviour variability within a same ecosystem (Renedo et al., 2018). Significant latitudinal variations of Hg isotopic signatures have also been observed in seabird tissues across distant colonies of the Southern Ocean (Renedo et al., submitted). These observations indicate that both specific foraging habitats and latitudinal movements of seabirds highly determine their exposure to distinct environmental MeHg sources in variable marine ecosystems.

Hg isotopic composition in seabirds has demonstrated to be a potent tool for the definition of MeHg sources and to highlight the variability of processes among the different marine compartments. Therefore, the use of Hg isotopic approaches in migratory seabirds, feather Hg isotopic signatures will reflect the integration of Hg from diverse sources during an entire year for species molting once a year. This integrative effect of adult feathers could be interesting for tracking whole annual Hg contamination, i.e. including the wintering areas. Furthermore, analyses of different type of feather of species moulting twice a year such as the case of alcids, appears as a suitable approach to obtain information about both the breeding and non-breeding Hg contamination. Measurements of Hg isotopic composition in feathers of flying migratory seabirds could then open a new horizon for exploring Hg exposure pathways at their breeding sites in summer but also in large zones of the marine environment during their non-breeding period outside the Arctic. Finally, stable isotopes of carbon and nitrogen (respectively indicators of foraging habitat and trophic level), together with the use of Hg isotopes (proxies of Hg biogeochemical characteristics in each marine compartment), would enable to help elucidating the Hg exposure pathways of migratory seabirds during their whole annual cycle, both during their breeding and non-breeding periods.

7. Limitations

Although highly valuable to inform about seabird Hg contamination through the Arctic, the use of blood and feathers also shows some limitations. Even if seabirds have been used in ecotoxicology since decades, detailed information about their trophic and behavioral ecology is essential to properly interpret their exposure to the environmental contamination. Such information is nonetheless often missing or ignored. For instance, information about seabird diet and molting patterns, mostly during the non-breeding period, are still scarce in many species. The present paper reviewed this existing knowledge of Arctic seabird molting patterns (pre- and/or post-breeding) (Annex 1) and should help future investigations using feathers to track seabird Hg contamination. Furthermore, Hg measurements are not always performed under the same protocols (e.g. different sample preparations leading to contrasting units (dry vs wet weights)). Although correction factors exist, they add uncertainties in the data and complicate comparisons. Such potential bias could be easily avoided by the use of homogenized methods, ensuring comparability of Hg measurements between areas and species. Finally, spatial ecotoxicology approaches combining biologging with Hg analyses should be developed to track the spatial origin of Hg contamination. However, large scale movements such as migration could also be associated to changes in diet, possibly affecting Hg intakes. Although nitrogen stable isotopes can be used to investigate seabird trophic status, isotopic values cannot be directly compared to Hg measurements in feathers as they do not provide information for the same periods (Bond, 2010). Therefore, feathers cannot be corrected to take into account potential diet effects as we can do for blood samples.

8. Conclusion

Although largely neglected in comparison to internal tissues or eggs, feathers and blood are two complementary and important tissues allowing large spatio-temporal investigations of Hg contamination in Arctic seabirds during both the breeding and non-breeding periods. They also provide information about risks encountered by seabirds depending on their Arctic breeding site, their migratory movements and their wintering grounds, within or outside the Arctic. Further studies monitoring long-term and large scale Hg contamination and potential subsequent risks for Arctic seabirds are now necessary, and require large international collaborations and monitoring programmes such as those implemented in Canada (Braune et al., 2001, Northern Contaminants Program (Chételat and Braune, 2012)), Norway (SEAPOP (Anker-Nilssen et al., 2017)) or at the pan-Arctic scale (newly established ARCTOX program). Such initiatives are indeed essential in order to determine temporal and spatial trends, to define baseline and hot-spots of Hg contamination, and to allow international actions (see the Minamata Convention, the Arctic Monitoring Assessment Program or the Global Mercury Assessment).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envres.2019.108588>.

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Annex 1: List of the 42 species referenced in the 101 articles who studied Hg contamination in seabirds breeding in the Arctic. For each species, pre or post breeding have been assessed.

Family	Species	Prebreeding molt	Postbreeding molt	Molt pattern unclear or unknow	References
Alcid	Atlantic puffin (<i>Fratercula arctica</i>)			x	Gaston and Jones, 1998 ; Harris and Yule, 1977
	Black guillemot (<i>Cepphus grylle</i>)	x	x		Gaston and Jones, 1998
	Mandt's black guillemot (<i>Cepphus grylle mandtii</i>)	x	x		Gaston and Jones, 1998
	Brünnich guillemot (<i>Uria lomvia</i>)	x	x		Gaston and Jones, 1998
	Common guillemot (<i>Uria aalge</i>)	x	x		Gaston and Jones, 1998
	Crested auklet (<i>Aethia cristatella</i>)	x	x		Gaston and Jones, 1998
	Horned puffin (<i>Fratercula corniculata</i>)	x	x		Piatt and Kitaysky, 2002
	Kittlitz's murrelet (<i>Brachyramphus brevirostris</i>)	x	x		Gaston and Jones, 1998
	Little auk (<i>Alle alle</i>)	x	x		Gaston and Jones, 1998
	Parakeet auklet (<i>Aethia psittacula</i>)	x	x		Gaston and Jones, 1998
	Pigeon guillemot (<i>Cepphus columba</i>)	x	x		Gaston and Jones, 1998
	Razorbill (<i>Alca torda</i>)	x	x		Gaston and Jones, 1998
	Tufted puffin (<i>Fratercula cirrhata</i>)	x	x	Post-breeding apparently complete	Gaston and Jones, 1998
	Whiskered auklet (<i>Aethia pygmaea</i>)			x	Gaston and Jones, 1998
Anatid	Black Scoter (<i>Melanitta nigra</i>)		x		Sea Duck Joint Venture, 2016
	Common eider (<i>Somateria mollissima</i>) female		x		Waltho and Coulson, 2015
	Common eider (<i>Somateria mollissima</i>) male	x	x	Experience an eclipse plumage before going back to breeding plumage	Waltho and Coulson, 2015
	King eider (<i>Somateria spectabilis</i>)		x		Fenneman and Toochin, 2013
	Long-tailed duck/oldsquaw (<i>Clangula hyemalis</i>)	x	x		Howell et al., 2003

	Spectacled eiders (<i>Somateria fischeri</i>)		x		US Fish & Wildlife Service, 2012
	Steller's eiders (<i>Polysticta stelleri</i>)		x		US Fish & Wildlife Service, 2012
	White-winged Scoter (<i>Melanitta fusca</i>)		x		Brown and Fredrickson, 1997; Dickson et al., 2012
Hydrobatid	Fork-tailed storm-petrel (<i>Oceanodroma furcata</i>)			x	But see Harris, 1974
	Leach's Storm-petrel (<i>Oceanodroma leucorhoa</i>)		x		Harris, 1974
Larid	Arctic tern (<i>Sterna paradisae</i>)	x	x		Voelker, 1997
	Black-legged kittiwakes (<i>Rissa tridactyla</i>)	x	x		Cramp and Simmons, 1983
	Glaucous gull/Arctic gull (<i>Larus hyperboreus</i>)	x	x		Hume, 1975
	Glaucous-winged gulls (<i>Larus glaucescens</i>)	x	x		Gabrielson and Lincoln, 1959
	Herring gull (<i>Larus argentatus</i>)	x	x		Cramp and Simmons, 1983
	Iceland gull (<i>Larus glaucooides</i>)	x	x		Cramp and Simmons, 1983
	Ivory gull (<i>Pagophila eburnea</i>)	x			Cramp and Simmons, 1983
	Ross's gull (<i>Rhodostethia rosea</i>)	x	x		Cramp and Simmons, 1983
	Sabine's gull (<i>Xema sabini</i>)	x	x		Cramp and Simmons, 1983
	Thayer's gull (<i>Larus thayeri</i>)	x	x		Howell and Gunn, 2007
Phalacrocoracid	European shag (<i>Phalacrocorax aristotelis aristotelis</i>)		x		Potts, 1970
	Pelagic cormorant (<i>Phalacrocorax pelagicus</i>)	x	x		Rohwer, 1999; Van Tets, 1959
	Red-faced cormorants (<i>Phalacrocorax urile</i>)	x	x		Toochin and Fenneman, 2014
Procellariid	Northern fulmar (<i>Fulmarus glacialis</i>)		x	Initiated around one week before hatching	Allard et al., 2008
Stercorariid	Long-tailed Jaeger (<i>Stercorarius longicaudus</i>)	x	x		Pyle, 2016
	Parasitic Jaeger (<i>Stercorarius parasiticus</i>)	x	x		Pyle, 2016
	Pomarine skua (<i>Stercorarius pomarinus</i>)	x	x		Pyle, 2016
Sulid	Northern gannet (<i>Morus bassanus</i>)			Continuous molt from post breeding to pre-breeding period	Nelson, 2010

Annex 2: List of the 101 articles studying Hg in seabirds breeding in the Arctic.

Article Nb	Authors	Title
1	Amélineau et al., 2019	Arctic climate change and pollution impact little auk foraging and fitness across a decade
2	Eckbo et al., 2019	Individual variability in contaminants and physiological status in a resident Arctic seabirds species
3	Miller et al., 2019	Interspecific patterns of trace elements in sea ducks: can surrogate species be used in contaminants monitoring?
4	Blévin et al., 2018	Organochlorines, perfluoroalkyl substances, mercury and egg incubation temperatures in an Arctic seabirds: insight from data loggers.
5	Braune B.M. et al., 2018	Variation in organochlorine and mercury levels in first and replacement eggs of a single-egg clutch breeder, the thick-billed murre, at a breeding colony in the Canadian Arctic
6	Keney et al., 2018	Mercury concentrations in multiple tissues of Kittlitz's murrelets (<i>Brachyramphus brevirostris</i>)
7	Mallory et al., 2018	Mercury concentrations in blood, brain and muscle tissues of coastal and peagic birds from northeastern Canada
8	Blévin et al., 2017	Contaminants and energy expenditure in an Arctic seabird: organochlorine pesticides and perfluoroalkyl substances are associated with metabolic rate in a contrasted manner.
9	Fenstad A.A. et al., 2017	Blood and feather concentrations of toxic elements in a Baltic and an Arctic seabird population
10	Ishii C., et al. 2017	Contamination status and accumulation characteristics of heavy metals and arsenic in five seabird species from the central Bering Sea
11	Mallory M.L. and Braune B.M. 2017	Do concentrations in eggs and liver tissue tell the same story of temporal trends of mercury in high Arctic seabirds?
12	Mallory C.D. et al., 2017	Hepatic trace element concentrations of breeding female common eiders across a latitudinal gradient in the eastern Canadian Arctic
13	Provencher J.F. et al 2017	Anti-parasite treatment, but not mercury burdens, influence nesting propensity dependent on arrival time or body condition in a marine bird
14	Braune B.M. et al., 2016	Temporal trends of mercury in eggs of five sympatrically breeding seabird species in the Canadian Arctic
15	Fenstad A.A. et al., 2016	DNA double-strand breaks in incubating female common eiders (<i>Somateria mollissima</i>): Comparison between a low and a high polluted area
16	Fort J. et al., 2016	Does temporal variation of mercury levels in Arctic seabirds reflect changes in global environmental contamination, or a modification of Arctic marine food web functioning?
17	Lucia M. et al., 2016	Trace Element Concentrations in Relation to the Trophic Behaviour of Endangered Ivory Gulls (<i>Pagophila eburnea</i>) During Their Stay at a Breeding Site in Svalbard

18	Miller et al., 2016	Trace elements in Sea ducks of the Alaskan Arctic Coast: patterns of variation among species, sexes, and ages
19	Provencher J.F. et al., 2016	Implications of mercury and lead concentrations on breeding physiology and phenology in an Arctic bird
20	Peck L.E. et al., 2016	Persistent organic pollutant and mercury concentrations in eggs of ground-nesting marine birds in the Canadian high Arctic
21	Provencher J.F. et al., 2016	Direct and indirect causes of sex differences in mercury concentrations and parasitic infections in a marine bird
22	Tartu S. et al., 2016	Mercury exposure, stress and prolactin secretion in an Arctic seabird: An experimental study
23	Bond A.L. et al., 2015	Rapidly increasing methyl mercury in endangered ivory gull (<i>Pagophila eburnea</i>) feathers over a 130 year record
24	Clayden M.G. et al., 2015	Mercury bioaccumulation and biomagnification in a small Arctic polynya ecosystem
25	Goutte A. et al., 2015	Survival rate and breeding outputs in a high Arctic seabird exposed to legacy persistent organic pollutants and mercury
26	Huber S. et al., 2015	A broad cocktail of environmental pollutants found in eggs of three seabird species from remote colonies in Norway
27	Lucia M. et al., 2015	Circumpolar contamination in eggs of the high-arctic ivory gull <i>Pagophila eburnea</i>
28	Mallory M.L. et al., 2015	Mercury concentrations in feathers of marine birds in Arctic Canada
29	Øverjordet I.B. et al., 2015	Effect of diet, location and sampling year on bioaccumulation of mercury, selenium and cadmium in pelagic feeding seabirds in Svalbard
30	Overjordet I.B. et al., 2015	Toxic and essential elements changed in black-legged kittiwakes (<i>Rissa tridactyla</i>) during their stay in an Arctic breeding area
31	Ruus A. et al., 2015	Methylmercury biomagnification in an Arctic pelagic food web
32	Tartu S. et al., 2015	Increased adrenal responsiveness and delayed hatching date in relation to polychlorinated biphenyl exposure in Arctic-breeding black-legged kittiwakes (<i>Rissa tridactyla</i>)
33	Braune B.M. et al., 2014	A geographical comparison of mercury in seabirds in the eastern Canadian Arctic
34	Braune B.M. et al., 2014	Changes in food web structure alter trends of mercury uptake at two seabird colonies in the Canadian Arctic
35	Fort J. et al., 2014	Spatial ecotoxicology: Migratory arctic seabirds are exposed to mercury contamination while overwintering in the northwest Atlantic
36	Kaler R.S.A. et al., 2014	Mercury concentrations in breast feathers of three upper trophic level marine predators from the western Aleutian Islands, Alaska
37	Provencher J.F. et al., 2014	Trace element concentrations and gastrointestinal parasites of Arctic terns breeding in the Canadian High Arctic
38	Brasso R.L. and Polito M.J., 2013	Trophic calculations reveal the mechanism of population-level variation in mercury concentrations between marine ecosystems: Case studies of two polar seabirds

39	Tartu S. et al., 2013	To breed or not to breed: Endocrine response to mercury contamination by an Arctic seabird
40	Braune B.M. et al., 2012	Toxicity of methylmercury injected into eggs of thick-billed murre and arctic terns
41	Day R.D. et al., 2012	Regional, temporal, and species patterns of mercury in Alaskan seabird eggs: Mercury sources and cycling or food web effects?
42	Day R.D. et al., 2012	Mercury stable isotopes in seabird eggs reflect a gradient from terrestrial geogenic to oceanic mercury reservoirs
43	Miljeteig C. et al., 2012	Eggshell thinning and decreased concentrations of vitamin E are associated with contaminants in eggs of ivory gulls
44	Point D. et al., 2011	Methylmercury photodegradation influenced by sea-ice cover in Arctic marine ecosystems
45	Akearok J.A. et al., 2010	Inter- and intraclutch variation in egg mercury levels in marine bird species from the Canadian Arctic
46	Braune B.M. et al., 2010	Persistent halogenated organic contaminants and mercury in northern fulmars (<i>Fulmarus glacialis</i>) from the Canadian Arctic
47	Wayland M. et al., 2010	Evidence of weak contaminant-related oxidative stress in glaucous gulls (<i>Larus hyperboreus</i>) from the Canadian arctic
48	Miljeteig C. et al., 2009	High levels of contaminants in ivory gull <i>Pagophila eburnea</i> eggs from the Russian and Norwegian arctic
49	Burger J. and Gochfeld M., 2009	Mercury and other metals in feathers of common eider (<i>Somateria mollissima</i>) and tufted puffin (<i>Fratercula cirrhata</i>) from the Aleutian chain of Alaska
50	Burger J. et al., 2009	Mercury and other metals in eggs and feathers of glaucous-winged gulls (<i>Larus glaucescens</i>) in the Aleutians
51	Burger J. and Gochfeld M., 2009	Comparison of arsenic, cadmium, chromium, lead, manganese, mercury and selenium in feathers in bald eagle (<i>Haliaeetus leucocephalus</i>), and comparison with common eider (<i>Somateria mollissima</i>), glaucous-winged gull (<i>Larus glaucescens</i>), pigeon guillemot (<i>Cephus columba</i>), and tufted puffin (<i>Fratercula cirrhata</i>) from the Aleutian chain of Alaska
52	Jæger I. et al., 2009	Biomagnification of mercury in selected species from an Arctic marine food web in Svalbard
53	Sagerup K. et al., 2009	Persistent organic pollutants and mercury in dead and dying glaucous gulls (<i>Larus hyperboreus</i>) at Bjørnøya (Svalbard)
54	Sagerup K. et al., 2009	Persistent organic pollutants, heavy metals and parasites in the glaucous gull (<i>Larus hyperboreus</i>) on Spitsbergen
55	Braune B.M. and Scheuhammer A.M., 2008	Trace element and metallothionein concentrations in seabirds from the Canadian arctic
56	Burger J. et al., 2008	Assessment of metals in down feathers of female common eiders and their eggs from the Aleutians: Arsenic, cadmium, chromium, lead, manganese, mercury, and selenium
57	Burger J. et al., 2008	Arsenic, cadmium, chromium, lead, manganese, mercury, and selenium in feathers of Black-legged Kittiwake (<i>Rissa tridactyla</i>) and Black Oystercatcher (<i>Haematopus bachmani</i>) from Prince William Sound, Alaska
58	Helgason L.B. et al., 2008	Levels and temporal trends (1983-2003) of persistent organic pollutants (POPs) and mercury (Hg) in seabird eggs from Northern Norway

59	Ricca M.A. et al., 2008	Sources of organochlorine contaminants and mercury in seabirds from the Aleutian archipelago of Alaska: Inferences from spatial and trophic variation
60	Wayland M. et al., 2008	Survival rates and blood metal concentrations in two species of free-ranging North American sea ducks
61	Wayland M. et al., 2008	Trace element concentrations in Blood of nesting king eiders in the Canadian Arctic
62	Yin X. et al., 2008	Animal excrement: A potential biomonitor of heavy metal contamination in the marine environment
63	Braune B.M., 2007	Temporal trends of organochlorines and mercury in seabird eggs from the Canadian Arctic, 1975-2003
64	Burger J. et al., 2007	Mercury levels and potential risk from subsistence foods from the Aleutians
65	Burger J. et al., 2007	Mercury, arsenic, cadmium, chromium lead, and selenium in feathers of pigeon guillemots (<i>Cepphus columba</i>) from Prince William Sound and the Aleutian Islands of Alaska
66	Burger J. and Gochfeld M., 2007	Metals and radionuclides in birds and eggs from Amchitka and Kiska Islands in the Bering Sea/Pacific Ocean ecosystem
67	Rigét F. et al., 2007	Transfer of mercury in the marine food web of West Greenland
68	Wayland M. et al., 2007	Year-to-year correlations in blood metal levels among individuals of two species of North American sea ducks
69	Borgå K. et al., 2006	Regional and species specific bioaccumulation of major and trace elements in arctic seabirds
70	Braune B.M. et al., 2006	Elevated mercury levels in a declining population of ivory gulls in the Canadian Arctic
71	Braune B.M. and Malone B.J., 2006	Mercury and selenium in livers of waterfowl harvested in northern Canada
72	Day R.D. et al., 2006	Murre eggs (<i>Uria aalge</i> and <i>Uria lomvia</i>) as indicators of mercury contamination in the Alaskan marine environment
73	Braune B.M. et al., 2005	Regional differences in collagen stable isotope and tissue trace element profiles in populations of long-tailed duck breeding in the Canadian Arctic
74	Campbell L.M. et al., 2005	Mercury and other trace elements in a pelagic Arctic marine food web (Northwater Polynya, Baffin Bay)
75	Rothschild R.F.N. and Duffy L.K., 2005	Mercury concentrations in muscle, brain and bone of Western Alaskan waterfowl
76	Wayland M. et al., 2005	Concentrations of cadmium, mercury and selenium in common eider ducks in the eastern Canadian arctic: Influence of reproductive stage
77	Braune B.M. et Simon M., 2004	Trace elements and halogenated organic compounds in Canadian Arctic seabirds
78	Franson J.C. et al., 2004	Contaminants in molting long-tailed ducks and nesting common eiders in the Beaufort Sea
79	Johansen P. et al., 2004	Human exposure to contaminants in the traditional Greenland diet

80	Mallory M.L. et al., 2004	Trace elements in marine birds, arctic hare and ringed seals breeding near Qikiqtarjuaq, Nunavut, Canada
81	Rocque D.A. et Winker K., 2004	Biomonitoring of contaminants in birds from two trophic levels in the North Pacific
82	Wilson H.M. et al., 2004	Concentrations of metals and trace elements in blood of spectacled and king eiders in northern Alaska, USA
83	Savinov V.M. et al., 2003	Cadmium, zinc, copper, arsenic, selenium and mercury in seabirds from the Barents Sea: Levels, inter-specific and geographical differences
84	Wayland M. et al., 2003	Biomarker responses in nesting, common eiders in the Canadian arctic in relation to tissue cadmium, mercury and selenium concentrations
85	Braune B.M et al., 2002	Contaminant residues in seabird eggs from the Canadian Arctic. II. Spatial trends and evidence from stable isotopes for intercolony differences
86	Christopher S.J. et al., 2002	Determination of mercury in the eggs of common murres (<i>Uria aalge</i>) for the seabird tissue archival and monitoring project
87	Stout J.H. et al., 2002	Environmental contaminants in four eider species from Alaska and arctic Russia
88	Wayland M. et al., 2002	Immune function, stress response, and body condition in arctic-breeding common eiders in relation to cadmium, mercury, and selenium concentrations
89	Braune B.M. et al., 2001	Contaminant residues in seabird eggs from the Canadian Arctic. Part I. Temporal trends 1975-1998
90	Wayland M. et al., 2001	Concentrations of cadmium, mercury and selenium in blood, liver and kidney of common eider ducks from the Canadian arctic
91	Wayland M. et al., 2001	Trace elements in king eiders and common eiders in the Canadian arctic
92	Riget F. and Dietz R., 2000	Temporal trends of cadmium and mercury in Greenland marine biota
93	Trust K.A. et al., 2000	Contaminant exposure and biomarker responses in spectacled eiders (<i>Somateria fischeri</i>) from St. Lawrence Island, Alaska
94	Donaldson G.M. et al., 1997	Organochlorine and heavy metal residues in breast muscle of known-age thick-billed murres (<i>Uria lomvia</i>) from the Canadian Arctic
95	Barrett R.T. et al., 1996	Recent changes in levels of persistent organochlorines and mercury in eggs of seabirds from the Barents Sea
96	Dietz R. et al., 1996	Lead, cadmium, mercury and selenium in Greenland marine animals
97	Kim E.Y. et al., 1996	Mercury levels and its chemical form in tissues and organs of seabirds
98	Kim E.-Y. et al., 1996	Metal accumulation in tissues of seabirds from Chaun, northeast Siberia, Russia
99	Wenzel C. and Gabrielsen G.W., 1995	Trace element accumulation in three seabird species from Hornøya, Norway
100	Dietz R. et al., 1990	Organic mercury in Greenland birds and mammals

101	Norheim G. and Kjos-Hanssen B., 1984	Persistent chlorinated hydrocarbons and mercury in birds caught off the west coast of Spitsbergen
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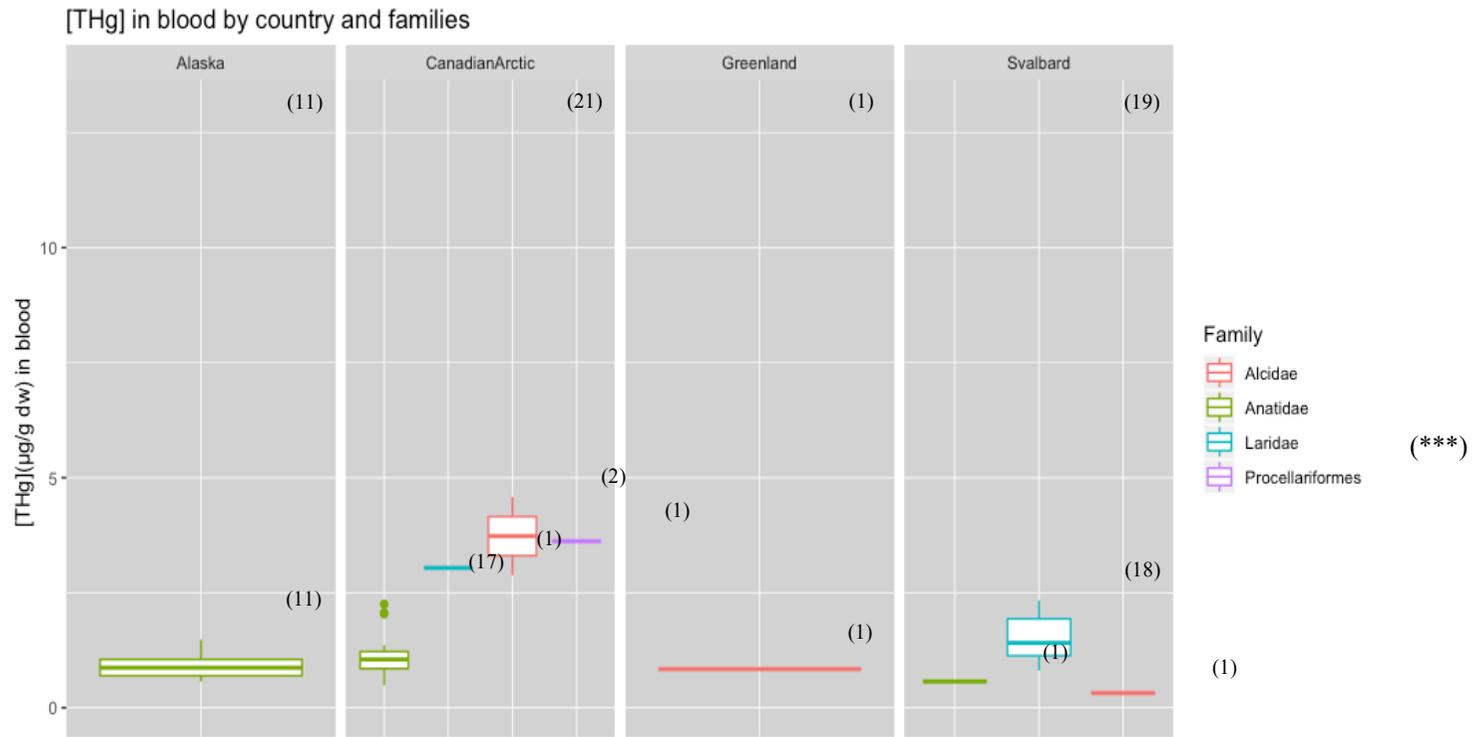


Figure S4: Hg concentration ($\mu\text{g/g dw}$) in blood, in addition to sample size (into bracket), per country and family. Hg concentrations comparisons have been done between the adults anatids ($n=29$) and the larids ($n=19$). Alcids ($n=4$) and the procellariiformes ($n=1$) have been removed of the statistics because of limited data. Significance in the legend into brackets.

