Environmental Research 177 (2019) 108588

Contents lists available at ScienceDirect

Environmental Research

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

Review article

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

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ARTICLE INFO

Keywords:
Seabirds
Spatial ecotoxicology
Molting pattern
Mercury

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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https://doi.org/10.1016/j.envres.2019.108588
Received 8 March 2019; Received in revised form 14 July 2019; Accepted 15 July 2019
Available online 15 July 2019

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 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.

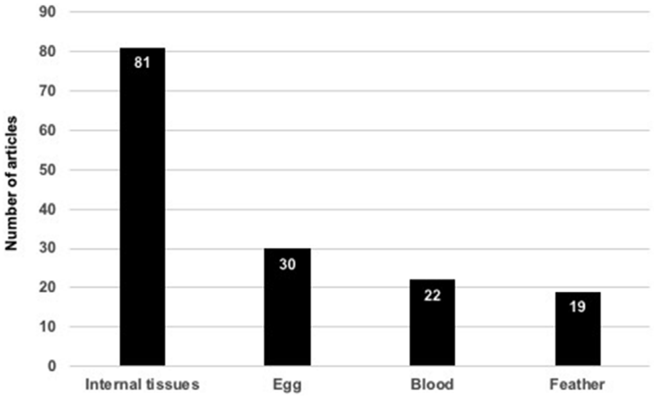

Fig. 1. Occurrence of tissues analysed for Hg in Arctic seabirds in the 101 reviewed articles. Some publications studied several tissues. Internal tissues comprise liver (n = 38), kidneys (n = 10), muscle (n = 25), heart (n = 1), bones (n = 1), brain (n = 4), gonads (n = 1) and lungs (n = 1). Feathers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

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 seaducks (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 (*Polysticta stelleri*), the spectacled eider (*Somateria fischeri*) and the long-

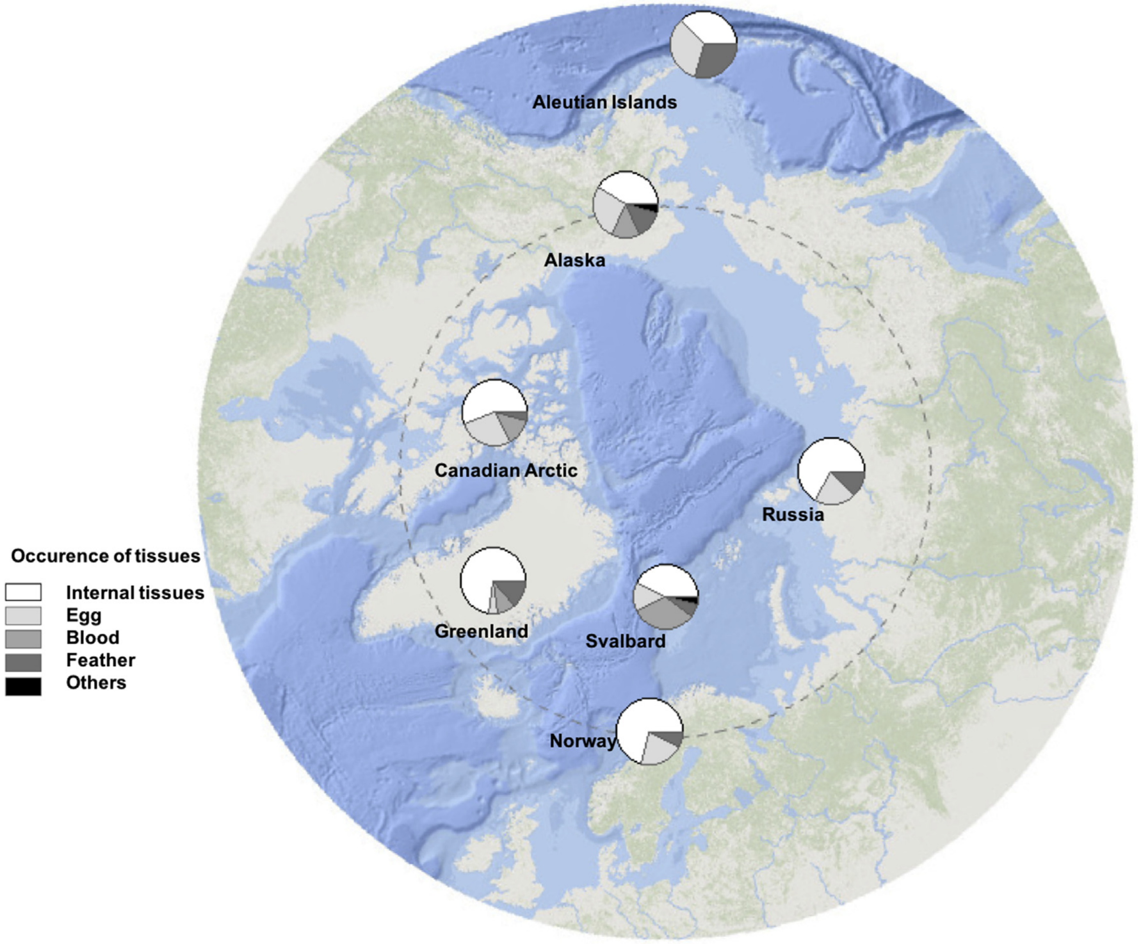

Fig. 2. Occurrence of tissues analysed for Hg in Arctic seabirds in the 101 reviewed articles, by Arctic region. Some publications studied several tissues. Internal tissues comprise liver (n = 38), kidneys (n = 10), muscle (n = 25), heart (n = 1), bones (n = 1), brain (n = 4), gonads (n = 1) and lungs (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 cirrhate*)), 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 µg/g dw mean ± SD or SE; red blood cell µg/g dw mean ± 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 nmol/g to µg/g, b overall sample size = 105.

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

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Table 1 (*continued*)

Family	Species	Sampling year	Colony location	Country	Whole blood	Red blood cell	References
	Ivory gull (*Pagophila eburnea*)	April 2012	Barentsøya	Svalbard	0.81 ± 0.05 (SD) (4)		Lucia et al. (2016)
	Ivory gull (*Pagophila eburnea*)	April 2013	Barentsøya	Svalbard	1.06 ± 0.16 (SD) (5)	1.39 ± 0.16 (SD) (40)	Lucia et al. (2016)
	Ivory gull (*Pagophila eburnea*)	April 2014	Barentsøya	Svalbard		1.20 ± 0.07 (SD) (42)	Lucia et al. (2016)
Procellariid	Northern fulmar (*Fulmarus glacialis*)	NA	Prince Leopold Island	Canada	3.62 ± 2.06 (SD) (7)		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 ± 0.04 µg/g dw (long-tailed duck, Franson et al., 2004) to 2.25 ± 0.29 µg/g dw (common eider, Provencher et al., 2017) (Table 