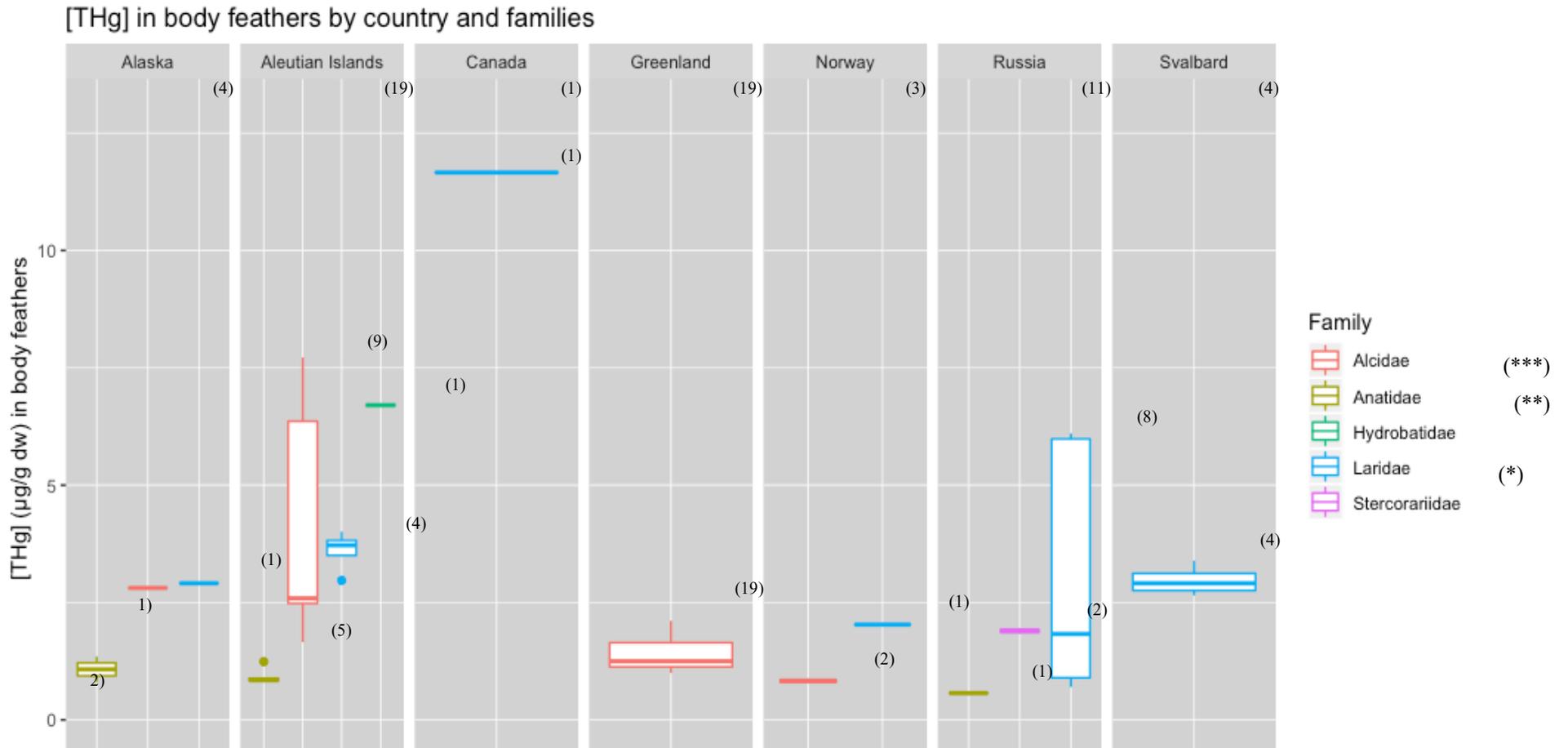


Figure S2: Hg concentration (µg/g dw) in body feathers, in addition to sample size (into bracket), per country and family. Hg concentrations comparisons have been done between the adults of alcids (n=31), anatids (n=8) and larids (n=19). The hydrobatids (n=1) and the stercorariids (n=2) have been removed from the statistical analyses because of limited data. Significance in the legend into brackets.

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Paper 2

Tracking mercury in Arctic seabirds and marine food webs.

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Paper 3

Seasonal variation of mercury contamination in Arctic seabirds: a pan-arctic assessment.

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Key words: polar, top predators, metal, seasonal variation, feathers

Abstract:

Mercury (Hg) is a natural trace element found in high concentrations in top predators including Arctic seabirds. Most current knowledge about Hg concentrations in Arctic seabirds concerns exposure during the summer breeding period when they can be accessed at colonies. However, the few studies focused on winter have shown higher Hg concentrations in several tissues. Hence, improving knowledge about Hg exposure during the non-breeding period is crucial to understanding the threats and risks encountered by these species. We used feathers of nine migrating alcid species occurring in high latitudes to study bird Hg exposure during both the breeding and non-breeding periods. Overall, Hg concentrations were higher during the non-breeding period and spatial differences within and between the Atlantic and Pacific regions were apparent, showing the highest Hg seasonal variations in seabirds breeding in the West Atlantic. In addition, Hg concentrations during the non-breeding period were above 5 $\mu\text{g}\cdot\text{g}^{-1}$ dw, considered to be the threshold for which deleterious effects are known for most of the seabird colonies, suggesting that some populations might be vulnerable to non-breeding Hg exposure. Wintering ground locations, migration routes and diet may influence seasonal Hg concentrations and it is crucial to improve our knowledge about spatial ecotoxicology to fully understand the risks associated with Hg contamination in Arctic seabirds.

Introduction

Mercury (Hg), a naturally-occurring non-essential element in marine ecosystems, has increased in concentration over the last decades because of anthropogenic emissions (UNEP, 2013). In its methylated, toxic form (methyl-mercury, MeHg), it is considered a pollutant of concern for both wildlife and human health (Dietz et al., 2019; Tan et al., 2009; Wolfe et al., 1998), even in remote areas such as the polar regions (Albert et al., 2019a; AMAP, 2018a; Johansen et al., 2007; Provencher et al., 2014). Hg is absorbed by organisms through diet and biomagnifies through food webs (Morel et al., 1998). The highest concentrations are generally found in top predators such as seabirds, although marine mammals such as seals, toothed whales and polar bears *Ursus maritimus* carry even higher body burdens (AMAP, 2005, 1998; Dietz et al., 2019, 2013a, 1996). In turn, elevated Hg may be associated with physiological, behavioural or reproductive impacts such as neurological deficiencies, immune disruption or lowered egg hatchability (Ackerman et al., 2016; Dietz et al., 2019), ultimately having long-term, deleterious effects on seabird population dynamics (Goutte et al., 2014a, 2014b). Spatial differences have been highlighted in numerous species with higher Hg concentrations found in the Canadian Arctic compared to the European Arctic (AMAP, 2018a), but with large interspecies variation (see Albert et al., 2019 for a review; AMAP, 2018). However, current knowledge about Hg concentrations in Arctic seabirds is restricted mostly to the breeding season (but see Fleishman et al., 2019a; Fort et al., 2014a), excluding the non-breeding period, which is mostly spent in areas outside the Arctic.

At the end of the reproductive season, most Arctic seabirds leave their breeding site and mostly migrate to southerly areas where conditions, including environmental Hg contamination, might differ from those encountered at their breeding areas. For example, little auks (or dovekies; *Alle alle*) had higher Hg concentrations during the non-breeding period spent east of Newfoundland than during the breeding season in East Greenland, negatively impacting their subsequent reproduction (Fort et al., 2014a). Outside of the Arctic, common guillemots (or common murre, *Uria aalge*) have higher and increasing Hg concentrations while wintering in the North Sea compared to their breeding grounds (Joiris et al., 1997). In this context, to better understand the Hg contamination risk of Arctic seabirds, the concentrations of this element should be studied at multiple spatial and temporal scales.

The alcid family of seabirds (Charadriiformes: Alcidae) comprises the most numerous species in the Arctic and shows a circumpolar distribution. Some species are endemic to the Pacific Arctic, such as murrelets *Brachyramphus*, auklets *Aethiini* and tufted puffins *Fratercula cirrhata*. Others are endemic to the Atlantic Arctic, such as little auks and razorbills *Alca torda*, or are common to both regions, such as black guillemots *Cepphus grylle*, common guillemots and Brünnich's guillemots (or thick-billed murre; *Uria lomvia*) (Gaston and Jones 1998). Alcids feed on zooplankton (e.g. little auks), on fish (e.g. common guillemots) or have a mixed diet (e.g. Brünnich's guillemot or tufted puffin) during or between seasons (i.e. switch of diet between season; Gaston & Jones, 1998). In the Arctic, alcids (except black guillemots) aggregate in large colonies for reproduction, then most migrate over large distances to overwinter in open seas, generally farther south. During these large-scale movements, alcids undergo two seasonal moults with important implications for their Hg contamination as approximately 70% to 90% of the Hg body burden is excreted into growing feathers (Agusa et al., 2005; Braune, 1987; Honda et al., 1986a). A partial moult (head, cheeks and neck) occurs after the non-breeding period in the spring leading to the nuptial plumage, and a total moult (body, head and wings) occurs after the breeding season, leading to the wintering plumage. Thus, Hg concentrations in head feathers mostly provide information about non-breeding exposure (from fall to early spring), while concentrations in body and wing feathers inform mostly about contamination at the breeding sites (from early spring to fall - Albert et al., 2019; Fort et al., 2014).

Here we focused on nine alcid species distributed around the circumpolar Arctic, representing different marine foraging niches and ocean systems. More specifically, we used body and head feathers to investigate seasonal, spatial and interspecific variations in Hg contamination. We predict higher Hg concentrations i) in head feathers (i.e. non-breeding period) than in body feathers (i.e. breeding period), ii) in the western Atlantic, iii) in species feeding at higher trophic level based on known foraging ecology and patterns of Hg exposure in the Pacific, and West and East Atlantic.

Material and methods

Sample collection

Body feathers and head feathers (hereafter BF and HF, respectively) were collected during the breeding season from ancient murrelets *Synthliboramphus antiquus*, Brünnich's guillemots, common guillemots, crested auklets *Aethia cristatella*, least auklets *Aethia pusilla*, little auks, razorbills, rhinoceros auklets *Cerorhinca monocerata* and tufted puffins. The sampling took place at 28 breeding colonies located in areas ranging from the North Atlantic and to North Pacific and from the sub-Arctic to the High Arctic between 2015 to 2017 ($n=1,331$, Fig. 1, Table 1, Table SI 1). Seabirds were captured during late incubation and chick-rearing with a noose pool, noose carpet, mist-net or by hand, handled for 5 - 10 min and released after sampling. In the Bering Sea (excluding St Lawrence Island), the Aleutian Islands and the Sea of Okhotsk, BF and HF were sampled on recently deceased birds.

Hg analysis

Prior to chemical analysis, any external contaminants were removed by plunging feathers for 3 min in a 2:1 chloroform:methanol solution in an ultrasonic bath. Feathers were then rinsed twice in a methanol solution and dried at 45°C for 48 h. Analyses were then performed on a pool of three homogenized feathers (Carravieri et al., 2014a). Total Hg concentrations were measured on a subsample of 0.50 - 2.00 mg using an Advanced Mercury Analyser spectrophotometer (Altec AMA 254) at the Littoral, Environnement et Sociétés laboratory (LIENSs, La Rochelle, France). For each sample, analyses were repeated two or three times, until the relative standard deviation for the aliquots was <10%. Subsequently, the mean of the repeated Hg measurements was used for statistical analysis. To ensure the accuracy of measurements, certified reference materials (dogfish liver DOLT-5 and lobster hepatopancreas TORT-2; NRC, Canada; reference values were 0.27 ± 0.06 SD and 0.44 ± 0.18 SD, respectively) were analysed every 15 samples. In addition, blanks were run prior to the analyses. and the detection limit was established at 0.05 ng dw.

Statistical analyses

To investigate seasonal Hg variations, we first classified seabird colonies into three regions: the Pacific, the East Atlantic and the West Atlantic (Fig. 1), following previous studies and known differences in the ecology of seabirds across these regions (see Descamps et al., 2019). We used linear

mixed models (package R “lme4”, Bates et al., 2015) to examine patterns in Hg seasonality and how that varied among species or region. We considered all species in a first step and focused in a second step on the two guillemot species individually, as these were the only two species sampled in all three regions. In each model, we always included the type of feather (representing the breeding or non-breeding period), plus the region and the species (1st step only) as fixed effects. Colonies and individuals were sampled for one to three breeding seasons during the study period (2015-2017). Hence, to address the potential non-independence among Hg concentrations taken within years, within individuals, and within colonies, all three were treated as random effects. The full model followed the form: [Hg] ~ fixed effects (Regions + Species + type of feathers) + random effects (years + Individuals + sampling sites). Hg concentrations were log_e transformed to meet the parametric assumptions of normality and homoscedasticity of residual distribution, and to avoid impossible predicted values below zero. We used the Akaike Information Criterion (AIC, Burnham and Anderson, 2002) to compare models. As all Δ AIC values between two models differed by more than 2, the model with the lowest AIC was selected. Statistical analyses were performed with R version 3.4.3 (R Core Team, 2017) and maps were created with ArcGIS Pro version 2.5.0 (ESRI, 2019). Means are reported with standard deviation (mean \pm SD) unless reported otherwise.

Results

Mean Hg concentrations in body and head feathers, by species and by breeding site are summarized in Table 1 (details per year in Table SI 1) and are further summarized by species and region in Table 2.

Hg contamination during the breeding period

Overall, the mean Hg concentration during the breeding season was $1.20 \pm 0.83 \mu\text{g.g}^{-1}$ dw. The highest mean Hg concentration during the breeding season was observed in the West Atlantic ($1.70 \pm 0.77 \mu\text{g.g}^{-1}$ dw; median = $1.59 \mu\text{g.g}^{-1}$ dw; range = $0.41\text{-}6.97 \mu\text{g.g}^{-1}$ dw), followed by the Pacific ($1.55 \pm 1.37 \mu\text{g.g}^{-1}$ dw; median = $0.95 \mu\text{g.g}^{-1}$ dw; range = $0.32\text{-}6.99 \mu\text{g.g}^{-1}$ dw) and the East Atlantic ($0.75 \pm 0.26 \mu\text{g.g}^{-1}$ dw).

¹ dw; median = 0.71 $\mu\text{g}\cdot\text{g}^{-1}$ dw; range = 0.28-2.29 $\mu\text{g}\cdot\text{g}^{-1}$ dw - Fig. SI 2A). The lowest mean Hg concentration during the breeding period was measured in crested auklets (1.00 ± 0.51 $\mu\text{g}\cdot\text{g}^{-1}$ dw) with the highest were observed in rhinoceros auklets (3.47 ± 1.63 $\mu\text{g}\cdot\text{g}^{-1}$ dw – Fig. SI 3A).

Hg concentrations during the non-breeding period

On average we found Hg concentrations during the non-breeding period to be 3.60 ± 2.40 $\mu\text{g}\cdot\text{g}^{-1}$ dw. The highest mean Hg concentrations during the non-breeding season was observed in the West Atlantic (5.42 ± 2.52 $\mu\text{g}\cdot\text{g}^{-1}$ dw; median = 5.21 $\mu\text{g}\cdot\text{g}^{-1}$ dw; range = 1.21-15.44 $\mu\text{g}\cdot\text{g}^{-1}$ dw) followed by the Pacific (2.64 ± 2.07 $\mu\text{g}\cdot\text{g}^{-1}$ dw; median = 2.06 $\mu\text{g}\cdot\text{g}^{-1}$ dw: range = 0.26-11.65 $\mu\text{g}\cdot\text{g}^{-1}$ dw) and the East Atlantic (2.52 ± 1.46 $\mu\text{g}\cdot\text{g}^{-1}$ dw; median = 2.01 $\mu\text{g}\cdot\text{g}^{-1}$ dw; range = 0.47-8.77 $\mu\text{g}\cdot\text{g}^{-1}$ dw – Fig. SI 2B). Mirroring the breeding season results, the lowest mean Hg concentration during the non-breeding season was measured in crested auklets (1.45 ± 0.58 $\mu\text{g}/\text{g}$ dw) while the highest observed was in rhinoceros auklets (6.89 ± 2.04 $\mu\text{g}/\text{g}$ dw – Fig. SI 3B). The number of individuals at risk per species and colonies is presented in Figure 3.

Hg seasonality

When testing for seasonal and species variation in Hg concentrations, the model that fitted the data better was composed of the interaction between species and season (AIC model with interaction = 2,433.08; AIC model without interaction = 2,667.68). This model indicated higher Hg concentrations during the non-breeding period, varying according to species. The largest seasonal differences were found in Brünnich's guillemots (estimate \pm SE = 1.15 ± 0.02), common guillemots (1.12 ± 0.02), little auks (0.82 ± 0.03) and razorbills (1.50 ± 0.07); while no significant seasonal variation in Hg concentrations was found in ancient murrelets (0.36 ± 0.21), crested auklets (0.41 ± 0.13), least auklets (0.23 ± 0.09), rhinoceros auklets (0.75 ± 0.11) and tufted puffins (0.29 ± 0.14). When testing for seasonal differences depending on regions, the model fitting better the data was composed of the interaction between seasons and regions (AIC model with interaction = 2,643.84; AIC model without interaction = 2,815.64). This model indicated higher Hg concentrations during the non-breeding period, varying

according to regions. The largest seasonal Hg difference was found in the West Atlantic (1.13 ± 0.02) and the East Atlantic (1.13 ± 0.02) followed by the Pacific (0.57 ± 0.04).

We tested for spatial and seasonal differences in Hg concentrations specifically for Brünnich's and common guillemots, the two species being sampled in all three regions. For Brünnich's guillemots, the model with an interaction between the type of feather and the region best fitted the data (AIC model with interaction = 943.16; AIC model without interaction = 1,014.28). It indicated higher Hg concentrations during the non-breeding period with variations being region-specific in this species. The largest seasonal difference in the West Atlantic (estimate \pm SE = 1.30 ± 0.03), followed by the East Atlantic (1.14 ± 0.03) and the Pacific (0.63 ± 0.06). For common guillemots, similar results were found with seasonal differences in Hg concentrations being different across the three regions (AIC model with interaction = 363.37; AIC model without interaction = 379.03), and following the same pattern of largest differences in the West Atlantic (1.12 ± 0.03) and the East Atlantic: 1.12 ± 0.03), followed by the smallest differences in the Pacific (0.83 ± 0.06).

Discussion

To date, limited data suggest considerable seasonal variation in seabird Hg concentrations, with higher contamination reported from the non-breeding period when seabirds are often outside of Arctic regions (Fort et al., 2014a; Joiris et al., 1997; Lavoie et al., 2014). This non-breeding Hg contamination can subsequently be associated with negative carry-over effects: birds with higher non-breeding Hg concentrations had smaller eggs and experienced enhanced contamination during the breeding period (Fort et al., 2014, Lavoie et al., 2014). As predicted, we found clear seasonal differences in Hg concentrations for most study sites and species, confirming higher Hg exposure during the non-breeding period for some species. However, populations of ancient murrelets, crested and least auklets, and tufted puffins from the Pacific Region, in addition to the little auks from the northwest Atlantic (Thule) presented low or no seasonal variation (BF/HF ratio >0.70) in their Hg concentrations.

Three non-exclusive hypotheses could explain Hg seasonal variations in migratory species: 1) the occurrence of a dietary shift between seasons, 2) different food chain lengths at breeding and non-breeding areas, 3) contrasting environmental contamination between sites. Additionally, higher energy

requirements (e.g. during storms weather, increased thermoregulation costs) and thus enhanced food intakes during the winter period (Dunn et al., 2020; Fort et al., 2009) could also contribute to differences in Hg concentrations between seasons.

Seasonal and interspecies differences

As food represents the main pathway of Hg exposure for seabirds and other marine predators, their contamination is closely related to their diet and trophic position (Carravieri et al., 2018), with Hg concentrations increasing along the food chain. Therefore, seasonal dietary shifts towards higher trophic level prey or the use of longer food chains during the non-breeding season may lead to higher Hg concentrations (Provencher et al., 2014). Nitrogen stable isotope ratios are the most common proxy used to study the diet and trophic level of marine predators. It has been widely used in the Arctic marine systems (e.g. Hobson et al., 2002), including characterization of Hg biomagnification processes (Atwell et al., 1998; Campbell et al., 2005; Pomerleau et al., 2016). However, nitrogen stable isotopes in feathers reflect bird trophic status during the feather growth period only (which lasts a few weeks), while Hg concentrations represent their contamination during the intermolt period (lasting several months). Hence, nitrogen stable isotopes in feathers to interpret Hg concentrations and their seasonal variations require stable diet over time and cannot be used in this study (Bond, 2010). In addition, comparing stable isotope values at large spatial scales requires accurate information about spatial baseline variation (e.g. isoscapes) (Hobson et al., 2012), which is currently still largely missing for the non-breeding period and the Arctic and subarctic regions. The diet of Arctic seabirds is generally well documented during the breeding season. In contrast, information about their winter diet is scarce, and only available for five of the nine studied species (Brünnich's and common guillemots, little auks, ancient murrelets and rhinoceros auklets) and only from a few wintering areas (see references below).

Our results showed that for most populations, higher Hg concentrations were found during the non-breeding period, in contrast with reported dietary shifts towards lower trophic levels during that period. For example, Brünnich's guillemots feed on both fish and zooplankton during the breeding season, both at their Atlantic and Pacific colonies (Gaston and Jones 1998). During the non-breeding period, they prefer low trophic level prey (mostly invertebrates and fish in the Atlantic and mostly

invertebrates in the Pacific, Gaston and Jones, 1998), although spatial differences might occur (Karnovsky et al., 2008; Linnebjerg et al., 2013; Moody and Hobson, 2007). Similarly, common guillemots (mostly feeding on small fish) or razorbills (feeding on fish during summer and on a mixed diet of fish and zooplankton during winter) are known to have a constant diet or a switch to lower trophic level prey in winter, respectively (e.g. Huettmann et al., 2005; Lilliendahl, 2009; Lorentsen et al., 1999). Again, this contrast with the higher Hg concentrations measured in the present study during the non-breeding period; as feeding lower in the food web during the winter would result in lower Hg exposure. A few studies investigated food web structure in the Arctic (e.g., Baffin Bay; Hobson and Welch, 1992; Linnebjerg et al., 2016) and subarctic marine systems (e.g., continental shelf off Newfoundland; Sherwood and Rose, 2005). These findings suggest slightly decreasing food chain lengths toward southern locations and again contrast with higher Hg contamination in birds during the non-breeding period in the sub-Arctic and North Atlantic, suggesting other processes. A larger food intake due to higher energy requirements during the cold season as well as less excretion of Hg in winter due to less plumage exchange/formation at this time of the year may serve as a possible explanation of higher Hg concentration in tissues of the studied seabirds recorded during the non-breeding period/winter. Only in rhinoceros auklets a seasonal change in diet [i.e. switch from a mix of fish and zooplankton in summer (Hobson et al., 1994) to fish and squids in winter (Takahashi et al., 2015)] could explain observed enhanced Hg contamination during the non-breeding period.

Nonetheless, much of the published evidence contrasts with our data, and collectively suggests that seasonal dietary changes alone cannot explain observed seasonal changes in Hg contamination and increased concentrations measured in most alcid species and areas during the non-breeding period. Thus, this may suggest a dominant role of bird migration strategies and distribution, both during summer and winter.

Seasonal and spatial differences

Our results highlight spatial differences in seabird Hg contamination during the non-breeding period, as reflected by HF measurements. We found higher mean Hg concentrations in seabirds populations breeding in the Northwest Atlantic (which are likely overwintering in the Labrador Sea,

McFarlane, Tranquilla et al., 2014) compared to the Northeast Atlantic seabirds, overwintering farther East (Fort et al., 2013; Stein et al., 2013 – Table 2). Such results are in agreement with the earlier reported higher Hg concentrations in little auks overwintering off Newfoundland (Fort et al. 2014) and suggest a Hg hotspot in the Labrador Sea.

In only a few cases, we found low or no seasonal variation in Hg concentrations, suggesting a high Hg contamination year-round. This almost exclusively occurred in Pacific species and populations (i.e. ancient murrelets, crested and least auklets) as well as in one little auk population breeding in northwest Greenland (Northwest Atlantic). This absence of seasonal variation in the Pacific region could be due either to a similar exposure at both their breeding and non-breeding sites or to a resident strategy of birds staying close to the breeding grounds year-round. For example, a few observations suggested that most of the least auklets are resident year-round, in ice-free areas (Gaston and Jones, 1998). Further investigations combining migratory movements and Hg contamination in Pacific seabirds are thus urgently required to understand seabird vulnerability to Hg in this region (see Fleishman et al., 2019). In the case of the little auk population, higher Hg concentrations are found in northwest Greenland compared to other Arctic regions, a result commonly found in seabirds and other marine top predators (Albert et al., 2019a; AMAP, 2011b). The high concentrations found in that population might be explained by close elevated concentration at both their breeding and non-breeding areas off Newfoundland (Fort et al., 2013b).

Finally, our results show spatial differences in bird Hg contamination and seasonal variations between the Pacific and Atlantic regions, with lower concentrations and smaller variation found in the Pacific (refer to figure, table, or both). Brünnich's and common guillemots, which are present in both regions also showed smaller seasonal differences and lower non-breeding Hg concentrations in the Pacific than in the Atlantic (see Table 2), suggesting differences in Hg environmental contamination between both regions. The Arctic Ocean is a sink for contaminants such as Hg, with inflows from both atmospheric and oceanic currents, as well as rivers (Sonke et al., 2018a). Nevertheless, important outflows of Hg also occur from the Arctic to lower latitudes (AMAP, 2011). Because the Atlantic region is widely opened with more complex and numerous atmospheric and oceanic fluxes than is the Pacific region, oceanic outflows of Hg (e.g. through the Arctic Canadian Archipelagoes and Fram Strait;

Outridge et al., 2008) could partly explain the higher concentrations in Hg found in species from the Western Atlantic region compared to the Pacific region.

Non-breeding inter-individual variability

In addition to species and population differences, our results show a high inter-individual variability in Hg concentrations during the non-breeding period (Table 2, Fig. 2, Fig. SI 5). Arctic seabirds are central place foragers, feeding mostly in the vicinity of their colony during the breeding season (Jakubas et al., 2016; Linnebjerg et al., 2015; Wilkinson et al., 2018). However, during the non-breeding period, they migrate towards the open seas to spend the winter within or outside the Arctic (McFarlane Tranquilla et al., 2013). Recent tracking studies highlight how individuals from the same population might engage in contrasting migratory strategies, and thus occupy very different wintering areas (Fort et al., 2013b; Frederiksen et al., 2016) where exposure to Hg and other pollutants could be substantially different (Leat et al., 2013; Miller et al., 2020). For instance, Brünnich's guillemots breeding at Coats Island and at the Gannet Islands (Northwest Atlantic) adopt variable individual strategies displaying wintering areas ranging from the Labrador Sea to Newfoundland (McFarlane Tranquilla et al., 2013). This finding is consistent with the observed increased inter-individual variability in Hg concentrations for non-breeding Brünnich's guillemots. In comparison, common guillemots from the same colonies show lower inter-individual variability in their non-breeding distribution as individuals winter close to their breeding colonies off Newfoundland (McFarlane Tranquilla et al., 2013). Similarly, this was reflected in our results by lower variability in Hg concentrations for that species. These inter-individual variations in both Hg concentrations and large-scale distribution strengthen the hypothesis that individual migratory strategies play an important role in Hg exposure and call for further studies linking individual movements to contamination risks.

Hg toxicity and risks for non-breeding Arctic seabirds

There is limited information about toxicity thresholds for seabird feathers. Nevertheless, Hg concentrations above 5 µg/g dw in feathers are usually considered to be associated with deleterious health and reproductive effects (Eisler, 1987). In addition, elevated Hg contamination during the non-

breeding period can exert carry-over effects on reproductive parameters in the following breeding season (e.g. Fort et al., 2014). Our results demonstrate that various populations and species of Arctic alcids had Hg values close to or above 5 µg/g dw (Fig. 3). In total, 26% of the sampled individuals for which non-breeding Hg was measured exceeded this toxicity threshold. These include rhinoceros auklets breeding in the Gulf of Alaska, and common and Brünnich's guillemot populations breeding in the Northwest Atlantic (Table 3, Fig. 3). This latter region has previously been associated with elevated Hg risk in seabirds. For instance, the investigation of Hg concentrations in blood of Brünnich's guillemot breeding at Coats Island (Canadian Arctic, Northwest Atlantic) showed that above ~90% of the sampled individuals were associated with at least a moderate risk (see Dietz et al., 2019 for a review).

Hence, many seabird species and populations might be at high risk of Hg contamination that negatively impacts their reproduction, with potential long-term impacts on population dynamics (Goutte et al., 2014a, 2014b). Arctic seabird populations are experiencing different trends according to their distribution (e.g., Frederiksen et al., 2016), but the role played by environmental pollution has been largely overlooked. Therefore, there is a need to include exposure to pollutants, notably during the non-breeding period, in population dynamic studies and conservation programs.

Conclusion

The present study is the first to investigate seabird seasonal Hg contamination at a large, pan-Arctic scale. Our results demonstrate that the non-breeding period is critical for Hg contamination of piscivorous and planktivorous seabirds. This period might represent an important risk for these sensitive populations, especially those breeding in the West Atlantic Arctic, and to a lesser extent in the Pacific Arctic. This study demonstrates that close monitoring should be pursued to understand both short- and long-term implications of Hg exposure on seabird population dynamics. Our study also shows that higher non-breeding contamination most likely depends on both individual and population, as well as species-specific migratory strategies and year-round spatial distribution. We therefore call for further studies linking non-breeding distribution of seabirds and Hg contamination at the individual and population scales. This would allow us to better understand which wintering areas are associated with higher Hg contamination and thus pose higher risks for Arctic seabirds. Further studies including

energetic modelling are also required to better understand the role of increased energy requirements on Hg seasonal changes. Therefore, there is a need for more integrative spatial ecotoxicology studies at multiple scales, combining tracking technologies, Hg measurements, dietary investigations, and including a large range of colonies and species.

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Table 1: Species and region- specific Hg concentrations (mean \pm SD; in $\mu\text{g}\cdot\text{g}^{-1}$ dw) measured in body feathers (BF – representing exposure during the breeding season) and head feathers (HF – representing exposure during the non-breeding period). Breeding: non-breeding period Hg concentrations (mean \pm SD BF : HF) calculated from individual values ; n = samples size

Species	Region	Sampling site	BF mean \pm SD in $\mu\text{g}/\text{g}$ dw (n)	HF mean \pm SD in $\mu\text{g}/\text{g}$ dw (n)	BF : HF	n
Ancient murrelet	Pacific Region	Talan Island	2.64 \pm 1.00	3.69 \pm 1.18	0.81 \pm 0.52	5
Brünnich's guillemot	Pacific Region	Saint George	0.90 \pm 0.20	3.42 \pm 1.10	0.29 \pm 0.11	10
		Saint Lawrence	0.77 \pm 0.33	1.43 \pm 0.89	0.79 \pm 0.49	40
	East Atlantic	Alkefjellet, Spitsbergen	0.63 \pm 0.20	1.70 \pm 0.48	0.39 \pm 0.14	32
		Bjørnøya	0.50 \pm 0.09	2.91 \pm 1.73	0.24 \pm 0.15	48
		Cape Flora	0.74 \pm 0.21	1.64 \pm 0.35	0.47 \pm 0.16	61
		Cape Gorodetskiy	0.66 \pm 0.10	2.06 \pm 0.39	0.33 \pm 0.07	19
		Hornøya, N Norway	0.75 \pm 0.20	2.45 \pm 1.08	0.34 \pm 0.13	59
		Isfjorden, Spitsbergen	1.17 \pm 0.37	6.05 \pm 1.56	0.21 \pm 0.08	32
		Kara Gate	0.64 \pm 0.20	1.65 \pm 0.52	0.41 \pm 0.13	40
		Oranskyi islands	0.70 \pm 0.20	1.79 \pm 0.35	0.39 \pm 0.10	34
	West Atlantic	Coats Island	1.63 \pm 0.33	8.22 \pm 3.27	0.23 \pm 0.12	19
		Gannet Islands	2.39 \pm 0.95	9.24 \pm 1.73	0.27 \pm 0.12	41
		Grimsey, Iceland	1.52 \pm 0.24	5.56 \pm 1.04	0.28 \pm 0.08	11
		Jan Mayen	1.63 \pm 0.32	5.70 \pm 1.77	0.32 \pm 0.14	65
Kippaku		1.79 \pm 0.44	7.38 \pm 2.32	0.26 \pm 0.07	20	
Langanes		1.38 \pm 0.21	4.83 \pm 1.14	0.30 \pm 0.08	35	
Common guillemot	Pacific Region	Thule	2.62 \pm 0.72	8.14 \pm 2.73	0.35 \pm 0.12	17
		Saint Lawrence	0.59 \pm 0.17	1.42 \pm 0.63	0.48 \pm 0.21	36
	East Atlantic	Bjørnøya	0.67 \pm 0.13	1.95 \pm 0.55	0.37 \pm 0.12	48
		Cape Gorodetskiy	0.64 \pm 0.14	1.79 \pm 0.42	0.37 \pm 0.08	40
		Hjelmsøya	0.85 \pm 0.20	2.85 \pm 0.94	0.32 \pm 0.10	58
		Sklinna	0.94 \pm 0.35	3.17 \pm 0.90	0.31 \pm 0.10	20
	West Atlantic	Faroes	2.26 \pm 1.05	4.91 \pm 2.37	0.54 \pm 0.26	8
		Funk Island	2.21 \pm 0.39	6.83 \pm 1.11	0.33 \pm 0.09	23
		Gannet Islands	2.17 \pm 0.51	6.44 \pm 1.25	0.35 \pm 0.11	32
		Gull Island	2.04 \pm 0.60	5.90 \pm 1.47	0.38 \pm 0.17	40
	Jan Mayen	0.98 \pm 0.36	3.88 \pm 1.65	0.28 \pm 0.11	61	
	Langanes	1.11 \pm 0.39	3.70 \pm 0.92	0.31 \pm 0.10	37	

Crested auklet	Pacific Region	Saint Lawrence	1.00±0.51	1.45±0.58	0.75±0.32	14
Least auklet	Pacific Region	Saint George	1.58±1.22	2.38±1.29	0.72±0.35	10
		Saint Lawrence	2.56±1.13	2.75±1.01	0.96±0.31	20
Little auk	East Atlantic	Bjørnøya	0.82±0.19	2.57±1.12	0.37±0.15	31
		Hooker Island	0.81±0.21	1.73±0.92	0.57±0.26	67
		Hornsund	0.82±0.35	2.80±1.01	0.34±0.23	37
		Isfjorden, Spitsbergen	0.90±0.16	2.97±0.70	0.31±0.03	5
	West Atlantic	Kap Hoegh	1.34±0.44	2.69±0.70	0.52±0.18	64
		Thule	2.08±0.52	2.15±0.40	1.00±0.30	10
Razorbill	East Atlantic	Hornoya, N Norway	0.79±0.31	4.68±1.54	0.20±0.14	39
	West Atlantic	Gannet Islands	2.89±1.84	4.84±1.55	0.65±0.47	13
Rhinoceros auklet	Pacific Region	Middleton	3.47±1.63	6.89±2.04	0.54±0.33	19
Tufted puffin	Pacific Region	Aiktak Island	3.12±1.42	4.04±1.35	0.80±0.26	11

3

4 Table 2: Geographical variations in Hg concentrations (mean \pm SD - $\mu\text{g.g}^{-1}$ dw) measured in body feathers (BF – representing
 5 exposure during the breeding season) and head feathers (HF – representing exposure during the non-breeding period). Breeding:
 6 non-breeding period Hg concentrations (mean \pm SD BF : HF) calculated from individual values ; n = samples size

Species	Regions	Mean \pm SD BF	Mean \pm SD HF	BF : HF	n
Ancient murrelet	Pacific Region	2.64 \pm 1.00	3.69 \pm 1.18	0.81 \pm 0.52	5
Brünnich's guillemot	Pacific Region	0.80 \pm 0.31	1.83 \pm 1.23	0.69 \pm 0.49	50
	East Atlantic	0.72 \pm 0.27	2.45 \pm 1.61	0.36 \pm 0.16	325
	West Atlantic	1.83 \pm 0.67	6.84 \pm 2.55	0.29 \pm 0.12	208
Common guillemot	Pacific Region	0.59 \pm 0.17	1.42 \pm 0.63	0.48 \pm 0.21	36
	East Atlantic	0.76 \pm 0.23	2.37 \pm 0.90	0.34 \pm 0.11	166
	West Atlantic	1.60 \pm 0.74	5.03 \pm 1.88	0.33 \pm 0.14	201
Crested auklet	Pacific Region	1.00 \pm 0.51	1.45 \pm 0.58	0.75 \pm 0.32	14
Least auklet	Pacific Region	2.24 \pm 1.23	2.63 \pm 1.10	0.88 \pm 0.34	30
Little auk	East Atlantic	0.82 \pm 0.24	2.24 \pm 1.10	0.46 \pm 0.25	140
	West Atlantic	1.44 \pm 0.51	2.61 \pm 0.69	0.58 \pm 0.25	74
Razorbill	East Atlantic	0.79 \pm 0.31	4.68 \pm 1.54	0.20 \pm 0.14	39
	West Atlantic	2.89 \pm 1.84	4.84 \pm 1.55	0.65 \pm 0.47	13
Rhinoceros auklet	Pacific Region	3.47 \pm 1.63	6.89 \pm 2.04	0.54 \pm 0.33	19
Tufted puffin	Pacific Region	3.12 \pm 1.42	4.04 \pm 1.35	0.80 \pm 0.26	11

7

Fig. 1: Breeding colonies locations of the studied piscivorous and planktivorous alcids and sampling sites. The study sites were divided into three regions.: Pacific, East Atlantic, West Atlantic. Colonies are colored based on the areas they belong to. Sub, Low and High Arctic are represented following the Conservation of Arctic Flora and Fauna delimitations (CAFF, 2001).

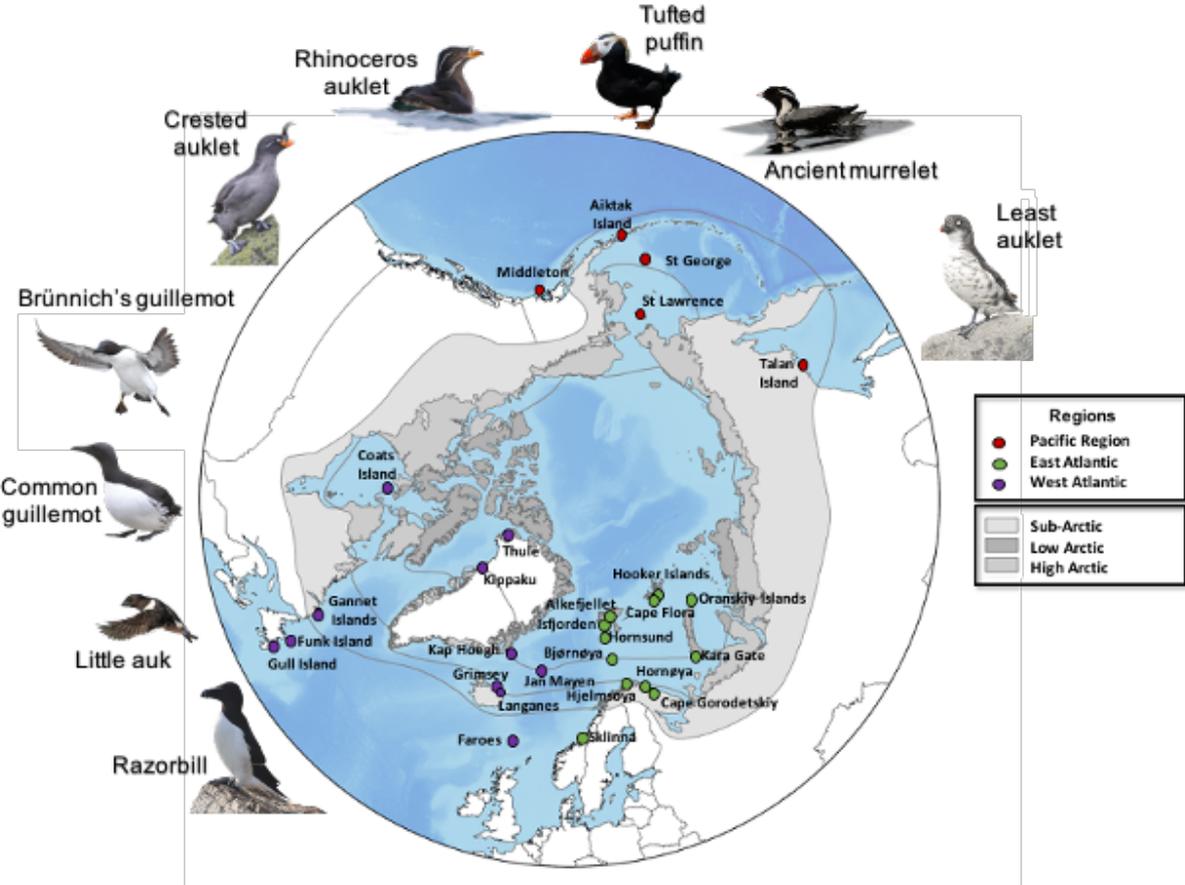


Fig. 2: Hg concentrations during the breeding (body feathers - green) and non-breeding period (head feathers - red) for each study colony. Mean Hg concentrations and SD per species and colony are presented in Table 1 and Fig. SI 4 and 5. Sub, Low and High Arctic are represented following the Conservation of Arctic Flora and Fauna delimitations (CAFF, 2001)

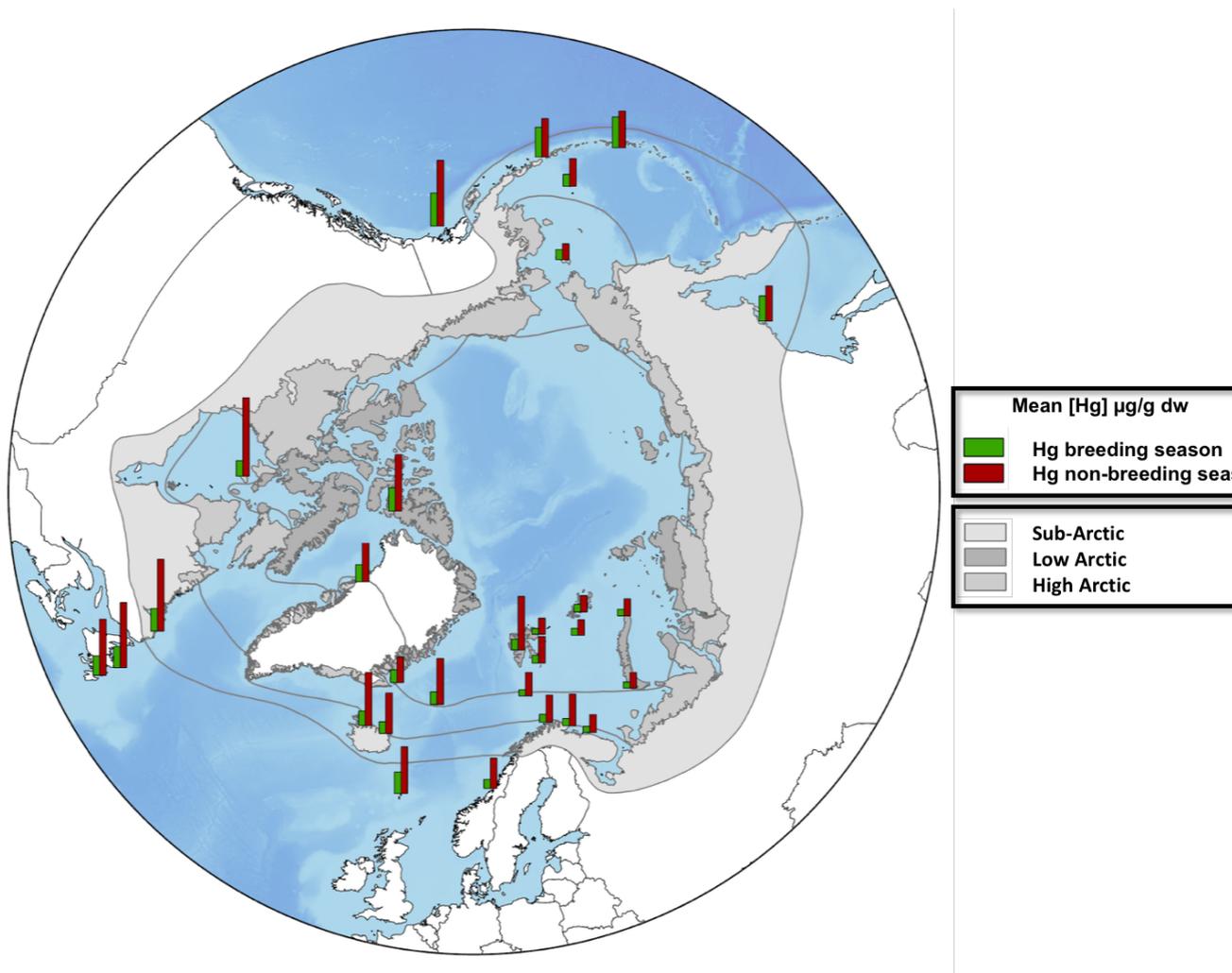
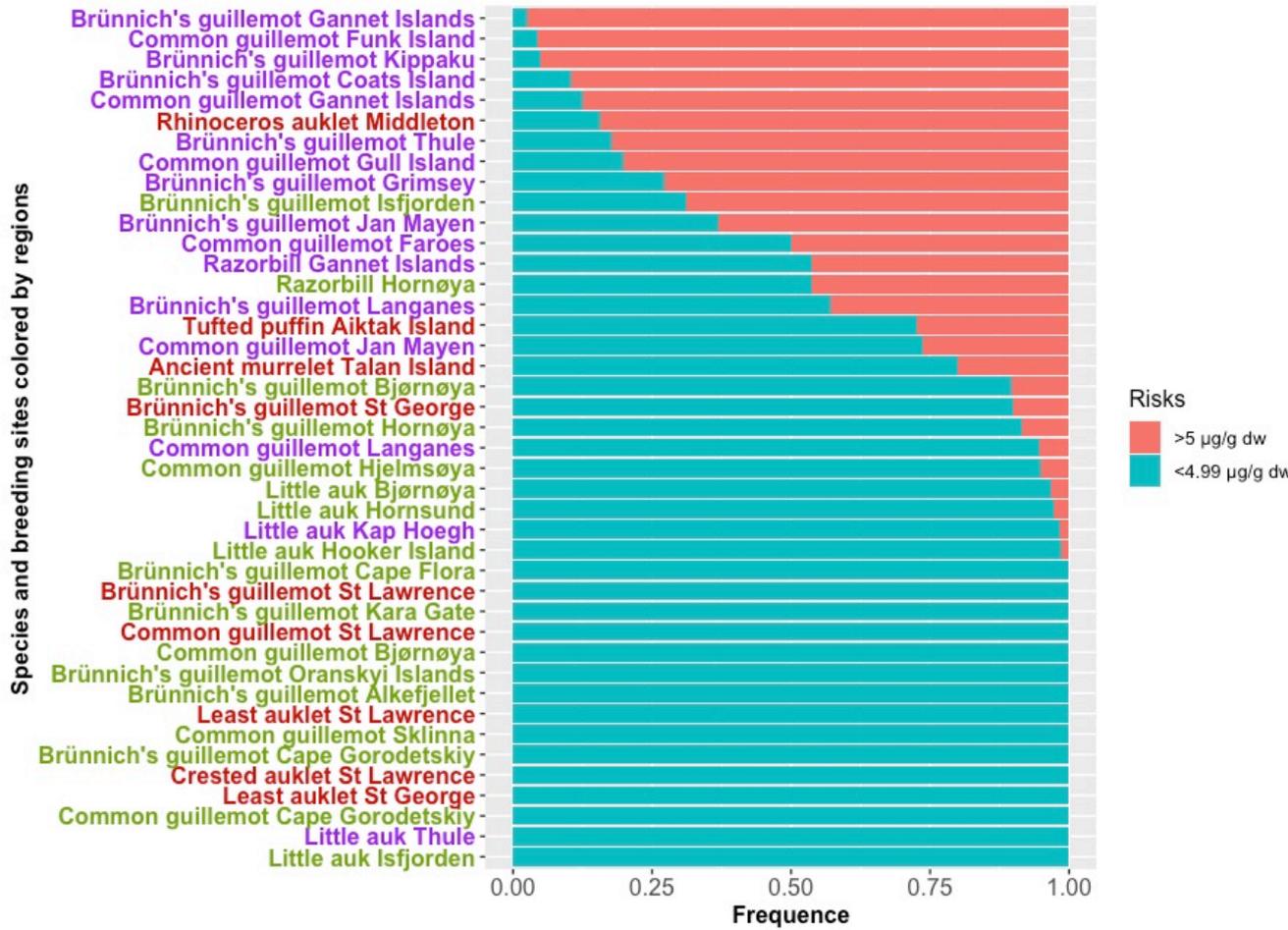


Fig. 3: Frequency of individuals below (blue) or above (red) the toxicity threshold (Eisler, 1987) of 5 $\mu\text{g/g}$ dw in head feathers (representing exposure during the non-breeding season). The species and breeding colony names are colored by regions (see Fig. 1).



Paper 4

Interannual variations in migratory movements impact individual seabird exposure to contaminants.

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Abstract:

Migratory seabirds are exposed to various pollutants throughout their annual cycle. Among them, mercury (Hg) is of particular concern given potentially large impacts on individual physiology and reproduction and long-term effects on population dynamics. Recent studies suggest that winter is a critical period for seabirds when exposure to Hg can be much higher than other times of year. However, individuals within many species have different migration strategies that potentially affect their Hg exposure. Here, we combined multi-year individual tracking data and Hg measurements from six Arctic seabird species that exhibit different migration strategies. Our primary aim was to investigate inter-annual variations in individual winter contamination to Hg, and whether wintering site fidelity is associated with repeated contamination in comparison to individuals and species showing variable migratory patterns. Our results show that individuals with high fidelity for their wintering grounds tend to have relatively constant exposure to Hg compared to individuals with low fidelity, suggesting an effect of their migration on Hg exposure. Further, we found that the directional change in wintering areas between years influenced their Hg exposure. More specifically, individuals migrating to the northwest direction of their previous wintering ground tend to have higher Hg exposure compared to the eastern directions. Those results suggest a spatial difference in Hg concentration throughout the North-Atlantic Arctic and more specifically an east to west gradient increase in Hg concentrations. This will require more large-scale ecotoxicological studies at a better resolution in order to confirm such trend and highlight potential hotspots of Hg contamination in order to fully apprehend the risks that Hg and other pollutants represent for seabirds and other marine organisms.

Introduction

Mercury (Hg) is a highly toxic pollutant for humans and wildlife causing severe health impairments, even at low concentrations (e.g. Wolfe et al. 1998, Tan et al. 2009, Dietz et al. 2019). Since the industrial revolution, anthropogenic activities have been responsible for an increase in Hg

concentrations in the environment (e.g. threefold increase in surface marine waters ; Lamborg et al. 2014). Hg is thus of major environmental concern worldwide, yet its spatial distribution is highly heterogeneous at the global scale (Zhang et al., 2020). For instance, within marine species where the highest Hg levels are measured, Dietz et al. (2000) found that polar bears (*Ursus maritimus*) from Northwest Greenland had Hg concentrations approximately 10 times higher than individuals from Northeast Greenland. A negative latitudinal gradient in Hg concentrations has also been found in the southern hemisphere in 35 albatross populations (Cherel et al., 2018). Therefore, species distribution can lead to highly different Hg exposure and contamination, potentially resulting in contrasting risks for individual health (Ackerman et al., 2016; Dietz et al., 2019). Among marine top predators, seabirds show some of the highest Hg concentrations due to biomagnification (Atwell et al. 1998) with deleterious effects on behavior, physiology or reproduction reported in some species and populations. For example, amongst adult male black-legged kittiwakes (*Rissa tridactyla*) breeding in Svalbard, individuals with the highest Hg concentrations were more likely to neglect their eggs (Tartu et al., 2015). Similarly, high Hg concentrations have been related to a reduction of hatchability and clutch size (Ackerman et al., 2016 for a review; Dietz et al., 2013a and), in addition to a decline in breeding success and population growth rate (Goutte et al., 2014b). Understanding the relationship between bird distributions and exposure to Hg is therefore important for the conservation of these species for which many populations are declining and deserve special attention (Croxall et al., 2012; Paleczny et al., 2015).

Seabirds are typically migratory species, many travelling hundreds to thousands of kilometers every year to reach their wintering grounds (e.g. Egevang et al., 2010, Frederiksen et al., 2016; McFarlane Tranquilla et al., 2014). However, these migratory strategies can be highly variable between individuals, conditioning their distribution and thus their exposure to Hg. First, different birds from the same population can use contrasting wintering areas. For instance, red-legged kittiwakes (*Rissa brevirostris*) breeding at Saint George Island (Pribilof Islands) overwinter from the Eastern Bering Sea to the Sea of Okhotsk, with higher Hg concentrations measured in individuals distributed in southern areas (Fleishman et al., 2019). Second, each individual can itself have different interannual migratory strategies and thus contrasting fidelity to its wintering grounds, with species showing high (e.g. common

eiders *Somateria mollissima*, Atlantic puffin *Fratercula arctica*, European shags *Phalacrocorax aristotelis*; Guilford et al. 2011, Petersen et al. 2012, Grist et al. 2014, Hanssen et al. 2016), low (e.g. Thin-billed prion *Pachyptila belcheri*; Quillfeldt et al. 2010) or variable (e.g. Brünnich's *Uria lomvia* and common guillemots *Uria aalge*, Northern fulmars *Fulmarus glacialis*; Merkel et al. *In press*, Hatch et al. 2010, McFarlane Tranquilla et al. 2014, Orben et al. 2015) individual fidelity to wintering grounds. Together, these different migratory patterns and overwintering strategies may affect the long-term exposure of seabirds to Hg and differentially impact their health, reproduction and consequently their population dynamics.

By combining individual tracking data with Hg analyses and focusing on some of the most abundant Arctic seabird species, we studied bird wintering ground fidelity across years and tested the hypothesis that winter migration and wintering ground fidelity condition their Hg contamination. More specifically, we predicted that individuals with a low fidelity to their wintering ground from one year to the other show more variable Hg contamination in comparison to faithful ones, with an effect of these wintering locations.

Material and methods

Species, study sites and sampling collections:

From June 2014 to July 2016, black-legged kittiwakes (n=13), Brünnich's guillemots (n=65), common guillemots (n=39), little auks (*Alle alle*, n=6), common eiders (females, n=20) and northern fulmars (n=38) were equipped with geolocators (GLS) at 17 breeding colonies widely distributed from Iceland to Russia (Fig. 1), for two or three years (details in Table S1). GLSs were retrieved every following breeding seasons. Geolocators are low-weight instrument that record ambient light-levels over multiple years, which later can be converted into geographical locations (Wilson et al., 1992). However, the positional precision derived from light-level data is considered low (Lisokvski et al., 2012), and is mostly suited for studies of large-scale movements and distribution.

In order to investigate individual Hg contamination during winter (i.e. non-breeding period – late fall and winter) periods, feathers were collected at deployment and recapture of GLS. Feathers are

useful tissues to monitor Hg contamination during the winter period (Albert et al., 2019) as seabirds, during the molt, excrete a large proportion of accumulated Hg into their feathers. Hence, feathers inform about Hg accumulated by an individual between two molting sequences, and specifically during the non-breeding period according to selected feathers and bird molting pattern (see Albert et al, 2019). For example, Alcids undergo a rapid total molt after the breeding season (during September or October) resulting in the winter plumage and a partial molt (cheek, neck, throat) at the end of the winter period (during March or April) resulting in the nuptial plumage. Black-legged kittiwakes (as most Larids) undergo a complete molt after the breeding period and a partial (e.g. back, head) molt at the end of the winter period (Cramp and Simmons, 1983). Female common eiders undergo a partial molt (e.g. body) after the breeding season and a complete molt at the end of the winter period (Baldassarre, 2015; Goudie et al., 2020). Hence, head, back and belly feathers provide information on Hg contamination specifically during the winter period in alcids, kittiwakes and eiders, respectively. Northern fulmars undergo one total molt per year after the breeding season lasting from mid-August to late October (Grissot et al. 2020). Recent studies indicated that Hg concentrations in fulmar body feathers reflect inter-individual variations in Hg contamination during the non-breeding period (Quinn et al., 2016).

We collected head feathers on Brünnich's guillemots, common guillemots and little auks, and body feathers on common eiders (belly), black-legged kittiwakes (back) and northern fulmars (belly) to assess Hg contamination during the non-breeding period. Feathers were stored in plastic bags at ambient temperature until Hg analyses.

Spatial analyses

GLS light-level data were first converted into positional data by identifying the timing of twilights, using a threshold method, from which we estimated two daily latitudes from the length of the night and day, and two daily longitudes from time of apparent midnight and noon, following procedures described in Merkel et al. (2016) and Bråthen et al. (2020). For individuals located above 70 degrees north, positions could not be obtained for days or weeks around winter solstice because of an apparent absence of daylight above this latitude.

The timing of Arctic seabird molt and migration can vary slightly between breeding sites (Fort et al., 2013b; Frederiksen et al., 2012). Nonetheless, to allow for inter-population comparisons, based on previous investigations (Fort et al., 2013b, 2012a; Frederiksen et al., 2016) and given that the variations in molting timing and migration are limited at the scale of the winter period, this later was homogenized within species. Therefore, we considered that birds were at their wintering ground from November to January for little auks (Fort et al., 2012a), common and Brünnich's guillemots (Fort et al., 2013c; Frederiksen et al., 2016; Merkel et al., 2019), black-legged kittiwakes (Frederiksen et al., 2012) and common eiders (Hanssen et al., 2016). As northern fulmars have been observed returning back to their colonies as early as January, the non-breeding period was defined from November to December in this species (Macdonald, 1980). We then calculated a median winter position for each individual for each year, to be used in further analyses.

Hg analyses

Prior to Hg analyses, feathers were cleaned to remove external contamination. Feathers were plunged into a 2:1 chloroform:methanol solution for three minutes in an ultrasonic bath, rinsed twice in a methanol solution and dried at 45°C for 48 hours. Hg analyses were performed on a ~0.20-1 mg subsample of a pool of three homogenized feathers, using an Advanced Mercury Analyser spectrophotometer (Altec AMA 254 – detection limit of 0.05 ng). The analysis of each sample was repeated (two to three times) until the relative standard deviation for two subsamples was <10%. Mean concentration for these two subsamples was then used for statistical analyses. Prior to Hg analyses, blanks were run and, to ensure the accuracy of measurements, certified reference materials (dogfish liver DOLT-5 and lobster hepatopancreas TORT-2, NRC, Canada) were used every fifteen samples. Hg concentrations are expressed in µg/g dry weight (dw).

Statistical analyses:

Variations in Hg concentrations (hereafter [Hg]) in the different species were analysed as a function of bird fidelity to their wintering grounds. To do so, the difference in [Hg] from one year to the other (Δ [Hg]; 2015-2016, 2015-2017, 2016-2017) was determined at the individual scale, as well as the distance (in km) between individual winter median positions each year (2015-2016, 2015-2017, 2016-2017). Phillips and colleagues (2004) showed that the accuracy of seabird positions using GLS is

186±114 (SD) km. Given this accuracy, the scale of the study area, and although the different study species can be more or less mobile during winter, we considered the limit of 372km to discriminate individuals with high or low fidelity to their wintering ground. Based on this, individual birds fell into the “high fidelity” group when their consecutive winter median positions were <372 km apart. Conversely, birds were considered to belong to the “low fidelity” group when they wintered in distinctly different geographical areas between years (i.e. winter median positions >373 km apart).

We performed Wilcoxon or student (unpaired) tests to investigate differences in Δ [Hg] (using absolute values) between low and high-fidelity groups. Within the low-fidelity group, we used the longitude and latitude of bird winter median positions during the different years to determine the direction from the previous to the newly used wintering ground (North-West, North-East, South-West or South-East). We then used Generalized Linear Models (GLMs) on raw values to test the correlation between the Δ [Hg], the change in their overwintering area (i.e. direction) and the distances (in km) considering all species together. The common eiders were not added to this analysis as all individuals showed high fidelity to their wintering grounds. We used the Akaike’s information criteria (AIC) to select the best model (Burnham and Anderson, 2002). If the difference in AIC between two models is <2, the models are considered to have equal statistical support and in case of nested models, the simplest (i.e. most parsimonious) was preferred. All statistical analyses were performed with RStudio version 1.2.1335 (R Core Team, 2017)

Results

The common eider was the only species where all individuals showed high fidelity to their wintering ground. This was reflected by low variations in Δ [Hg] (Table 1, Fig. S1). All other species showed both a mixture of individuals with high and low fidelity (Table 1, Fig. 2). Individuals sampled for three years presented variable level of fidelity (Table 1). Black-legged kittiwakes, little auks and common guillemots (species for which only two years of tracking information were available) presented an equal number of individuals with either a low or a high fidelity. Most of the Brünnich’s guillemots were faithful to their wintering ground, while most of the northern fulmars showed low fidelity. Overall,

the inter-annual variation in mean [Hg] was significantly different between the high and low fidelity groups, both when common eiders were included ($W = 5649.5, p = 0.0002$) and when they were excluded ($W = 5294.5, p = 0.009$). Individuals experiencing high fidelity to their wintering ground showed less inter-annual variations in Hg concentrations (mean absolute $\Delta[\text{Hg}] \pm \text{SD}$, including eiders = $0.80 \pm 0.81 \mu\text{g/g dw}$; excluding eiders = $0.89 \pm 0.84 \mu\text{g/g dw}$) than individuals with low fidelity to their wintering grounds ($1.19 \pm 0.98 \mu\text{g/g dw}$ - Fig. 2). The lower variations in Hg concentrations for individuals with high winter ground fidelity was observed in all species but the little auk (Fig. S1). This difference between low and high-fidelity individuals was mostly pronounced in black-legged kittiwakes and common guillemots (Fig. S1).

Results of the GLM model selection for seabirds with low fidelity to their wintering grounds are summarized in Table 2. When including all species (but the common eider), we found that the inter-annual variation in [Hg] varied among species and are associated with both the direction and the distance (in km) between two median winter positions (Table 2). Δ Hg variation increased when individuals changed their winter distribution to the northwest direction and decreased when individuals changed to the southwest direction, and these effects were similar among species (i.e. the interaction between direction and species was not selected; Table 2; Fig. 3, Fig. S2). However, the effect of the distance on variation in [Hg] varied among species (as indicated by the selected distance \times species effect in the best model; Table 2). More specifically, Δ [Hg] is positively correlated with the distance for the black-legged kittiwakes (estimate \pm SE = -1.62 ± 1.02) and the Brünnich's guillemots (-0.07 ± 0.47) and negatively correlated with the common guillemots (-0.52 ± 0.45), the northern fulmars (-0.29 ± 0.42) and the little auks (1.19 ± 1.00) (Fig. S3).

Discussion

Seabirds can adopt different non-breeding strategies and show contrasting fidelity to their wintering sites, at species, population and individual scales (refs). Hence, while some birds use the same wintering ground every year, others change and occupy different places from one year to the other. Such different strategies can have large effects on their exposure to environmental stressors, particularly pollutants, which could in turn impact their condition, reproduction and ultimately their population dynamics. By

combining for the first time the use of tracking devices on multiple species with pollutant analyses, we demonstrated that inter-annual fidelity of individuals to their wintering areas affect their Hg contamination, with a major effect of the location of these areas in the North Atlantic and sub-Arctic.

Interannual changes in Hg contamination might be driven by several non-exclusive factors, including different diets, changing migratory strategies between years (resident *vs* migrating, fidelity to wintering sites) or different regions used during winter in the North-Atlantic Arctic. Hg concentrations in Arctic seabirds, like in other predators, are closely related to their diet. For example, previous studies found that Hg concentrations increased in sub-Antarctic seabirds, from those feeding at low trophic levels (i.e. on crustaceans) to those feeding at higher trophic levels (i.e. on fish, squid and carrion consumers) (Carravieri et al., 2014b). During winter, Brünnich's guillemots, common guillemots and northern fulmars generally feed on relatively high trophic levels (Erikstad 1990, Gaston & Jones 1998) compared to black-legged kittiwakes, little auks, (mainly zooplankton; Karnovsky et al. 2008, Fort et al. 2010, Frederiksen et al. 2012, Reiertsen et al. 2014) or common eiders (Goudie et al. 2020) generally feed at lower trophic levels. Therefore, higher Hg concentrations, if explained by diet differences only, should be expected in the two guillemot species and in northern fulmars, followed by black-legged kittiwakes, little auks and common eiders. However, our results showed a different contamination order with the Brünnich's guillemots having the highest mean Hg concentrations, followed by black-legged kittiwakes, little auks and common guillemots and being more contaminated than the northern fulmars and common eiders (Table 3). Such results suggest that interannual variations in winter Hg contamination cannot be explained by the diet alone, but potentially by seabird movements and distribution.

Our study focused on six species breeding at 17 different colonies, showing contrasting fidelity to their wintering grounds. The common eider was the only species for which all individuals showed a high fidelity to their wintering grounds, overwintering year after year in the same areas. This is the case for both eiders breeding in the Low Arctic and resident year-round, and populations from the High Arctic migrating to southernly, but constant, locations during winter (Bustnes and Erikstad, 1993; Hanssen et al., 2016). As a consequence, all individual common eiders in this study showed similar constant and low Hg concentrations from one year to the other. The five other study species

encompassed individuals with variable level of fidelity to their wintering grounds (Table 1). Accordingly, seabirds with low inter-annual fidelity had a higher Δ [Hg] mean than individuals with high inter-annual fidelity, although this result was mostly driven by black-legged kittiwakes and common guillemots (Figs. 2 & S1), confirming an effect of adopted migratory movements on bird winter exposure to Hg, and suggesting spatial variations over the North-Atlantic Arctic.

In addition, in individuals experiencing low fidelity to their wintering grounds, we found that while the effect of the direction was relatively similar between species, the effect of the distance was not. More specifically, regardless of the species, our results indicated that Hg concentrations tended to be higher when individuals migrated northwest of their previous wintering ground. In comparison, Hg concentrations tended to be lower when migrating in the three other directions (Figs. 3 and S2). We found that when the distance to their previous wintering ground increased, Hg concentrations decreased for little auks, common guillemots and northern fulmars, and increased for black-legged kittiwakes and Brünnich's guillemots. Overall, this support the hypothesis on a heterogeneous distribution of Hg in the marine environment at large spatial scale. In the present study, wintering Brünnich's guillemots and northern fulmars covered a large part of the North-Atlantic, while common guillemots, black-legged kittiwakes and little auks were distributed over a part of it only (Fig. 1). With such a large-scale study, seabirds found on the eastern (or western) areas first the first year of our assessment had more chance to migrate in the western (or eastern) areas the following years and such a point was difficult to incorporate into the statistical analysis. But despite these differences, our results showed that their Hg concentrations varied the same way between years according to changes in their distribution, strengthening our interpretation. We therefore believe that our results confirm the role of seabird winter movements and distribution on their exposure to Hg and highlight spatial differences in Hg concentrations through the North-Atlantic Arctic, with an increasing east-west gradient. Previous studies on the Arctic wildlife also suggested a positive east-west Hg gradient during spring and summer (breeding period), with species distributed in the Canadian Arctic being more contaminated than those in the European Arctic (Albert et al., 2019a; AMAP, 2018a; Provencher et al., 2014). The present data set covers a large but fragmented part of the North-Atlantic Arctic. But a more precise dataset could

allow us to confirm and extend such a trend during the winter period in North-Atlantic Arctic marine areas.

Seabirds are exposed to various environmental threats during the winter period (e.g. oil spills, storms, contaminants; Frederiksen et al. 2012, Petersen et al. 2012, Fort et al. 2014, Guéry et al. 2017) which might impact their condition, survival or subsequent reproduction (Anker-Nilssen et al., 2016; Mesquita et al., 2015; Votier et al., 2005). The large-scale distribution of Hg in the marine environment is assumed to be relatively stable over years. Hence, high fidelity to their wintering ground likely expose birds to repeated threats and constant pollution issues associated to this specific area (e.g. human industries), possibly impacting their short-term survival and reproduction with long-term impacts on their population dynamics (Guéry et al., 2019, 2017). On the other hand, seabirds wintering constantly in an area with low level of such threats might only be exposed to limited acute pollution events, which could have severe effects on their survival and subsequent reproduction but limited long-term impacts on their populations. Conversely, seabirds with low fidelity to wintering grounds and winter distribution will exploit contrasting wintering environments with different Hg concentrations years after years (Quillfeldt et al., 2010) which might lower long-term risks associated with Hg contamination. Therefore, the inter-individual variability in Hg concentrations depending on their winter ground fidelity should be taken into account while studying population dynamic of migratory species as it could have large effects on the survival, reproduction and subsequently on population dynamics.

Our study demonstrates that individual migratory strategy and the consistency of occupied wintering grounds affect their exposure to Hg and likely of other contaminants. Given the high diversity of areas used by single individuals over two to three winters only, we urge to extend ecotoxicological studies investigating seabird winter contamination and its impacts in order to consider these interannual variations and fully apprehend the risks that Hg and other pollutants represent for seabirds and other marine organisms.

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Table 1: Difference in Hg concentrations (mean \pm SD Δ [Hg] in $\mu\text{g/g dw}$, absolute value)) from one year to the other for low and high fidelity groups, in addition to the results of the associated student or Wilcoxon tests (differences in $|\Delta$ [Hg]| between low and high fidelity groups) and the mean [Hg] \pm SD per species (pool of 2015-2017 details are summarized in Table SI 1)

Species	$ \Delta$ [Hg] in High fidelity group (n obs)	Range	$ \Delta$ [Hg] in Low fidelity group (n obs)	Range	t.test/Wilcoxon test	Mean [Hg] \pm SD (n obs)	Nb of individual with high fidelity	Nb of individual with low fidelity	Nb of individual with both fidelities
Black-legged kittiwakes	1.03 \pm 0.98 (5)	0.39-2.71	1.48 \pm 0.91 (8)	0.02-2.82	$W = 13, p = 0.341$	3.10 \pm 1.67 (26)	5	8	0
Brünnich's guillemots	1.00 \pm 0.89 (67)	0.00-3.79	1.31 \pm 1.22 (32)	0.07-4.64	$W = 917.5, p=0.249$	4.09 \pm 2.24 (157)	40	18	7
Common guillemots	0.68 \pm 0.76 (31)	0.07-3.73	1.23 \pm 0.86 (30)	0.04-2.95	$W = 263, p=0.003$	2.73 \pm 1.18 (89)	18	15	6
Little auks	1.26 \pm 0.93 (3)	0.37-2.22	1.17 \pm 1.21 (3)	0.15-2.50	$t = 0.10, df = 3.75, p = 0.924$	2.93 \pm 1.23 (12)	3	3	0
Northern fulmars	0.81 \pm 0.74 (20)	0.14-2.78	0.97 \pm 0.81 (32)	0.01-3.37	$W = 280, p = 0.461$	2.34 \pm 1.16 (83)	13	21	4
Common eiders	-0.27 \pm 0.28 (22)	0.03-1.23	-	-	-	0.81 \pm 0.29 (41)	22	0	0

Table 2: AIC model ranking for winter Hg concentrations within species and individuals with low fidelity for their wintering ground from one year to the other. The GLM results are presented in a decreasing order, from the best model (lowest AIC and highest delta AIC) explaining variations in delta [Hg]. The table includes the AICc, the Δ AICc and the AIC weight.

Variable	Model	AICc	Δ AICc	AIC Weight
All species				
Delta [Hg]	~ Distance * Species + Direction	360.62	0.00	0.59
	~ Distance + Species + Direction	362.46	1.85	0.23
	~Species * Direction	364.14	3.52	0.10
	~ Distance + Species * Direction	365.46	4.84	0.05
	~Species + Direction	366.66	6.05	0.03
	~ Distance * Species	378.11	17.50	0.00
	~ Distance + Species	378.47	17.85	0.00
	~ Species	378.75	18.14	0.00
	~ Distance * Species * Direction	381.54	20.92	0.00
	~ Direction	381.98	21.36	0.00
	~ Distance	388.92	28.30	0.00
	~1	390.09	29.48	0.00

Fig. 1: Winter distribution of black-legged kittiwakes, Brünnich's guillemots, common eiders, common guillemots, little auks (median position from November to January) and northern fulmars (median position from November to December). Sampling sites (Table S1) are identified by (▲).

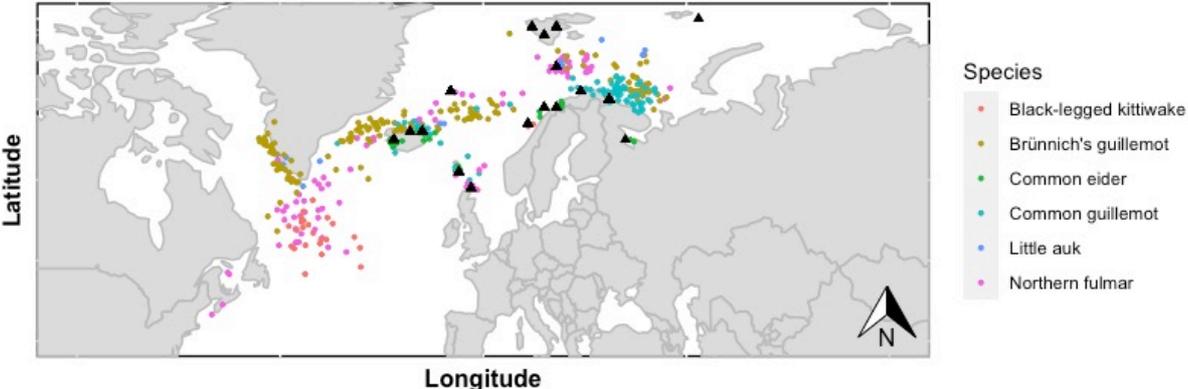


Fig. 2: Δ [Hg] (absolute) between one year to the other for birds with low ($>372\text{km}$) or high fidelity ($<372\text{km}$) for their wintering grounds, per species. Each species is represented by a specific color (see legend). The mean Δ [Hg] is represented by a red dotted line (detailed boxplot per species is provided in Fig. S1).

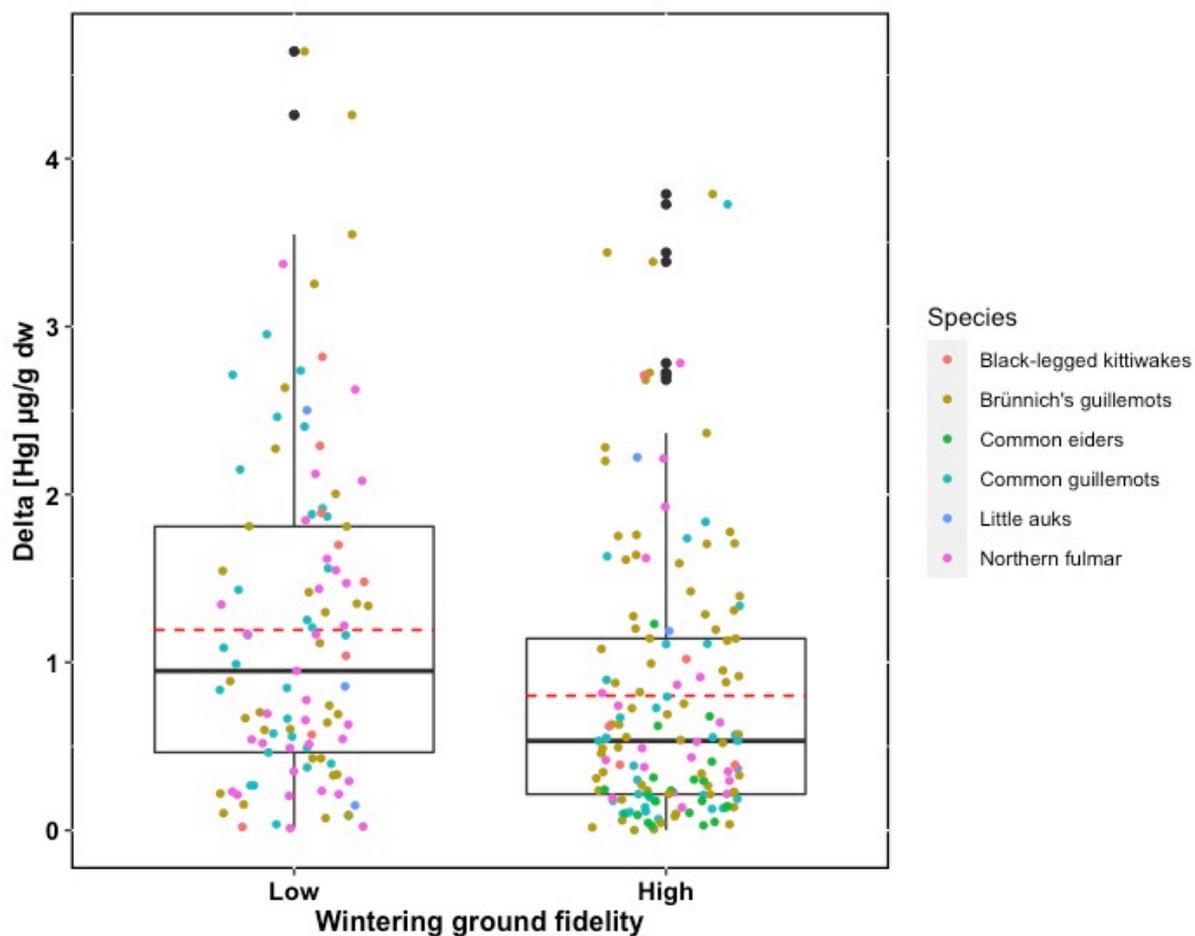
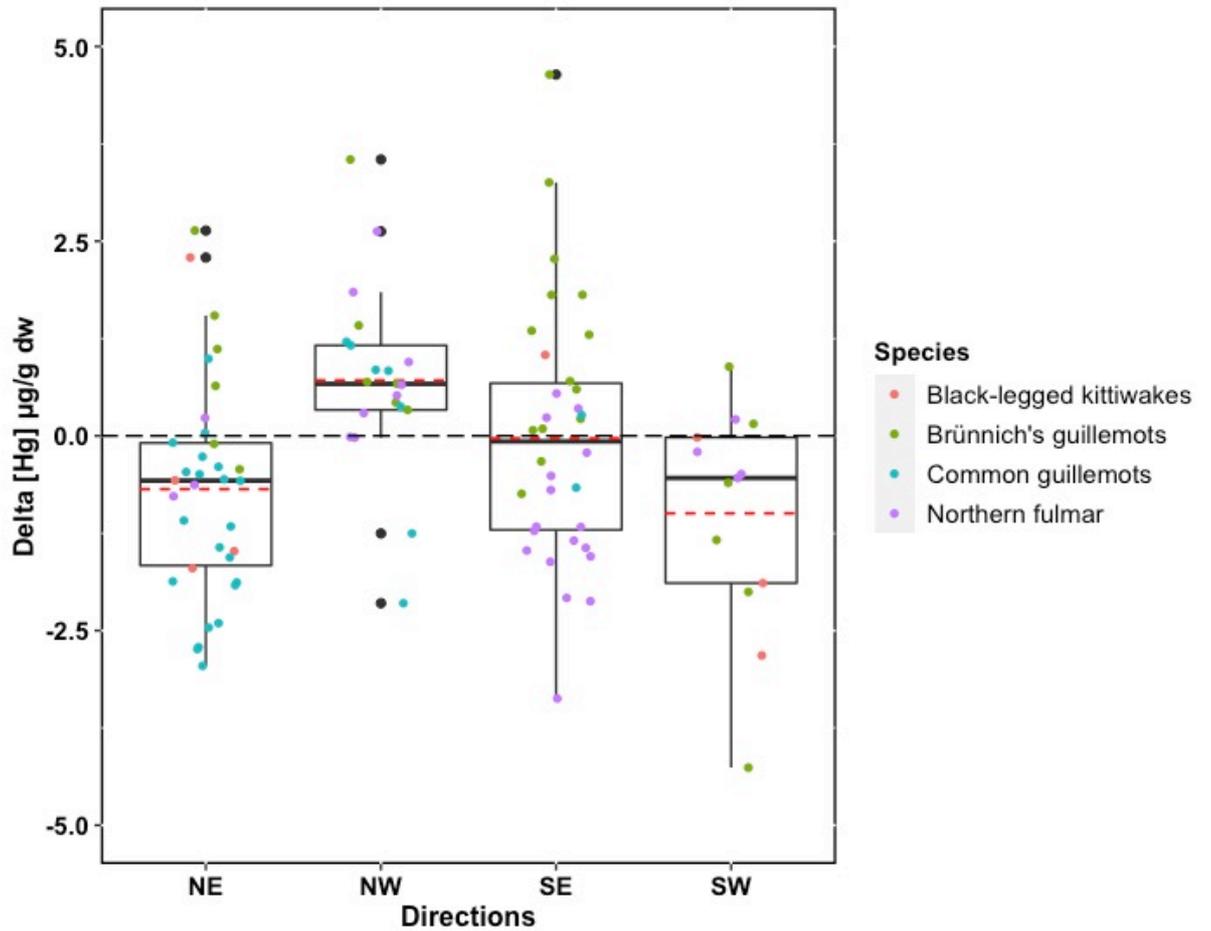


Fig. 3: Δ [Hg] in the fourth different directions (NE=North-East, NW=North-West, SE=South-East, SW=South-West) in species and individuals with low fidelity for their wintering grounds. The mean Δ [Hg] is represented by a red dotted line. The zero axis is represented by a black dotted line (detailed boxplot per species is in Fig. S2).



Supplementary information

Table S5 : sampling effort (n) and mean Hg concentrations (mean ± SD µg/g dw) per species and sampling sites from 2015 to 2017.

Species	Sampling site	2015	2016	2017
Black legged kittiwake	Anda	3.29±0.54 (2)	3.29±2.84 (3)	2.88±1.19 (3)
	Hornøya	-	0.85±0.04 (2)	1.36±0.71 (2)
	Langanes	4.36±1.23 (4)	4.16±0.79 (4)	
	Rost	-	3.55±2.20 (3)	2.11±1.23 (3)
Brünnich's guillemot	Alkefjellet	-	1.85±0.49 (5)	1.57±0.82 (5)
	Bjørnøya	2.45±1.42 (13)	3.50±2.26 (9)	2.52±1.15 (6)
	Grimsey	-	5.89±1.24 (5)	5.87±1.04 (5)
	Hornøya	2.73±1.38 (17)	2.31±1.32 (12)	2.59±1.14 (16)
	Isfjorden	6.36±1.43 (2)	5.97±1.21 (2)	-
	Jan Mayen	5.58±1.12 (11)	6.77±2.36 (22)	5.19±0.97 (17)
	Langanes	4.33±0.98 (4)	5.67±0.64 (3)	4.69±0.41 (3)
Common eider	Breidafjordur	-	0.82±0.23 (8)	1.03±0.37 (8)
	Faroes	-	0.65±0.13 (4)	0.49±0.16 (4)
	Kongsfjorden	0.72±0.13 (4)	0.98±0.21 (4)	-
	Solovetsky archipelago	0.82 (1)	-	1.44 (1)
	Tromsø	0.80±0.24 (2)	0.74±0.13 (3)	0.46±0.00 (2)
Common guillemot	Bjørnøya	2.72±0.95 (13)	1.85±0.31 (17)	1.91±0.56 (16)
	Hjelmsøya	4.29±1.18 (4)	2.35±0.39 (5)	2.94±0.90 (4)
	Jan Mayen	3.23±1.46 (3)	3.71±1.86 (5)	2.25±1.17 (4)
	Langanes	3.84±0.55 (8)	4.31±0.98 (6)	2.79±1.14 (4)

Little auk	Bjørnøya	4.12±0.33 (2)	2.35±0.41 (3)	3.34±1.41 (5)
	Hooker Island	-	1.67 (1)	1.52 (1)
Northern fulmar	Bjørnøya	1.57±0.84 (11)	1.93±1.34 (6)	1.09±0.60 (8)
	Eynhallow	-	3.49±0.42 (5)	3.06±1.14 (5)
	Jan Mayen	3.35±0.78 (3)	2.95±1.15 (3)	1.79±0.84 (6)
	Langanes	2.87±1.42 (11)	2.79±0.86 (12)	2.28±0.88 (13)

Fig. S1: Δ [Hg] (absolute) from one year to the other for birds with low (<372km) or high fidelity (>372km) for their wintering grounds, per species. The mean Δ [Hg] is represented by a red dotted line.

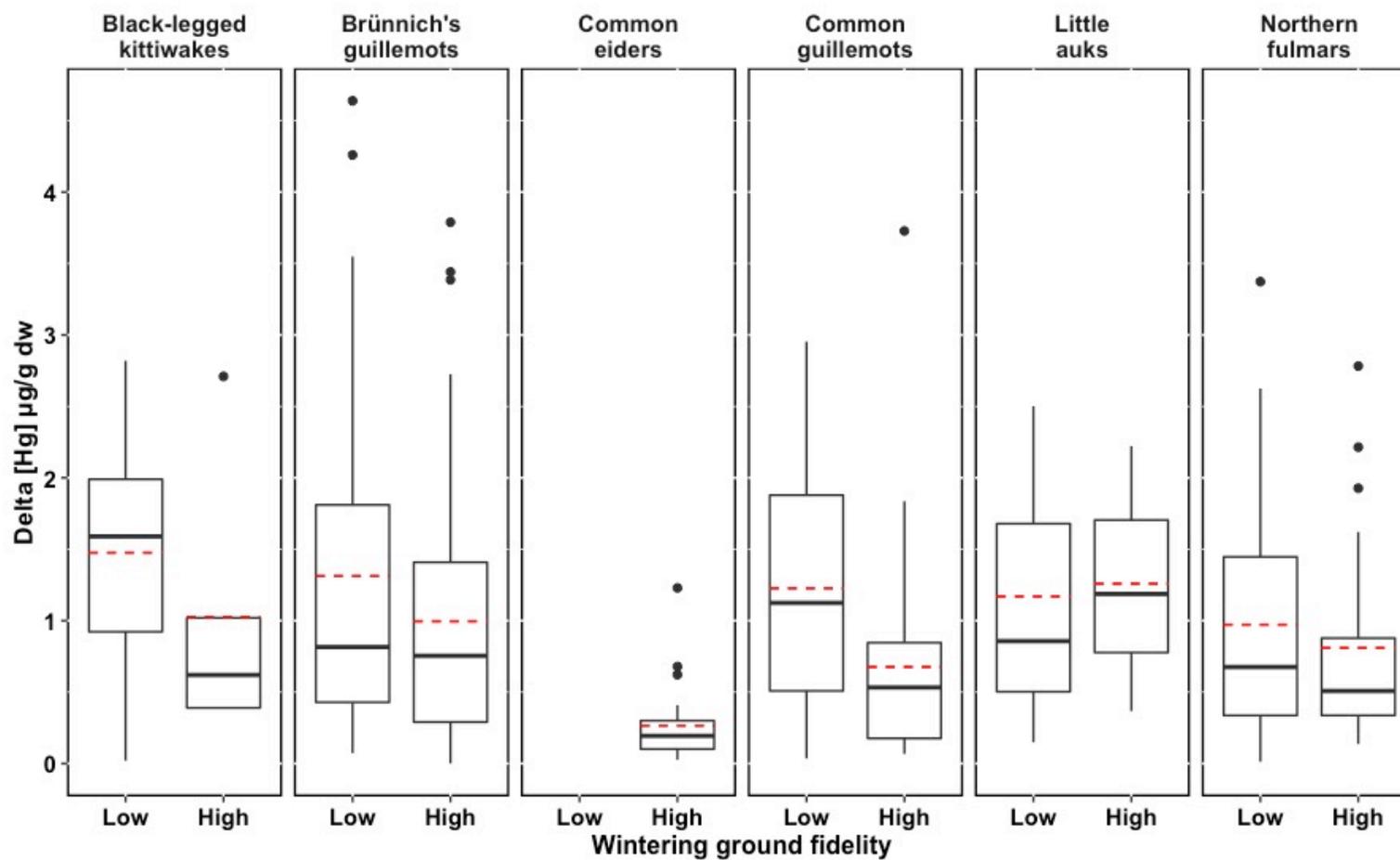


Fig. S2: Δ [Hg] in the four different directions (NE=North-East, NW=North-West, SE=South-West, SW=South-West) per species for individuals with low fidelity for their wintering grounds. The mean Δ [Hg] is represented by a red dotted line. The zero axe is represented by a black dotted line.

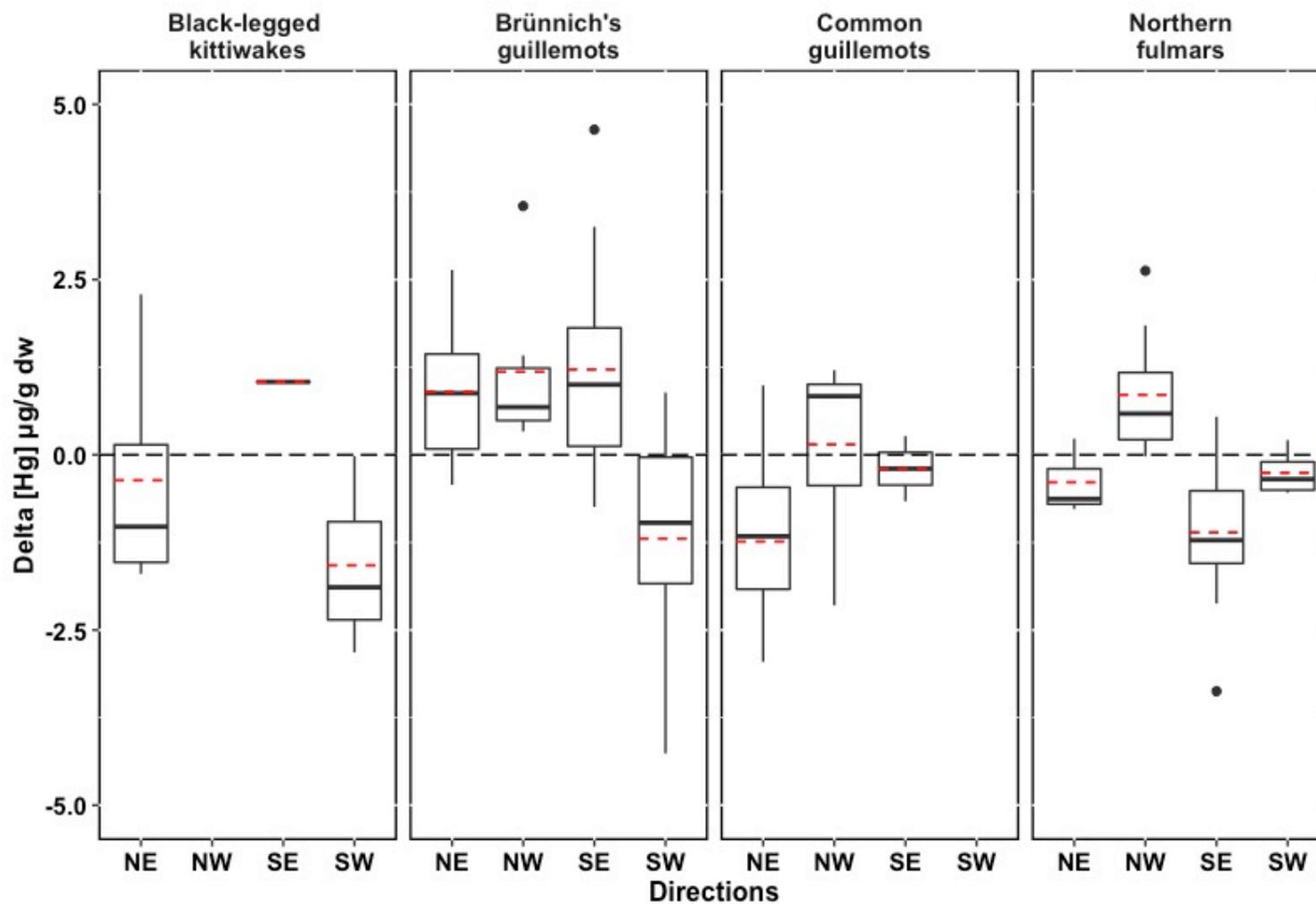
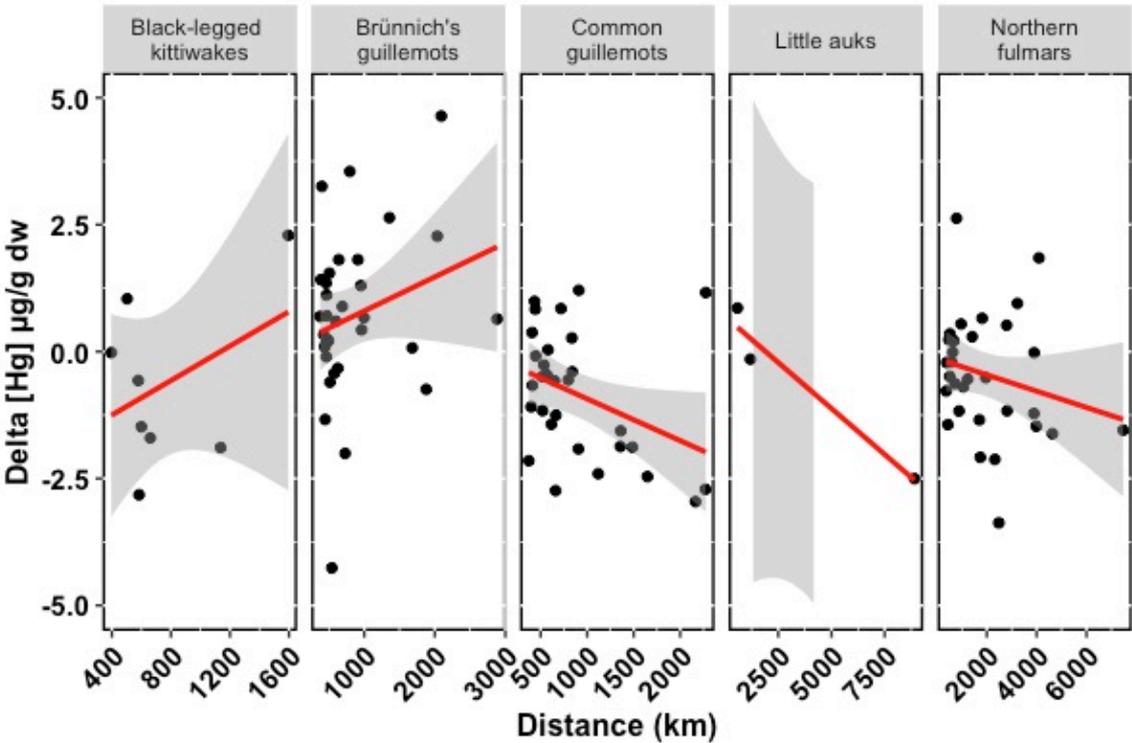


Fig. S3: Δ [Hg] in relation to the distance (in km) for the five studied species and individuals with low fidelity for their wintering grounds.



Paper 5

Seabirds as bioindicators of winter mercury distribution through the North-Atlantic Arctic.

Albert C., Moe B., Sandøy V.B., Helgason H.H., Astrom J., Amélineau F., Angelier F., Anker-Nilssen T., Chastel O., Christensen-Dalsgaard S., Danielsen J., Descamps S., Elliott K., Erikstad K.E., Ezhov A., Gavrilov M., Grémillet D., Krasnov Y., Langset M., Lorentsen S.-H., Merkel B., Olsen B., Patterson A., Reiertsen T.K., Systad G.H., Thompson P., Thórarinnsson T.L., Brault Favrou M., Bustamante P., Strøm H., Fort J.



Introduction

Mercury (Hg), a widely distributed contaminant, is a neurotoxic affecting human and wildlife health (UNEP, 2013). To protect the environment from its toxicity, the United Nations Environment Programme (UNEP) established the Minamata convention in 2017, engaging the signatory countries to decrease their emissions. To follow and improve the efficiency of such programme, extensive and large-scale Hg studies are essential to understand Hg transportations, origins of spatial contamination (Bowman et al., 2016; Evers and Sunderland, 2019; Heimbürger et al., 2015a; Pomerleau et al., 2016; Wang et al., 2018) and associated risks for human and wildlife (Dietz et al., 2019; Tan et al., 2009a; Wolfe et al., 1998).

Even far from intensive human activities, high Hg concentrations have been measured in the Arctic (AMAP 2018), with the Canadian Arctic presenting some of the highest concentrations. Additionally, global change has induced rapid rise of temperature (ACIA, 2004), causing a fast decrease of sea ice cover, potentially exacerbating Hg release in the Arctic. This strengthen the need to closely monitor this pollutant through the Arctic. However, and mostly because of logistical reasons, this knowledge is limited to the summer period and neglect a large part of the Arctic and Pan-Arctic marine areas (see AMAP, 2018).

We combined Hg analyses and wildlife tracking (Fort et al., 2014) to provide new insight about winter Hg distribution at the North Atlantic Arctic scale. As migratory species, Arctic seabirds cover large marine areas. In addition, as top predators they reflect different habitat and/or trophic chains. Therefore, we used seven Arctic seabirds as bioindicators of the winter Hg contamination status of the North Atlantic Arctic.

In the present study, we used seven species of Arctic seabirds' species as proxies to map the spatial origin of winter Hg contamination through the North-Atlantic Arctic and highlight potential hotspots of contaminations. We tested the hypotheses that winter Hg concentrations were spatially different and predict that the Canadian Arctic will present higher Hg concentrations than the Barents Sea (AMAP, 2018).

Material and methods

Species, study sites and sampling collections

From June 2014 to July 2018, Atlantic puffins (*Fratercula arctica*, n=42), black-legged kittiwakes (*Rissa tridactyla*, n=120), Brünnich's guillemots (*Uria lomvia*, n=244), common eiders (*Somateria mollissima*, n=118 females), common guillemots (*Uria aalge*, n=130), little auks (*Alle alle*, n=64), and northern fulmars (*Fulmarus glacialis*, n=124) were equipped with geolocators (GLS) at 27 breeding colonies (Table 1), for two to four years. GLS were retrieved every following breeding seasons (one to three times). Geolocators are low-weight instruments that record ambient light-levels over multiple years, which later can be converted into geographical locations. However, the positional precision derived from light-level data are considered low (Lisokvski et al., 2012), and is mostly suited for studies of large-scale movements and distribution.

In order to investigate individual Hg contamination during winter (non-breeding) periods, feathers representing the winter period (Albert et al., 2019) were collected at deployment and recapture of GLS. During the moult, seabirds excrete a large proportion of accumulated Hg into their feathers. Hence, feathers inform about Hg accumulated by an individual between two moulting sequences, and specifically during the non-breeding period according to selected feathers and bird moulting pattern (see Albert et al, 2019). For example, Alcids undergo a total moult after the breeding season (September - October) resulting in the winter plumage and a partial moult (cheek, neck, throat) at the end of the winter period (March - April) resulting in the nuptial plumage. Black-legged kittiwakes (as most Larids) undergo a complete moult after the breeding period and a partial (e.g. back, head) moult at the end of the winter period (Cramp and Simmons, 1983). Female common eiders undergo a partial moult (e.g. body) after the breeding season and a complete moult at the end of the winter period (Baldassarre, 2015; Goudie et al., 2020). Hence, head, back and belly feathers provide information on Hg contamination specifically during the non-breeding period in alcids, kittiwakes and eiders, respectively. Northern fulmars undergo one total moult per year after the breeding season (from mid-August to late October - Grissot et al. 2020). However, recent studies indicated that Hg concentrations in body feathers reflect inter-individual variations in Hg contamination during the non-breeding period (Quinn et al., 2016).

We therefore collected head feathers on Atlantic puffins, Brünnich's guillemots, common guillemots and little auks, and body feathers on common eiders (belly), black-legged kittiwakes (back) and northern fulmars (belly). Feathers were then stored in plastic bags at ambient temperature until Hg analyses.

Spatial analyses

GLS light-level data were first converted into positional data by identifying the timing of twilights, using a threshold method, from which we estimated two daily latitudes from the length of the night and day, and two daily longitudes from time of apparent midnight and noon, following procedures described in Bråthen et al. (2020). For individuals located above 70 degrees north, positions could not be obtained for days or weeks around winter solstice because of an apparent absence of daylight above this latitude.

Winter positions were then extracted to investigate individual distribution during this period. The timing of Arctic seabird moult and migration can slightly vary between breeding sites (Fort et al., 2013b; Frederiksen et al., 2012). Nonetheless, to allow inter-population comparisons, based on previous investigations (Frederiksen et al., 2016, Fort et al., 2012, 2013) and given that these variations are limited at the scale of the winter period, this latter was homogenised within species. Therefore, we considered that birds were at their wintering ground from November to January for little auks (Fort et al., 2012a), common and Brünnich's guillemots (Fort et al., 2013c; Frederiksen et al., 2016; Merkel et al., 2019), black-legged kittiwakes (Frederiksen et al., 2012) and common eiders (Hanssen et al., 2016). As northern fulmars have been observed reaching back their colonies as early as January, the winter period was defined from November to December in this species (SEATRACK, *Unpublished*). In order to have the most accurate seabirds' accurate distribution, we then calculated a median winter position per week for each individual for each year, to be used in further analyses (Table 1).

Hg analyses

Prior to Hg analyses, feathers were cleaned to remove external contamination. Feathers were plunged into a 2:1 chloroform:methanol solution for three minutes in an ultrasonic bath, rinsed twice in a methanol solution and dried at 45°C for 48 hours. Hg analyses were performed on a ~0.20-1 mg

subsample of a pool of three homogenized feathers, using an Advanced Mercury Analyser spectrophotometer (Altec AMA 254 – detection limit of 0.05 ng). The analysis of each sample was repeated (two to three times) until the relative standard deviation for two subsamples was <10%. Mean concentration for these two subsamples was then used for statistical analyses. Prior to Hg analyses, blanks were run and, to ensure the accuracy of measurements, certified reference materials (dogfish liver DOLT-5 and lobster hepatopancreas TORT-2, NRC, Canada) were used every fifteen samples. Hg concentrations are expressed in $\mu\text{g/g}$ dry weight (dw).

Statistical analyses

The mapping of the spatial distribution of Hg concentration in the North Atlantic Arctic was realised with interpolation methods (package “*gstat*”-Pebesma 2004). Prior to the interpolations, we ran Generalized Additive Models (GAMs) to take into account different variability in the data. In a first step, we considered all species and in a second step, focused on every species individually. In each model, we always included winter longitudes and latitudes as fixed effects, species (1st step only) and individuals as random effects. Indeed, as individuals were attributed several winter median points (see Spatial analyses section), this variable needed to be treated as a random effect (see Table 1). In addition, as we aimed to map Hg distribution depending on a multi-species analysis, this variable also needed to be treated as a random effect in order to take into account their different ecologies. species were used as bioindicators of their environment. Then, we used the residuals of the GAMs to run the interpolation (kriging) on a grid (resolution: $1.5^\circ \times 0.5^\circ$). Predictions and variances of the GAMs and kriging were summed in order to have the most accurate values. Those results were mapped for all the species and then individually (Figs. 1-2). Hg concentrations were log transformed to meet the parametric assumptions of normality and homoscedasticity of residual distribution. Statistical analyses were performed with R version 3.4.3 (R Core Team, 2017). Means are reported with standard deviation (mean \pm SD) unless reported otherwise.

Results

The results of the GAMs indicate that Hg concentration variate depending on the winter latitudes and longitudes, but also the individuals (except the black-legged kittiwakes) and the species (Table 2). The multi-species interpolation indicates an east-west gradient in Hg concentrations increase. More

specifically, the highest Hg concentrations are predicted on the South-East of Greenland and along the Canadian Coast (where we have some of the lowest variance). On the other hand, the lowest predictions are found in the Barents Sea and along the Norwegian coast.

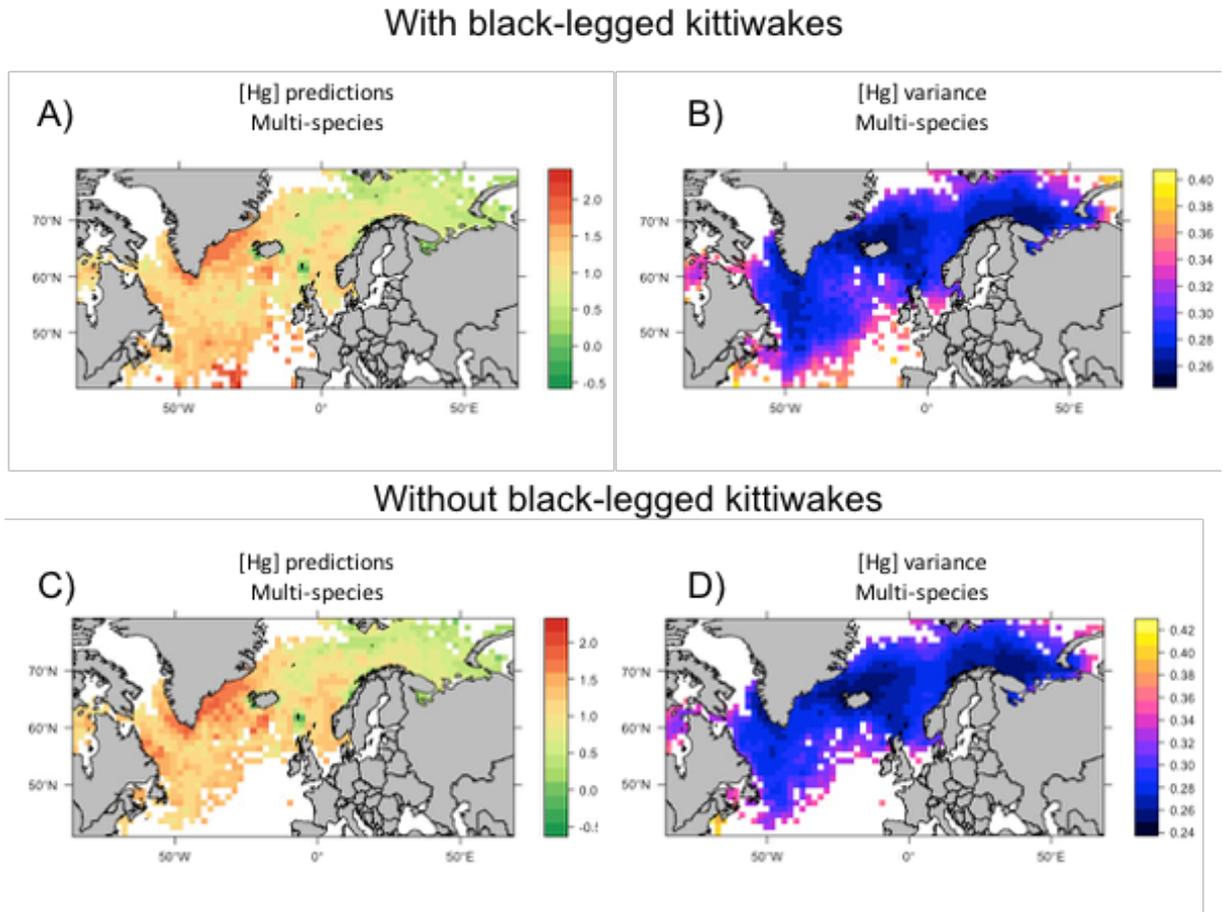


Figure 5: spatial representation of the [Hg] predictions and variances A),B) with or C), D), without black-legged kittiwakes.

When looking specifically at the intraspecies variations, the alcid species unravel the highest Hg predictions on the East coast of Greenland. In addition, an east-west Hg increase appears for all species covering large areas. However, the results are less clear for the black-legged kittiwakes, species for which the individuals did not show significant effects on Hg concentrations and for which the variances are the highest.

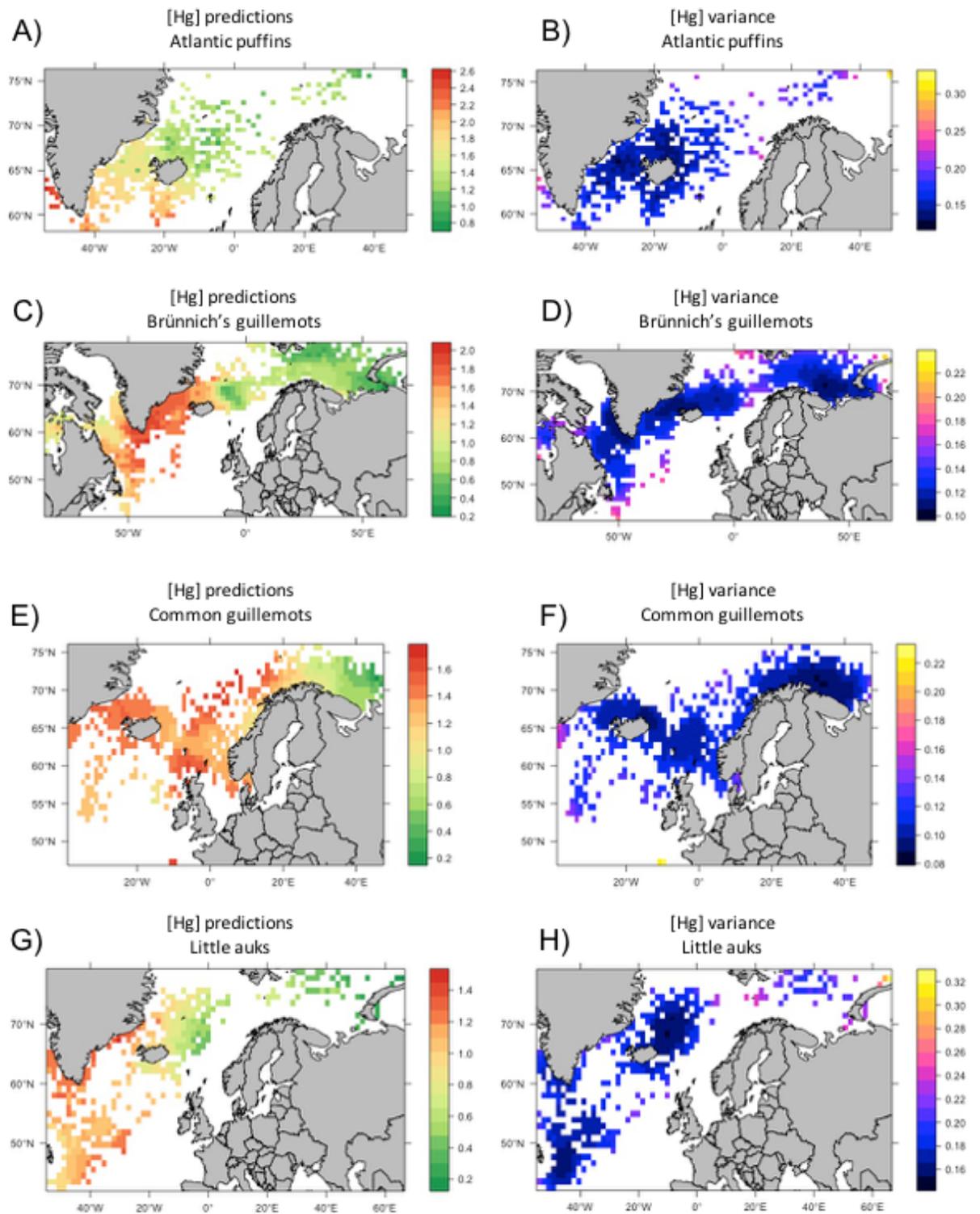


Figure 6 : Hg prediction and variances for the A), B) Atlantic puffins, the C), D) Brünnich's guillemots, E), F) common guillemots and G) H) little auks.

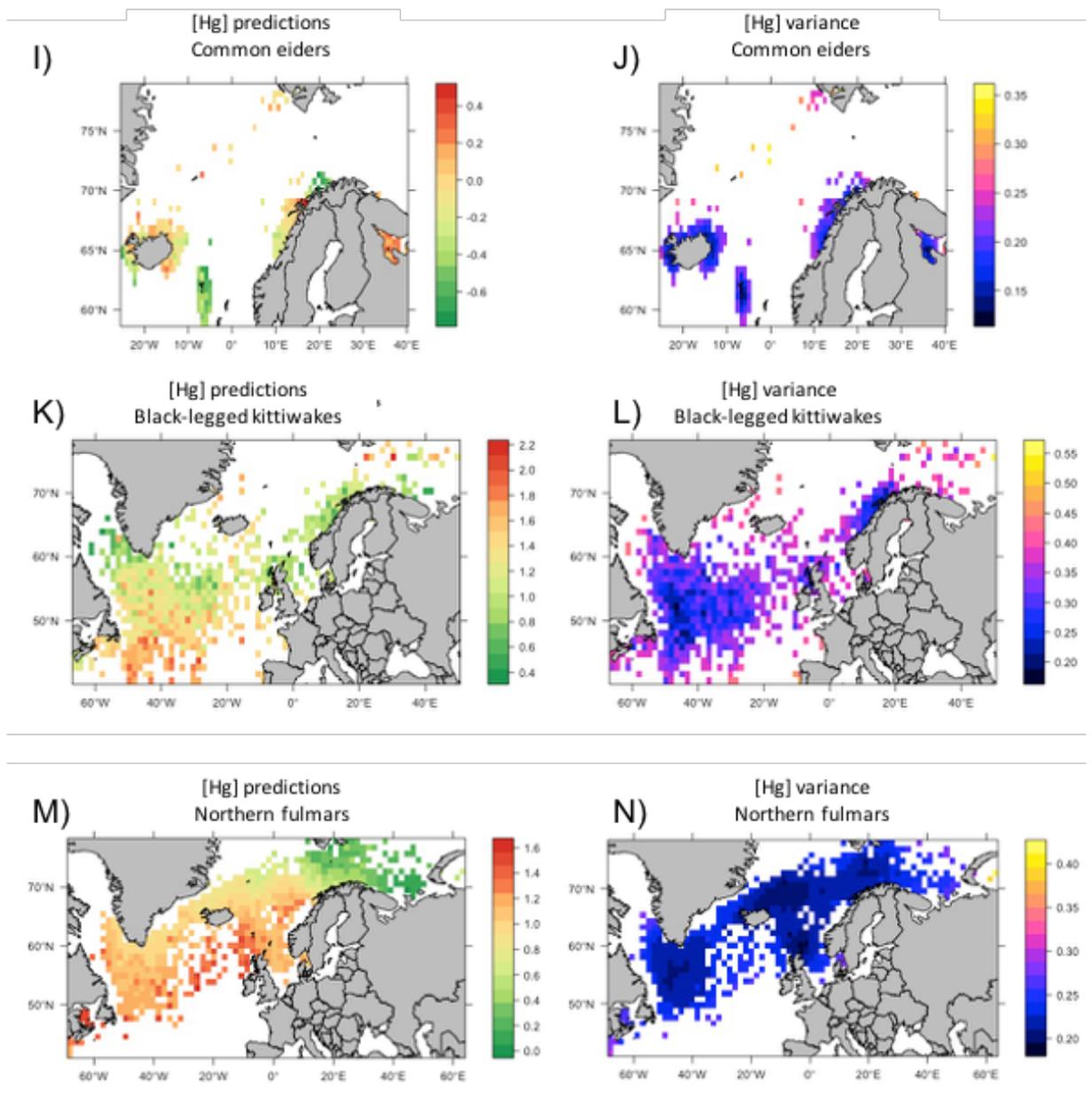


Figure 2 (suite): Hg prediction and variances for the I), J) common eiders, the K), L) black-legged kittiwakes, M), N) Northern fulmars.

Discussion

The use of seabirds as bioindicators unravels spatial variations in Hg concentrations through the North Atlantic Arctic, with an east-west increase in Hg concentrations. These results follow precedent studies focused on the breeding season (AMAP, 2018; Provencher et al., 2014), which highlighted that

some of the highest Hg concentrations were measured in the Canadian Arctic, which was mostly attributed to the atmospheric deposition coming from Asia (AMAP, 2011). In addition, we highlighted hotspots of Hg on the East coast of Greenland and along the Canadian coast (Labrador Sea, Newfoundland). The Arctic Ocean is a sink for contaminants such as Hg, with inflows from both atmospheric and oceanic currents, as well as rivers (Sonke et al., 2018a). Nevertheless, important outflows of Hg also occur from the Arctic to lower latitudes through the Arctic Canadian Archipelagoes and Fram Strait (Outridge et al., 2008). As the Hg hotspots found in the present study are following the Greenlandic (East and West) and Labrador oceanic currents, we believe they could partly explain this high Hg concentrations. On the contrary, the lowest concentrations are found in the Barents Sea and might be explained by lower atmospheric deposition and oceanic transportation in comparison to the West Atlantic. Indeed, the Minamata convention engaged signatory countries to decrease their Hg emissions. Important decrease has been observed in Europe. Oceanic currents from south Europe to the Barents Sea may have seen their load in Hg concentration decrease and might therefore explain the lower Hg concentrations found in this area. In addition, atmospheric Hg deposition in the Barents Sea comes from Russia and North America, countries where emissions are much lower than is Asia, and might explain part of the spatial variations between East and West Atlantic-Arctic (AMAP, 2011b).

Large-scale Hg assessment during winter is essential. It allows to extend the existing knowledge about the breeding period to this specific period (e.g. contamination, threats), in addition to cover marine habitats only used during winter. Consequently, this should allow to set up appropriate conservation measures specifically for this period. The results of the present study show that seabird populations breeding through the Arctic winter through a large marine part of the North-Atlantic Arctic. Previous studies based on multi-colonies analysis show that the North Atlantic was a key area for many seabird species during winter and played a role on population trends (Fort et al., 2013b; Frederiksen et al., 2016, 2012). For instance, when Brünnich's guillemots populations breeding in Canada and mostly wintering off Canada were stable, the populations breeding in Svalbard and mostly wintering off Greenland declined (Frederiksen et al. 2016). To explain such a decrease, direct human impacts like hunting has been proposed as one possible threat (Frederiksen et al., 2012). Our study also reveals the importance of winter by highlighting the existence of Hg hotspots. The fact that hotspots of Hg concentrations

occurs in areas with high seabird densities during winter suggest that this contaminant should also be considered as a possible threat for populations survival and trends. Knowing the impact of this pollutant (and other pollutants) on wildlife, the conservation measures about contaminants should be extended to the winter period in marine areas highly used by wildlife.

Hg cycle and its transportation is complex and involve many mechanisms. Therefore, there is a need to monitor this pollutant at different level (biotic and abiotic) in order to have a large and precise idea of its distribution (Evers et al., 2016). Previous work modeled Hg distribution at a large scale through the study of oceanic current and air deposition. For instance, Zhang et al (2020) modelled MeHg at different marine layers. We compared our results with their surface modelization (0-100m) where most of our studied species feed (but some seabirds plunge higher than 100m), but only few areas seem to have similar pattern (off East Greenland). On the four atmospheric deposition models summarized in the AMAP (2011b), our results only seems to follow a similar trend, but only along the Greenlandic coast (Dastoor et al. 2008, GRAHM, net depositions). These differences might be explained by temporal variations (e.g. season, difference in assessment years) or incorporation time into the food web (see Foster et al., 2019). Overall, this strengthen the need to use both biotic and abiotic studies in Hg distribution research.

Mapping Hg distribution is also essential for acknowledging the efficiency of international program of Hg reduction. By signing the Minamata convention, many countries agreed to reduce their emissions in order to locally and internationally protect the environment and therefore, human health. This convention calls for international collaboration, studying Hg on different compartments for improving our knowledge. Subsequently, conservation and international programs can be set up, adapted and/or reinforced, and new gaps and questions of research can be arisen. In this particular case, we believe that new modelization research including oceanic currents and wildlife measurements will be needed in order to improve knowledge about Hg incorporation into the food webs and potential large-scale transportation through the food chains, until top predators and human beings.

Table 6: mean [Hg] ± SD (µg/g dw), winter median longitude and latitude (mean ± SD), per species and sampling sites. Number of observations and individuals are also mentioned

Sampling colonies	Colonies coordinates	Species	Mean [Hg] ± sd µg.g-1	Winter median longitude ± sd	Winter median latitude ± sd	Observations (n)	Individuals (n)
Alkefjellet	<i>79.58°N, 18.51°E</i>	Brünnich's guillemot	1.64±0.44	26.79±10.02	76.06±1.66	86	23
		Northern fulmar	1.48±1.01	37.72±13.93	71.97±2.23	38	5
Anda	<i>69.07°N, 15.17°E</i>	Black-legged kittiwake	3.39±1.42	-25.58±24.48	57.20±6.84	286	19
Bjørnøya	<i>74.50°N, 18.96°E</i>	Brünnich's guillemot	3.01±1.76	-5.35±16.98	69.00±2.79	521	25
		Common guillemot	2.06±0.65	34.29±6.00	70.67±1.38	706	32
		Little auk	2.65±1.09	-17.21±18.06	66.90±4.83	417	28
		Northern fulmar	1.59±0.92	22.75±9.54	72.88±1.86	234	22
Breidafjordur	<i>65.08°N, 22.74 °W</i>	Common_eider	1.01±0.43	-22.55±0.80	64.62±0.58	435	26
Cape Flora	<i>79.95°N, 50.09°E</i>	Black-legged kittiwake	5.84±2.13	-34.94±21.73	53.03±8.23	120	9
		Brünnich's guillemot	1.58±0.32	41.65±8.08	75.86±1.75	12	4
Cape Krutik	<i>69.15°N, 35.95°E</i>	Black-legged kittiwake	2.34±0.83	-32.84±30.89	55.72±8.32	105	8
Coats Island	<i>62.47°N, 83.10°W</i>	Brünnich's guillemot	4.34±3.52	-62.64±11.93	57.29±4.49	315	23
Eynhallow	<i>59.14°N, 3.12°W</i>	Northern fulmar	3.76±1.51	-8.39±15.57	59.28±3.58	332	20
Faroës	<i>61.98°N, 6.65°W</i>	Common eider	0.65±0.25	-6.41±0.55	61.32±0.80	282	17
		Common guillemot	5.15±2.24	-8.05±8.40	60.74±4.52	94	7
		Northern fulmar	3.48±1.28	-15.58±18.70	60.04±4.87	158	12
Cape Gorodetskiy	<i>69.58°N, 32.94°E</i>	Brünnich's guillemot	2.12±0.38	27.92±19.49	70.75±1.59	89	7
Grimsey	<i>66.54°N, 18.00°W</i>	Brünnich's guillemot	5.76±0.83	-40.91±13.03	61.72±4.17	136	10
Hjelmsoya	<i>71.11°N, 24.73°E</i>	Common guillemot	3.03±1.15	30.44±6.24	70.92±1.07	316	20
Hooker Island	<i>80.23°N, 53.02°E</i>	Little auk	1.60±0.50	19.4±26.53	73.4±3.28	90	16
Hornøya	<i>70.39°N, 31.16°E</i>	Black-legged kittiwake	2.38±1.26	-36.74±20.47	55.09±5.71	135	10
		Brünnich's guillemot	2.67±1.25	25.14±29.22	69.81±4.12	714	38
Isfjorden	<i>78.25°N, 15.51°E</i>	Brünnich's guillemot	6.31±1.36	-39.47±13.66	63.47±3.64	216	14
		Little auk	4.03	-10.82	72.05	1	1

Jan Mayen	<i>71.03°N, 8.29°W</i>	Brünnich's guillemot	5.88±1.81	-39.41±13.44	64.14±3.47	837	33
		Common guillemot	3.75±1.48	1.99±25.62	67.42±4.22	445	27
		Northern fulmar	2.81±1.19	-12.34±19.98	66.21±6.87	542	33
Kap Hoegh	<i>70.72°N, 21.55°W</i>	Little auk	2.99±0.62	-41.00±8.42	50.11±5.15	253	19
Kara gate	<i>71.42°N, 51.95°E</i>	Brünnich's guillemot	1.81±0.51	50.10±6.15	70.53±1.01	476	36
Kongsfjorden	<i>79.00°N, 11.67°E</i>	Black-legged kittiwake	6.65±2.46	-35.8±16.46	50.67±6.69	234	15
		Common eider	1.03±0.33	-12.36±8.99	66.24±2.42	495	28
Langanes	<i>66.18°N, 15.99°W</i>	Black-legged kittiwake	5.56±3.10	-36.66±10.00	52.42±5.42	204	15
		Brünnich's guillemot	4.89±1.16	-29.53±13.55	64.8±3.00	409	25
		Common guillemot	3.71±0.86	-14.29±8.05	64.94±2.30	511	27
		Northern fulmar	2.90±1.19	-28.22±21.65	61.86±7.20	673	32
Oranskyie islands	<i>77.07°N, 67.64°E</i>	Brünnich's guillemot	1.71±0.20	47.05±11.37	74.06±2.31	29	6
Rost	<i>67.45°N, 11.91°E</i>	Atlantic puffin	5.09±1.99	-18.41±15.33	66.14±3.43	728	42
		Black-legged kittiwake	3.10±1.15	-22.76±27.79	57.54±7.75	264	16
Runde Alesund	<i>62.44°N, 5.87°E</i>	Black-legged kittiwake	4.85±1.27	-29.4±22.35	53.74±5.85	154	11
Selvaer	<i>66.59°N, 12.23°E</i>	Common eider	0.91±0.24	12.18±1.32	66.06±1.10	107	8
Sklinna	<i>64.74°N, 10.77°E</i>	Black-legged kittiwake	2.14±0.70	-23.42±22.97	55.52±6.55	237	17
		Common guillemot	3.19±0.83	16.91±11.87	65.89±4.98	226	17
Solovetsky archipelago	<i>65.05°N, 35.79°E</i>	Common eider	1.23±0.28	36.26±1.08	64.99±0.89	218	15
Tromsø	<i>69.64°N, 18.85°E</i>	Common eider	0.77±0.75	19.29±1.09	69.16±0.54	378	24

Table 7: results of the generalized additive models (GAMs)

	Variables	edf	Ref.df	F	p
All species	s(longitude, latitude)	28.78	29.00	183.70	< 2e-16
	s(individual)	1.00	1	1073.93	< 2e-16
	s(species)	0.95	1	24.85	9.51e-07
Atlantic puffins	s(longitude, latitude)	21.04	25.44	24.60	< 2e-16
	s(individual)	0.98	1.00	28.91	5.86e-08
Black-legged kittiwakes	s(longitude, latitude)	2.463e+01	27.88	12.46	< 2e-16
	s(individual)	1.732e-04	1.00	0.00	0.518
Brünnich's guillemots	s(longitude, latitude)	26.73	28.65	196.7	< 2e-16
	s(individual)	0.9891	1.00	123.5	< 2e-16
Common guillemots	s(longitude, latitude)	26.59	28.63	109.39	< 2e-16
	s(individual)	0.99	1.00	71.66	< 2e-16
Common eiders	s(longitude, latitude)	26.05	28.44	29.17	< 2e-16
	s(individual)	0.99	1.00	22.03	1.52e-06
Little auks	s(longitude, latitude)	21.40	25.73	9.82	< 2e-16
	s(individual)	0.99	1.00	12.82	0.00025
Northern fulmars	s(longitude, latitude)	25.23	28.14	31.99	< 2e-16
	s(individual)	1.00	1.00	41.79	8.1e-11

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Paper 6

Contrasted spatial and seasonal trends of methylmercury exposure pathways of Arctic seabirds: combination of large-scale tracking and stable isotopic approaches

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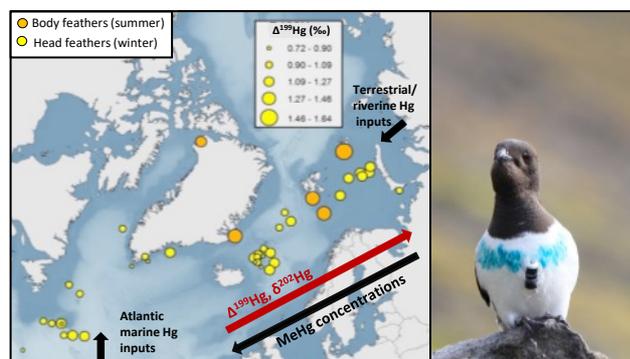
Submitted in Environmental Science and Technology



Abstract

Despite the limited direct anthropogenic mercury (Hg) inputs in the circumpolar Arctic, elevated concentrations of methylmercury (MeHg) are accumulated in Arctic marine biota. However, little is known about the MeHg production and bioaccumulation pathways in these ecosystems. We measured Hg concentrations and stable isotopes of Hg, carbon and nitrogen in feathers and blood of geolocator-tracked little auk *Alle alle* from five Arctic breeding colonies. The wide-range spatial mobility and tissue-specific Hg integration times of this planktivorous seabird allowed the exploration of their spatial (wintering quarters/breeding grounds) and seasonal (non-breeding/breeding periods) MeHg exposure. An east-to-west increase of head feather MeHg concentrations ($1.74\text{-}3.48\ \mu\text{g}\cdot\text{g}^{-1}$) was accompanied by longitudinal trends of Hg isotopic composition (e.g. $\Delta^{199}\text{Hg}$, $\delta^{202}\text{Hg}$), suggesting distinct mixing/proportion of MeHg sources and bioaccumulation pathways between western North Atlantic and eastern Arctic regions. Higher $\Delta^{199}\text{Hg}$ values ($+0.4\text{‰}$) in northern colonies indicate an accumulation of more photochemically impacted MeHg, supporting shallow MeHg production and bioaccumulation in High Arctic waters. Lower $\delta^{13}\text{C}$ values in High Arctic (-20.6‰) relative to North Atlantic wintering seabirds (-19.5‰) may indicate higher contribution of terrestrial/riverine Hg inputs to northern Arctic regions. The combination of seabird tissue isotopic analysis and spatial-tracking helps tracing the MeHg sources at various spatio-temporal scales.

TOCArt- graphical abstract



Introduction

Mercury (Hg) induces major risks for wildlife and human health, especially under its methylated form (methylmercury, MeHg), a potent bioaccumulative neurotoxin (Tan et al., 2009b). In the ocean, MeHg production mainly occurs by biotic *in situ* methylation of inorganic Hg (Blum et al., 2013; Lehnherr et al., 2011). Once formed, MeHg incorporates into the food webs and biomagnifies its concentrations leading to life-impacting levels in top predators and humans. Despite little direct anthropogenic pressure in the Arctic region, total Hg concentrations measured in the Arctic surface seawater are high compared to other regions (Heimbürger et al., 2015b; Mason et al., 2012). Sea-ice melting, direct atmospheric deposition and continental inputs originating from soil erosion and riverine circulation are considered major drivers of the high Hg levels in the Arctic (Beattie et al., 2014; Larose et al., 2011; Obrist et al., 2017b; Anne L Soerensen et al., 2016; Sonke et al., 2018b). However, the MeHg production pathways and zones in the Arctic Ocean are still not completely identified. Several studies demonstrated that Hg in Arctic marine environments may be methylated in the water column or sediments (Kirk et al., 2008; Lehnherr et al., 2011). Potential Hg methylating bacteria were also identified in Antarctic sea ice (Gionfriddo et al., 2016). Recent findings and modelling studies evidenced that the largest net MeHg production in Arctic water columns may occur in oxic waters at the subsurface layer (20–200 m) (Heimbürger et al., 2015b; Anne L Soerensen et al., 2016). A new study also reported the highest abundance of Hg methylating gens in the oxic subsurface waters of the global ocean (Villar et al., 2020), where the highest MeHg concentrations are typically observed (Mason et al., 2012). All these findings suggest that Hg methylation in oxic waters could be a significant source of MeHg towards Arctic marine food webs. Although policy implementations for the reduction of anthropogenic Hg emissions were achieved over the last 30 years in some parts of the world, Hg levels continue to increase in biota from several regions of the Arctic (AMAP, 2011c). Medium to high predators such as seabirds are exposed to significant environmental MeHg concentrations through their diet (AMAP, 2018b, 2011c) and have been extensively studied as bioindicators of Hg exposure in marine food webs (e.g (Carravieri et al., 2016; Thompson et al., 1992)), including the Arctic (Albert et al., 2019b; Dietz et al., 2013b; Rigét et al., 2011). Specific foraging habitats and migratory movements of Arctic seabirds

strongly determine their exposure to distinct environmental MeHg sources in marine ecosystems (Fleishman et al., 2019b; Fort et al., 2014b). However, studies on Hg exposure in Arctic seabirds have commonly put the focus towards the breeding season when seabirds are more accessible for researchers. Consequently, the investigation of Hg exposure during the non-breeding season is still scarce due to sampling logistical difficulties.

The combination of carbon and nitrogen stable isotopes with Hg stable isotopes has demonstrated its suitability for the identification of Hg sources and the associated geochemical processes in the different marine compartments (Day et al., 2012a; D Point et al., 2011; Renedo et al., 2018a). Therefore, its use can help understanding Hg exposure pathways of seabirds according to their migratory behaviour. Hg has seven stable isotopes (196 to 204) and fractionates dependently and independently of the isotopic masses. The combined use of Hg isotopic mass-dependent (MDF, e.g. $\delta^{202}\text{Hg}$) and mass-independent (MIF, e.g. $\Delta^{199}\text{Hg}$) fractionation enables the quantification of processes and the identification of sources and pathways of Hg in the environment (Blum et al., 2014), including marine ecosystems (Cransveld et al., 2017; Day et al., 2012a; Li et al., 2014b; D Point et al., 2011). MDF of Hg isotopes occurs during many physical, chemical or biological processes (Kritee et al., 2009, 2008; Kwon et al., 2013; Zheng et al., 2007). However, large Hg MIF of odd isotopes ($\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$) is observed during light-induced reactions, such as inorganic Hg photoreduction and MeHg photodemethylation. Hg MIF signature is not affected by biological or trophic processes, so it is preserved up to the food webs (Kwon et al., 2012b), then presenting a significant advantage to trace Hg marine sources. For instance, Arctic marine top predators reported much higher Hg odd MIF values (more photochemically impacted Hg) in non-ice covered regions, relating the importance of the accelerated melting of sea ice on the Hg polar cycle (Masbou et al., 2015; D Point et al., 2011). Also, consistent decrease of Hg odd MIF (and MDF) in pelagic fish according to their foraging depth in the North Pacific Ocean demonstrated the dilution of surface MeHg by *in situ* methylated Hg at depth (Blum et al., 2013). More recently discovered, Hg MIF of even Hg isotopes (reported as $\Delta^{200}\text{Hg}$) seems to occur during complex atmospheric mechanisms such as photo-oxidation in the tropopause (Chen et al., 2012). Even-MIF is not induced during any biogeochemical nor photochemical processes in the lower

troposphere or the photic zone (Chen et al., 2012; Gratz et al., 2010; Sherman et al., 2012), therefore the signature is preserved and useful to identify major potential Hg sources of atmospheric origin (Jason D. Demers et al., 2013; Enrico et al., 2016b; Obrist et al., 2017b). Due to the different combinations of the processes involving Hg transformations in the environment, Hg isotopes fractionate differently and with different degrees of magnitude in every specific environmental compartment. Thus, the analysis of Hg stable isotopes of mobile predators such as Arctic seabirds can give access to interesting information about MeHg trophic sources at large scales of the Arctic Ocean and neighbouring water bodies.

Here we propose an original approach consisting in the combination of isotopic analyses (Hg, C and N) and wildlife tracking to provide new information about MeHg exposure pathways of seabirds at both temporal and spatial scales. For this purpose, we focused on the little auk (or dovekie, *Alle alle*), the most numerous seabird species breeding in the High Arctic (between 37 to 40 million breeding pairs estimated (Keslinka et al., 2019; K. Kovacs and Lydersen, 2006)). Little auks have several ecological advantages for their use as a bioindicator models. 1) They are zooplanktivorous and mainly feed on copepods belonging to two *Calanus* species (i.e., *C. glacialis* and *C. hyperboreus*) during the breeding period (Harding et al., 2008). Therefore, they reflect MeHg accumulation in a short food chain that is strongly dependent on sea ice abundance and seawater temperature (Grémillet et al., 2012). 2) They exhibit colony specific wintering areas (Fort et al., 2013a), then reflecting wide-ranging spatial variability of Hg exposure (Jerome Fort et al., 2016). 3) Little auks moult their feathers twice during their annual cycle: a partial moult (head, neck and throat feathers, hereafter ‘head feathers’) during the pre-breeding period (in ca. April) and a complete post-breeding moult in September¹³. During moult, seabirds excrete the Hg accumulated in their body tissues into feathers (Furness et al., 1986b). Thus, feathers reflect the Hg levels accumulated since the last moulting sequence (Honda et al., 1986b). Thereby, the different Hg integration times between the types of feathers allow studying Hg exposure during both the non-breeding (head feathers) and the breeding (body feathers) periods in a same individual (Jerome Fort et al., 2016). Besides, C and N isotopic ratios of blood sampled during the breeding – chick rearing – period can provide information about summer diet and then be compared to Hg levels and isotopic composition in body feathers of little auks (Bearhop et al., 2002; K. A. Hobson

and Clark, 1992). In this study, feathers (head and body) and blood samples of five distant colonies of little auks from the Arctic Ocean were considered with a double objective: i) to investigate tissue-specific Hg isotopic signatures according to their seasonal variability (summer vs winter), and ii) to use both spatial and isotopic information to track sources of Hg contamination along with their migratory circulation.

Material and methods

Sampling sites and description of sample collection

This study was conducted during the seabird breeding seasons of 2015 and 2016 at five colonies of the Arctic Ocean: Franz Josef Land (FJL) (Hooker Island; 80.23°N, 53.01° E), Bear Island (Bjørnøya; 74.45°N, 19.04° E), East Greenland (Kap Høegh; 70.72°N, 21.55°W), Spitsbergen (Hornsund; 76.97°N, 15.78 °E) and North West Greenland (Thule; 77.47°N, 69.22° W). Blood and feathers were sampled from ten individuals per colony (n=50 for the 5 colonies). Individuals from all sites, but Thule, were equipped with a miniature geolocator data-logger (GLS, Biotrack MK4083 or Migrate Technology C65) to track their non-breeding movements and distribution, as described in previous works (Fort et al., 2013a, 2012b; Grissot et al., 2019). We treated GLS tracking data from 1st December to 30th January (period when all little auks were at their winter grounds) and calculated the median individual winter latitude and longitude for each individual separately.

Description of analytical methods

Sample preparation, analyses of total Hg and Hg species concentrations

Body and head feathers were cleaned, oven-dried and homogenized as detailed in a previous work (Jérôme Fort et al., 2016a). Blood samples were dried, lyophilized and ground to powder as previously described (Jérôme Fort et al., 2016a). Feather and blood total Hg concentration (hereafter expressed as $\mu\text{g}\cdot\text{g}^{-1}$, dry weight) was quantified by using an advanced Hg analyser (AMA-254, Altec).

Prior to Hg speciation analyses, blood and feathers (0.01–0.05 g) were digested following previously developed method by microwave-assisted extraction (details in (Renedo et al., 2018b, 2017b)). Quantification of Hg species was carried out by isotope dilution analysis, using a GC-ICP-MS Trace Ultra GC equipped with a Triplus RSH autosampler coupled to an ICP-MS XSeries II (Thermo

Scientific, USA) as detailed elsewhere (Renedo et al., 2017b). Human hair reference material (NIES-13) and feather internal reference material (F-KP, king penguin feathers) were used for validation of feather analyses (keratin-based matrixes). Blood analyses were validated with internal reference material (RBC-KP, king penguins red blood cells) as detailed elsewhere (Renedo et al., 2018b).

Total Hg isotope analyses

Feather (and blood) samples (0.05–0.10 g) were digested with 3 or 5 mL of HNO₃ acid (65%, INSTRA quality) after a predigestion step overnight at room temperature. Hg extraction was carried out by Hotblock heating at 75 °C during 8 h (6 h in HNO₃ and 2 h more after addition of 1/3 of the total volume of H₂O₂ 30%, ULTREX quality). Hg isotopic composition was determined using cold-vapor generator (CVG)-MC-ICPMS (Nu Instruments), detailed in previous work (Renedo et al., 2018b). Hg isotopic values were reported as delta notation, calculated relative to the bracketing standard NIST SRM-3133 reference material to allow interlaboratory comparisons, as described in the SI. NIST SRM-997 thallium standard solution was used for the instrumental mass-bias correction using the exponential law. Secondary standard NIST RM-8160 (previously UM-Almadén standard) was used for validation of the analytical session (Table S1).

Recoveries of extraction were verified for all samples by checking the signal intensity obtained on the MC-ICPMS for diluted extracts relative to NIST 3133 standard (with an approximate uncertainty of ±15%). Total Hg concentrations in the extract solution were compared to the concentrations found by AMA-254 analyses to assess method recovery. Accuracy of Hg isotopic analyses for keratin matrixes was evaluated with validated human hair material NIES-13 isotopic composition (Yamakawa et al., 2016). Hg isotopic results for blood samples were validated with reference values of Lake Michigan fish tissue NIST SRM 1947. Internal reference samples of feathers (F-KP) and avian blood (RBC-KP) were also measured. Uncertainty for delta values was calculated using 2SD typical errors for each internal reference material (Table S1).

Carbon and nitrogen stable isotope analyses

Homogenized feather and blood subsamples (aliquots mass: ~0.3 mg) were weighed with a microbalance and packed in tin containers. Carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) stable isotope ratios were

determined with a continuous flow mass spectrometer (Thermo Scientific Delta V Advantage) coupled to an elemental analyser (Thermo Scientific Flash EA 1112). Results are in delta notation relative to Vienna PeeDee Belemnite and atmospheric N₂ for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, respectively. Replicate measurements of internal laboratory standards (acetanilide) indicated measurement errors < 0.15‰ for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values.

Statistical analyses

Statistical analyses were performed using the software R 3.3.2 (R Core Team, 2018) (“R Core Team, 2016: A language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria,” n.d.). Before statistical analyses, the data were checked for normality of distribution and homogeneity of variances using Shapiro–Wilk and Breusch-Pagan tests, respectively. Since data did not meet specificities of normality and homoscedasticity, non-parametrical tests (Kruskal–Wallis with Conover-Iman *post-hoc* test) were performed. Statistically significant results were set at $\alpha = 0.05$. Statistical significance of Hg concentration and isotopic differences between head and body feathers were assessed using a randomization procedure. A 99% confidence interval was calculated by means of bootstrap estimation method (n=1000 iterations).

We examined the correlations between Hg concentrations, $\delta^{13}\text{C}$, Hg MDF ($\delta^{202}\text{Hg}$) and MIF ($\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$), latitude and longitude using linear regressions and Spearman correlation rank tests. The influence of the latitude and longitude of their individual breeding and non-breeding distribution on feather Hg isotopic signatures were tested using linear mixed models (LMMs) with colonies as random effect on the whole data set, using the R package “lme4” (Douglas Bates et al., 2015). Summer latitude, summer longitude and both summer latitude + longitude together were used as predictors for Hg isotopic signatures of body feathers. Similarly, median winter latitude, median winter longitude and both median together were used as predictors of Hg isotopic signatures in head feathers. Variance inflation factors were always < 3 (Salmerón et al., 2018), ensuring that there was not collinearity between latitude and longitude in summer (breeding colonies) and median latitude and longitude in winter (wintering areas) (Zuur et al., 2009). The different models were ranked based on Akaike's Information Criteria adjusted for small sample sizes (AICc) and compared using ΔAICc and Akaike

weights (w) using the R package “wqid” (Meredith, 2020). To assess the explanative power of these models, marginal R^2 was obtained using the R package “r2glmm” (Jaeger, 2017).

Results and discussion

Seasonal and geographical variations of feather MeHg concentrations related to changing foraging habits ($\delta^{15}\text{N}$)

We observed a dominant proportion of MeHg in body feathers and blood ($94 \pm 2\%$, $n=20$ and $90 \pm 3\%$, $n=10$; respectively) for all the studied populations of little auks (Table S2). This result is in good agreement with previous studies (Alexander L Bond and Diamond, 2009; Davis et al., 2004; Renedo et al., 2018b, 2017b) and supports that both tissues of little auks principally present Hg as MeHg. Based on the different Hg integration times between types of feather of little auks, we considered that the Hg excreted in head and body feathers reflects respectively the exposure during their non-breeding and breeding periods. Since birds are known to excrete between 70 and 90% of their Hg body burden by feather moult (Honda et al., 1986b), we cannot exclude that some residual Hg accumulated during the non-breeding period could also be excreted during body feather moult, and vice versa, but this fraction would be minor. Therefore, we assumed that most of the Hg excreted through head feathers integrated the non-breeding period acquisition (October to April) whereas Hg in body feathers corresponded to the acquisition during the breeding period (May to September).

Overall, individuals presented higher Hg (MeHg) concentrations in head compared to body feathers, exhibiting up to 2-fold higher concentrations in head feathers in the case of East Greenland and Bear Island populations (Table S3). We also observed high variations of Hg concentrations in head feathers among individuals of the same colony, especially in Bear Island (from 0.81 to $4.35 \mu\text{g g}^{-1}$) and Spitsbergen populations (from 1.67 to $3.79 \mu\text{g g}^{-1}$) (Fig. S1). During the non-breeding period, little auks occupy a wide range of areas and feed on a higher prey spectrum (Jérôme Fort et al., 2010). The widespread individual foraging specialisation of little auks during the non-breeding period may be the reason for the higher variability in head feather Hg concentrations in the same colony (Jérôme Fort et al., 2010; Rosing-Asvid et al., 2013). Conversely, little auks occupy more restricted foraging areas during the breeding season due to the need to frequently feed their chicks (Welcker et al., 2009). The lower mobility

and the predominance of local prey captured near their respective colonies could lead to less intra-population variability of Hg concentrations in their body feathers.

Higher Hg concentrations of head feathers compared to body feathers of little auks are coherent with previous observations (Fort et al., 2014b) and suggest a higher exposure to MeHg during the non-breeding period outside the High Arctic. Little auks breeding in areas of Spitsbergen and East Greenland are known to mainly forage on copepods (*Calanus* spp.) during the breeding season (Harding et al., 2008). However, the seasonal vertical migration of their main prey *Calanus* spp. to inaccessible depths produces a seasonal shift in their diet towards krill *Meganyctiphanes norvegica*, hyperiid amphipods *Themisto* spp., and fish larvae (Rosing-Asvid et al., 2013). The consumption of higher trophic level prey during winter could explain the higher Hg levels excreted during the spring moult (head feathers), whereas they are probably less exposed to Hg in summer.

We observed a consistent longitudinal trend of body feather Hg concentrations ($R^2=0.57$, $p<0.0001$) with increasing Hg levels from eastern (Bear Island and FJL, 0.71 and $0.75 \mu\text{g}\cdot\text{g}^{-1}$, respectively) to western colonies (NW Greenland, $2.07 \mu\text{g}\cdot\text{g}^{-1}$). When applying mixed models, summer longitude was the most supported predictor of body feather Hg concentrations (Table S5). Head feather Hg concentrations were positively correlated both with winter latitude and longitude for the four spatially tracked populations ($R^2=0.54$ and $R^2=0.60$, respectively; both $p<0.0001$). Both variables together were considered as predictors of Hg head feather concentrations by linear mixed models (Table S6). Head feather concentrations were higher in populations wintering in western zones ($3.48 \mu\text{g}\cdot\text{g}^{-1}$, East Greenland population) and decreased gradually and significantly ($H=20.13$, $p=0.001$) in those wintering in northeast areas ($1.74 \mu\text{g}\cdot\text{g}^{-1}$, FJL population). The consistent longitudinal patterns both in summer and winter reflect a higher accumulation of MeHg in little auks from western regions, whereas colonies breeding in Arctic northern regions seem to be exposed to lower concentrations. Seabird blood $\delta^{15}\text{N}$ values provide short- to medium-term information (about 1–5 weeks) while feathers $\delta^{15}\text{N}$ values reflect the diet at the time they were grown (Bearhop et al., 2002; K. a Hobson and Clark, 1992). The distribution of little auk populations in winter was limited to the North Atlantic and the Arctic areas, where large-scale $\delta^{15}\text{N}$ values are known to be relatively homogeneous at the base of the food web

(Graham et al., 2010), then allowing the inter-population comparison. The lower body feather Hg concentrations and blood $\delta^{15}\text{N}$ values observed in little auks (Table S4) confirm that all birds from the different populations mostly feed at low trophic levels and on *Calanus* copepods in summer. Therefore, the increase of Hg concentrations in body feathers from east to west colonies is not driven by trophic effects but may be mostly related to complex Hg oceanic dynamics or distinct environmental sources that control the level of exposure to MeHg at the different breeding colonies.

Spatio-temporal trends of Hg MDF ($\delta^{202}\text{Hg}$) in feathers related to ecological aspects

Specific integration times of feather types may influence the seasonal integration of MeHg from different spatial origin. Based on this assumption, both changing seasonal behaviour and geographical environmental parameters of their foraging grounds between summer (body feathers) and winter (head feathers) could lead to different levels of MeHg exposure and induce variations of $\delta^{202}\text{Hg}$ values between types of feather.

Regarding the spatial trends on $\delta^{202}\text{Hg}$ signatures, head and body feathers showed large ranges, varying from -0.24 to 1.43‰ and from -0.11 to 1.28‰, respectively. FJL population exhibited significantly heavier $\delta^{202}\text{Hg}$ values relative to the other four populations, both in head ($H=29.42$, $p<0.0001$) and body feathers ($H=27.69$, $p<0.0001$) (Table S3). Positive linear relationships were found for body feather $\delta^{202}\text{Hg}$ values with summer latitude ($R^2=0.25$, $p<0.0001$) and summer longitude ($R^2=0.08$, $p=0.03$) (Fig 1). When applying mixed models, summer latitude was considered the explanatory variable of body feather $\delta^{202}\text{Hg}$ variations (Table S5). Head feather $\delta^{202}\text{Hg}$ signatures were positively and highly related to winter latitude ($R^2=0.51$, $p<0.0001$) and winter longitude ($R^2=0.62$, $p<0.0001$) (Fig 1). The model considering median winter latitude and longitude together is the most supported, evidencing that both variables together are drivers of head feather $\delta^{202}\text{Hg}$ trends (Table S6).

Geographical variations in $\delta^{202}\text{Hg}$ values are generally difficult to distinguish since metabolic processes also induce Hg MDF. Although we focused on the study of multiple colonies of the same seabird species to minimize the metabolic or trophic-related effects, we cannot exclude that the variability of $\delta^{202}\text{Hg}$ signatures among colonies is led only by the specific isotopic baseline of their

respective foraging habitats. For instance, it is known that little auks from FJL are morphologically bigger than those of the populations from Svalbard due to more severe climate conditions in this area (Stempniewicz et al., 1996). Thus, potentially different biological characteristics associated to their bigger size could contribute to higher feather $\delta^{202}\text{Hg}$ values in this colony. Consequently, we cannot discern spatial MeHg sources related to different migratory routes of the different populations only by the interpretation of their $\delta^{202}\text{Hg}$ signatures. The utilisation of feather $\delta^{202}\text{Hg}$ values as a proxy of geographical patterns or to changing environmental conditions requires a complete knowledge of all the processes and factors driving Hg MDF (i.e., trophic ecology and intrinsic metabolic/physiological processes).

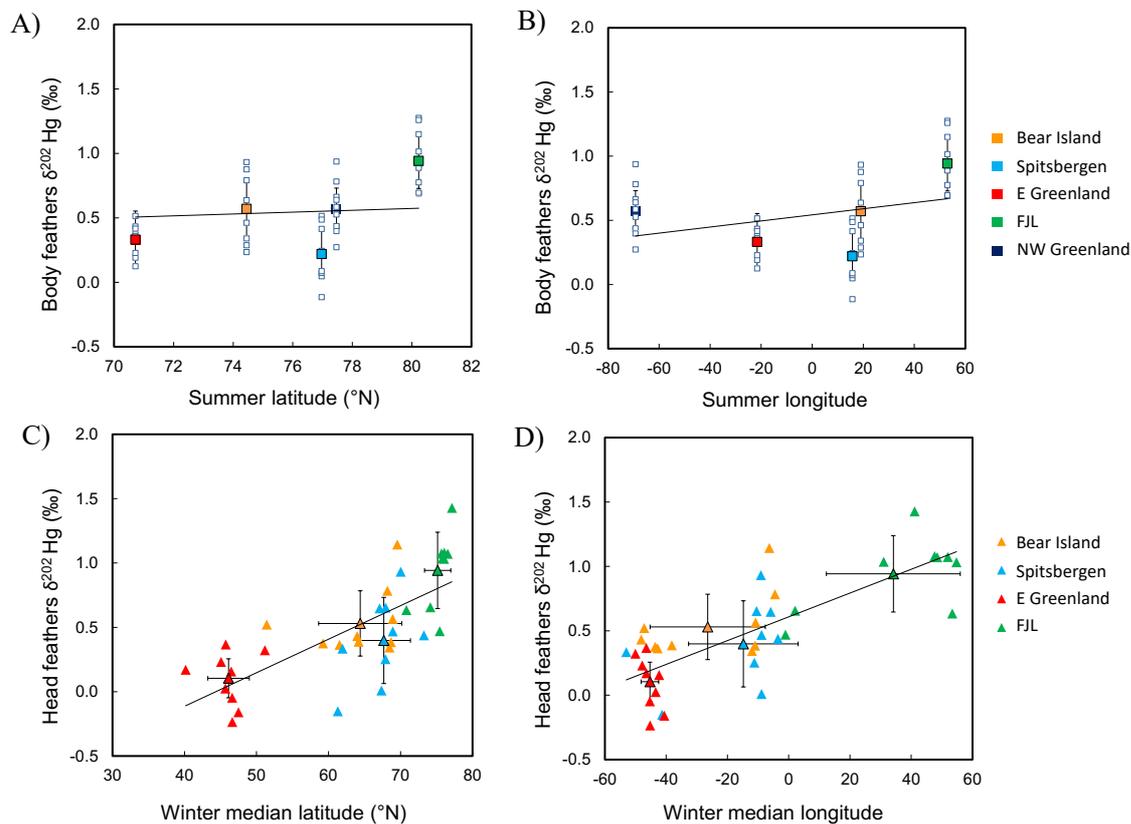


Figure 1. Hg MDF ($\delta^{202}\text{Hg}$) of little auk body feathers (summer) as a function of latitude (A) and longitude of their breeding sites (B) and head feathers (winter) as a function of the median latitude (C) and longitude (D) of their winter grounds. Regression equations are A) $y = 0.05x - 3.42$, $R^2=0.25$, $p<0.0001$; B) $y = 0.01x + 0.54$, $R^2=0.08$, $p=0.03$; C) $y = 0.02x - 0.95$, $R^2=0.50$, $p<0.0001$; D) $y = 0.01x + 0.64$, $R^2=0.62$, $p<0.0001$.

Hg odd-MIF ($\Delta^{199}\text{Hg}$): seasonal and spatial differences of Hg marine photochemistry

Head and body feather odd-MIF values ($\Delta^{199}\text{Hg}$) ranged from 0.72 to 1.26‰ and 0.90 to 1.91‰, respectively. Significantly higher $\Delta^{199}\text{Hg}$ values in body compared to head feathers (Table S7) clearly evidence a seasonal variability in odd-MIF values. This may be mainly associated to the higher extent of Hg marine photochemistry during summer (body feathers) than during winter (head feathers). In summertime/ spring, little auks are known to return to their breeding sites, located at northern latitudes, where they are exposed to longer daily photoperiod at this moment of the year (polar day). A seasonal higher angle of solar incidence at northern latitudes could affect the transmission of the UV/visible light towards the surface waters, as well as the higher magnitude of inorganic Hg or MeHg photochemical processes (Rose et al., 2015).

We observed a positive linear relationship between body feather $\Delta^{199}\text{Hg}$ values and summer longitude ($R^2=0.20$, $p<0.0001$) and between head feather $\Delta^{199}\text{Hg}$ values and winter longitude ($R^2=0.22$, $p<0.0001$) (Fig 2). Summer and winter longitudes were respectively the most supported explanatory factors of body and head feather $\Delta^{199}\text{Hg}$ values (Tables S5 and S6). No significant relationships were observed with latitude in summer ($R^2=0.01$, $p=0.20$) nor winter ($R^2=0.07$, $p=0.06$) (Fig 2).

The consistent longitudinal trends of $\Delta^{199}\text{Hg}$ values suggest different extent of Hg photochemical processes between the distant colonies and the wide-ranging spatial foraging habitats of little auks during winter. FJL population showed significantly higher $\Delta^{199}\text{Hg}$ values both in head and body feathers comparing to other studied colonies ($H=11.96$, $p=0.018$ and $H=18.55$, $p=0.001$; respectively). Potentially higher solar irradiance conditions in northern zones of the Arctic Ocean in summer could mostly contribute to more positive body feather $\Delta^{199}\text{Hg}$ values of FJL population ($1.64 \pm 0.15\%$, $n = 10$) compared to other studied colonies ($1.31 \pm 0.20\%$, $n=37$). However, FJL individuals also presented higher $\Delta^{199}\text{Hg}$ values of their head feathers ($1.13 \pm 0.06\%$) compared to the other colonies ($1.00 \pm 0.12\%$). Head feathers are assumed to integrate Hg during late autumn and winter seasons when this region is highly covered by sea ice and under scarce or even inexistent light availability. Therefore, we would expect lower $\Delta^{199}\text{Hg}$ values in little auks wintering in these northern zones compared to southern zones where the sea ice extent is lower. Previous studies on Alaskan seabirds revealed that the

presence of sea ice inhibits light penetration and strongly inhibits Hg marine photochemistry, reporting around 2-fold higher mean $\Delta^{199}\text{Hg}$ signatures in low-ice-covered oceanic areas ($1.13 \pm 0.16\%$; 56-58°N) than highly ice-covered regions ($0.53 \pm 0.15\%$; 68°N) (D Point et al., 2011). Although we observed weaker differences on $\Delta^{199}\text{Hg}$ values between FJL (80°N) and other colonies (70-77°N), our north to south trend is inverted to the one observed in Alaska. Based on our observations, we cannot presume that the presence of sea ice is the only driving factor controlling MeHg photochemistry and the related odd-MIF signatures registered in feathers of little auks. The existence of an opposed latitudinal trend of $\Delta^{199}\text{Hg}$ values between the Eastern and Western Arctic Ocean regions reveals different Hg dynamic systems, especially for Hg accumulation pathways in food webs. Further, the vertical migration of their main prey (copepods) and consequent seasonal shift on their diet to krill/amphipods during the winter season could also enhance the accumulation of pelagic MeHg that is less connected to the photic zone.

According to previous findings, the largest MeHg production in the Arctic water column seem to occur in oxic surface waters just below the productive surface layer (Heimbürger et al., 2015b; Anne L Soerensen et al., 2016). In the Arctic, additional sources of Hg and carbon are provided by sea ice algae during spring blooms (Burt et al., 2013). The presence of terrestrial organic matter and sea ice layers that concentrates phytoplankton near the MeHg production zone may favour the Hg microbial methylation at shallow depths of the Arctic water column (Gionfriddo et al., 2016; Schartup et al., 2015). Shallower methylation occurring in Arctic waters may result in higher photochemical impact on MeHg before its assimilation in Arctic biota compared to North Atlantic marine food webs. This phenomenon could contribute to the higher feather $\Delta^{199}\text{Hg}$ values of FJL little auks compared to populations breeding at lower latitudes.

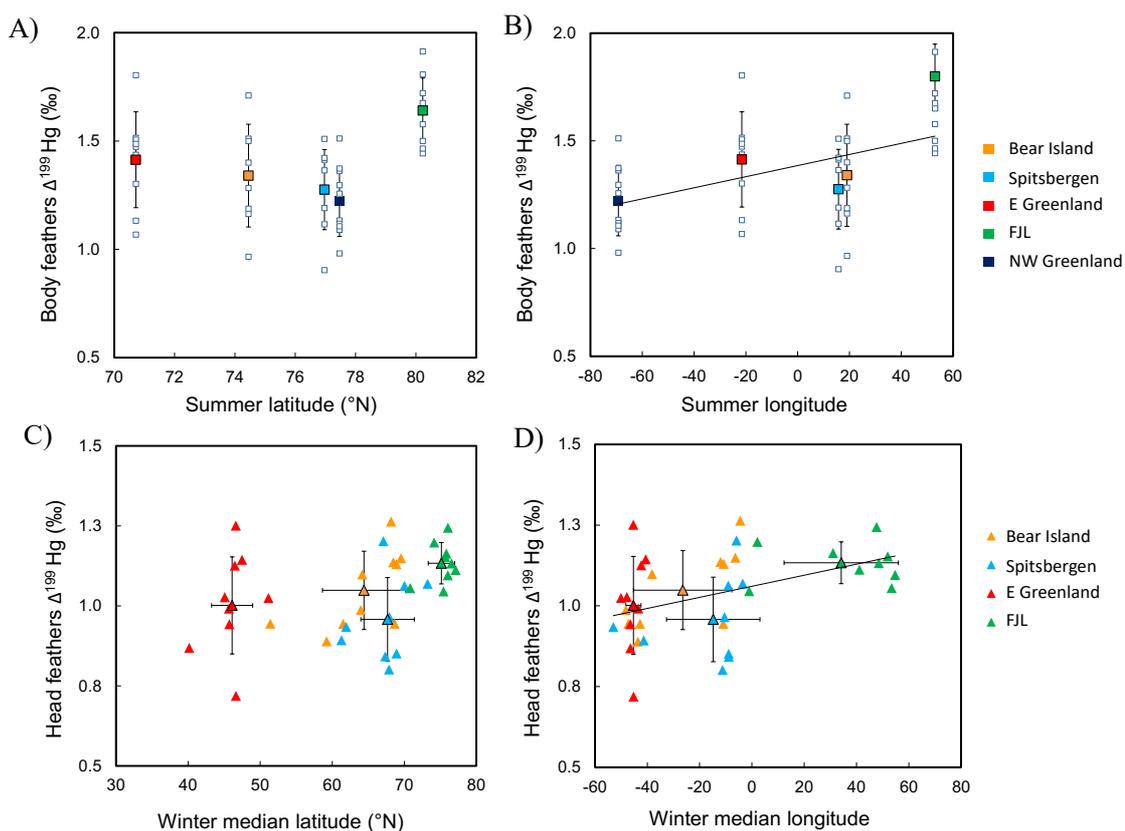


Figure 2. Hg odd-MIF ($\Delta^{199}\text{Hg}$) of little auk body feathers (summer) as a function of latitude (A) and longitude of their breeding sites (B) and head feathers (winter) as a function of the median latitude (C) and longitude (D) of their wintering grounds. Regression equations are A) $y = 0.01x + 0.32$, $R^2=0.01$, $p=0.20$; B) $y = 0.01x + 1.38$, $R^2=0.20$, $p<0.0001$; C) $y = 0.01x + 0.83$, $R^2=0.07$, $p=0.06$; D) $y = 0.01x + 1.07$, $R^2=0.22$, $p=0.002$. Regression lines presented only for significant relationship between the two variables.

Photochemical reactions of MeHg and inorganic Hg in the water column are characterized by a different $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ ratio. Therefore, this ratio is used to identify mechanisms involving MIF variations (Bergquist and Blum, 2007). $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ slopes of head and body feather samples were 1.20 ± 0.08 (SE, $R^2=0.65$, $p<0.0001$) and 1.16 ± 0.06 (SE, $R^2=0.85$, $p<0.0001$), respectively (Fig. S2). Individual $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ ratios of head and body feathers calculated for each population (Table S3) are not statistically different between colonies (respectively $H=2.030$ and $H=2.766$, both $p=0.05$). The $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ slopes of little auk feathers fell between the range of experimentally derived ratio for photodemethylation of MeHg (1.36 ± 0.04) and photoreduction of divalent Hg^{2+} (1.00 ± 0.02) in freshwater with natural dissolved organic carbon (Bergquist and Blum, 2007). $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ slope for body feathers (1.16 ± 0.06) was closer to slopes previously obtained in Arctic marine mammals ($1.07 \pm$

0.01) (Masbou et al., 2018) and Arctic seabird eggs from northern regions of the Bering Sea (1.09 ± 0.04) (Day et al., 2012a). Meanwhile, $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ slope of head feathers (1.20 ± 0.08) was closer to the slope observed in subarctic marine fish (1.29 ± 0.04) (Li et al., 2016) and Arctic seabird eggs from the Southern Bering Sea and the Gulf of Alaska (D Point et al., 2011). The lower odd-MIF slopes in northern Arctic regions (body feathers) than in subarctic or North Atlantic regions (head feathers) suggest a north to south trend on $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ ratios. The particular conditions of the Arctic Ocean may therefore drive the different photochemical pathways of MeHg (terrestrial and autotrophic organic matter, ice and melted ice/seawater photochemistry, etc.), contributing to distinct oceanic mixing sources of MeHg accumulated in Arctic food webs.

Hg stable isotopes and carbon stable isotopes ($\delta^{13}\text{C}$): terrestrial/marine Hg inputs

The deposition of atmospheric Hg from mid-latitude anthropogenic emissions into the Arctic Ocean could contribute to the accumulation of MeHg from distinct origin in Arctic-North Atlantic food webs (Larose et al., 2011). Although body feathers of little auk presented a relative high range of $\Delta^{200}\text{Hg}$ signatures (from -0.23 to 0.17‰), the inter-population differences were not significant ($H=3.685$, $p=0.45$) (Fig. S3). Low variability of $\Delta^{200}\text{Hg}$ values was observed for head feathers of little auks (-0.14 to 0.12‰). These results seem to indicate similar atmospheric sources in all the studied populations both in summer and winter areas. However, large-scale ocean circulation and vertical transport processes throughout the water column could also influence the distribution of distinct MeHg sources between the widely distributed compartments used by little auks. Seabird samples with high fractions of MeHg (e.g., blood, feathers, eggs) typically exhibit positive $\delta^{202}\text{Hg}$ values (Day et al., 2012a; D Point et al., 2011; Renedo et al., 2018b). However, some individuals from East Greenland exhibited low (including negative) feather $\delta^{202}\text{Hg}$ and slight $\Delta^{199}\text{Hg}$ values. These values are closer to those typically observed in Arctic sediments or vegetation (Day et al., 2012a; Obrist et al., 2017b). Arctic snow also show close-to-zero $\delta^{202}\text{Hg}$ values (-1 to 1‰) and extremely negative $\Delta^{199}\text{Hg}$ (-0.95 to -5.08‰) (Laura S. Sherman et al., 2010). The progressive increase of feather $\Delta^{199}\text{Hg}$ and $\delta^{202}\text{Hg}$ values from North Atlantic to High

Arctic Regions of little auks (Fig. S4) could therefore suggest a gradient of MeHg sources and progressive increase of photochemical demethylation of MeHg.

The exploration of carbon stable isotopes ($\delta^{13}\text{C}$) of little auks could help discriminating the potential contributions of distinct MeHg sources (terrestrial vs marine) probably linked to the widely specific foraging habitats of little auks. Contrary to Hg isotopes, body feather $\delta^{13}\text{C}$ values do not reflect the period of summer but the moult period in late summer/early autumn (September) when they are grown (Fort et al., 2013a). To ensure only the integration of the summer, breeding period, we compared body feather Hg isotopes with blood $\delta^{13}\text{C}$ values. Little auks from FJL exhibited the lowest blood $\delta^{13}\text{C}$ values ($-23.13 \pm 0.84\text{‰}$) and NW Greenland individuals the highest ($-20.07 \pm 0.35\text{‰}$) relative to the rest of the colonies ($H=40.74$, $p<0.0001$) (Table S4). Head feather $\delta^{13}\text{C}$ values separated little auk populations in those overwintering in western areas of the North Atlantic Ocean and those wintering in north-eastern areas ($H=26.28$, $p<0.0001$). Significant negative linear relationships were obtained for both $\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ of body feathers with blood $\delta^{13}\text{C}$ values ($R^2=-0.447$, $p=0.002$ and $R^2=-0.558$, $p<0.0001$) (Fig 3). No relationship was observed between $\delta^{202}\text{Hg}$ and $\delta^{13}\text{C}$ values of head feathers ($R^2=-0.025$, $p=0.863$) but it was significant for $\Delta^{199}\text{Hg}$ and $\delta^{13}\text{C}$ values ($R^2=-0.350$, $p=0.017$) (Fig 3). Interestingly, the negative relationship between $\Delta^{199}\text{Hg}$ and $\delta^{13}\text{C}$ values of little auks contrasts with those previously reported on eggs from guillemot species (or murre, *Uria aalge* and *U. lomvia*) breeding in the Alaskan Arctic (Day et al., 2012a). These authors reported a co-enrichment of egg $\delta^{13}\text{C}$, $\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ values linked to the transition from terrestrial to marine Hg sources and the subsequent reduction of Hg photochemistry in coastal reservoirs due to higher turbidity (Day et al., 2012a). However, the wintering areas of little auks mainly correspond to more opened oceanic areas as the study in the Bering Sea and probably do not present such a remarkable coastal-oceanic gradient. Our negative trend between Hg isotopes and $\delta^{13}\text{C}$ seems to be dominated by higher magnitude of Hg photochemical processes at northern Arctic regions and a potential transition of Hg inputs (terrestrial/riverine to marine inputs) from High Arctic to North Atlantic waters.

Head feather $\delta^{13}\text{C}$ values of little auks wintering in north-eastern Arctic regions, i.e. FJL (-20.59‰) and Bear Island (-20.35‰), were close to values of Arctic sediments such as the Yermak

Plateau (-23.0‰) and central Arctic Ocean (-21.4‰) (Carsten J. Schubert and Calvert, 2001). The FJL archipelago and surrounding waters are highly covered by glaciers and extensive year-round sea-ice cover (Grémillet et al., 2015). These high Arctic areas are strongly impacted by the Makarov and Arctic cold currents flowing southward from the north and potentially transporting terrigenous organic matter from river discharge and from coastal erosion by sea ice and glaciers (Winkelmann and Knies, 2005b). Current high rates of annual glacial melting in the European Arctic fjord ecosystems are favouring the release of organic matter originating from the meltwater or terrestrial materials in these regions (Kim et al., 2020). Besides, recent observations reported higher concentrations of total dissolved Hg in the Arctic Ocean compared to the North Atlantic Ocean (Cossa et al., 2017a). These observations support an important supply of terrestrial Hg entering the High Arctic marine systems. Contrarily, head feather $\delta^{13}\text{C}$ values of little auks from Northwest (-19.41 ‰) and East Greenland (-16.61‰) were closer to marine carbon endmembers, such as particulate organic matter of meltwater ponds and Arctic ice-cores (-15.3 to -20.6‰) (Carsten J. Schubert and Calvert, 2001). Indeed, these populations winter in regions near the Newfoundland Island (Fort et al., 2013a), where the North Atlantic Current appears to enhance the supply of marine organic matter (Fontela et al., 2016b) and Hg (Lamborg et al., 2014a) from deeper layers. It is known that North Atlantic deep-waters are enriched in Hg relative to other oceans as a result of the incorporation of anthropogenic inputs (Lamborg et al., 2014a). Therefore, the contribution of Hg from deep-water masses in Atlantic regions could explain the isotopically distinct MeHg accumulated in head feathers of little auks wintering in these areas.

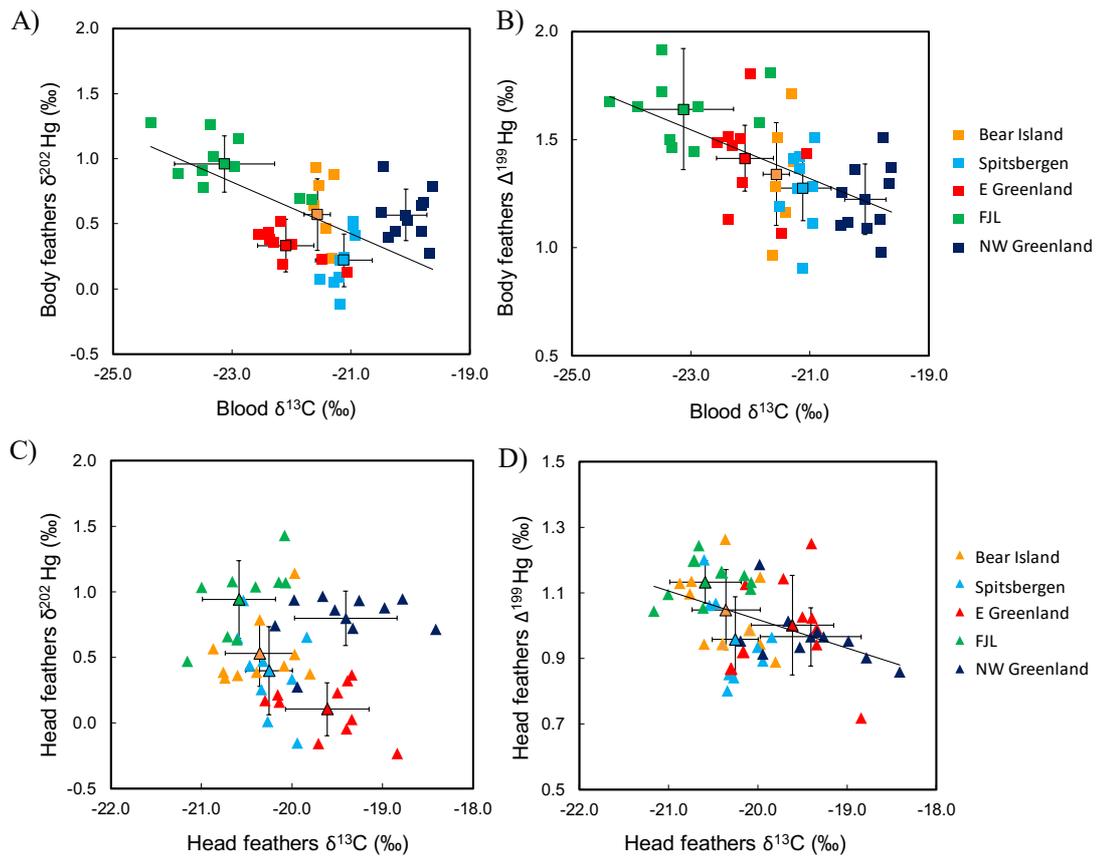


Figure 3. Carbon ($\delta^{13}\text{C}$) vs MDF and MIF Hg signatures for summer (body feathers) and winter (head feathers) periods. Regression equations are A) $y = -0.20x + 3.75$, $R^2 = -0.45$, $p = 0.002$; B) $y = -0.12x - 1.05$, $R^2 = -0.56$, $p < 0.0001$; C) $y = -0.06x - 0.78$, $R^2 = -0.025$, $p = 0.863$; D) $y = -0.09x - 0.72$, $R^2 = -0.35$, $p = 0.017$. Regression lines presented only for significant relationship between the two variables.

Geographically distinct Hg source mixing across the Arctic and North Atlantic Oceans

Our results suggest that the longitudinal variability of Hg concentrations, stable isotopes ($\Delta^{199}\text{Hg}$, $\delta^{202}\text{Hg}$) and $\delta^{13}\text{C}$ values of little auks is linked to the assimilation of isotopically distinct MeHg depending on their wintering grounds, resulting from higher contribution of terrestrial Hg inputs in High Arctic regions and higher supply of Hg marine inputs by Hg-enriched deep waters in North Atlantic regions. Fig. 4 shows a compilation of Hg odd-MIF values observed in little auks compared to previous studies in Arctic biota over a wide longitudinal scale. The observed isotopic spatial variability across the different regions of the Arctic Ocean suggests the existence of two different Hg systems between East (Atlantic) and West (Pacific) Arctic Ocean regions. Our opposed latitudinal trend of $\Delta^{199}\text{Hg}$ values relative to Western Arctic compartments (Masbou et al., 2015; D Point et al., 2011) indicates that the presence of sea ice cover is not the only driving factor controlling Hg photochemistry in the Eastern

Arctic Ocean. Possibly, an additional supply of Hg and carbon sources by sea-ice algae may enhance the microbial/photochemical methylation and demethylation processes at shallower depths (Heimbürger et al., 2015b; Anne L Soerensen et al., 2016) in East Arctic regions, therefore contributing to the higher odd-MIF values (and associated MDF) of Hg accumulated in biota. Besides, the progressive enrichment of $\delta^{13}\text{C}$ values from Arctic regions to North Atlantic waters used by little auks corroborates higher terrestrial Hg supply from northern Arctic regions in Eastern Arctic environments. Our close-to-zero $\Delta^{200}\text{Hg}$ values are slightly lower than those obtained in Western Arctic biota (Masbou et al., 2018) and seem to indicate a higher contribution of Hg inputs deriving from riverine/terrestrial compartments in Eastern Arctic regions. The inversed relationship of $\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$ values of little auks with latitude is opposed to the latitudinal covariation of $\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$ in biota from Western Arctic regions (Masbou et al., 2018) and from Antarctic regions (Renedo et al., n.d.) and evidences a completely different functioning of Hg cycling compared to other polar marine environments. Complex Hg dynamics and ocean control factors seem to drive the longitudinal increasing pattern of Hg isotopes from west to east regions of the Arctic Ocean. Future research assessing large scale and long-term Hg contamination are necessary to have a complete understanding of the Hg exposure pathways and of the associated risks for the whole marine Arctic environments.

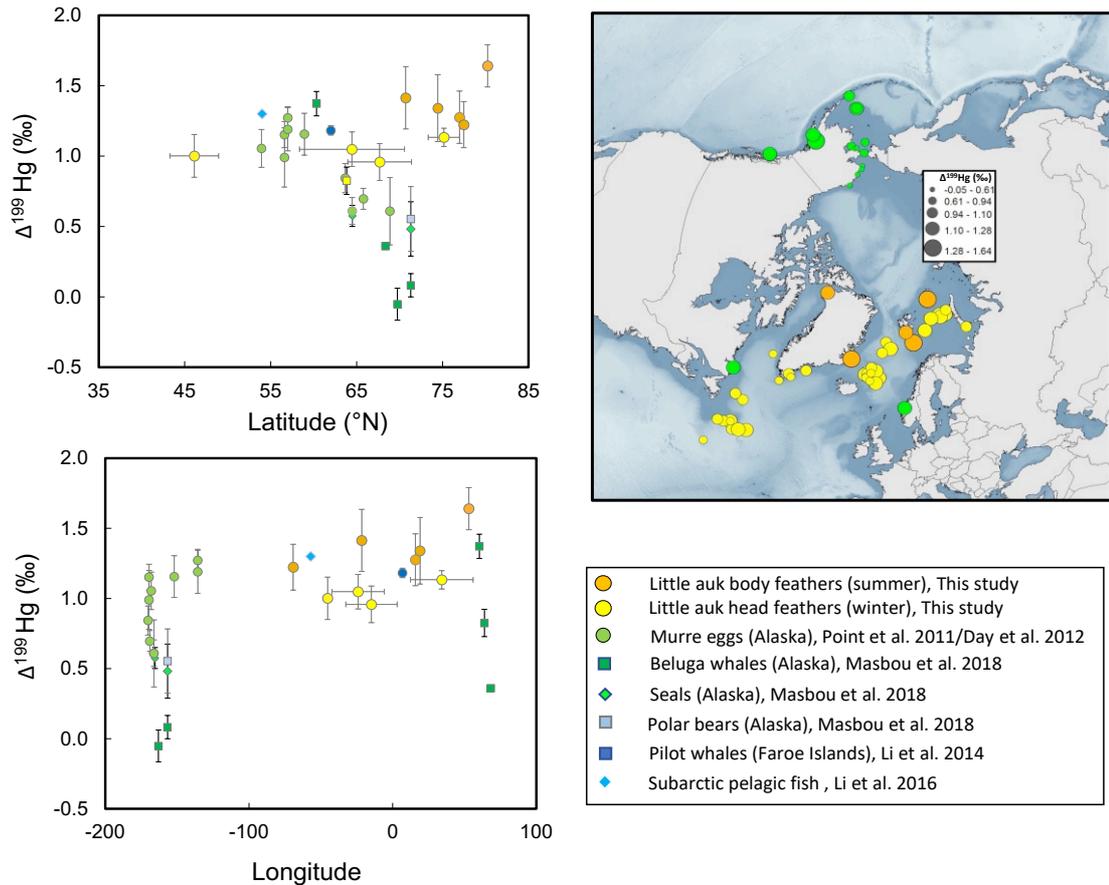


Figure 4. Compilation of Hg odd-MIF values ($\Delta^{199}\text{Hg}$) of marine biota from spatially distant Arctic Regions. The map comprises both little auk breeding sites (orange), individual little auk median winter positions (yellow) and previous published data (green) including seabirds (Day et al., 2012a; D Point et al., 2011), beluga whale, seals and polar bears (Masbou et al., 2018) from Alaskan Regions; pilot whales from Faroe Islands (Li et al., 2014a), and fish from the Labrador Sea (Li et al., 2016).

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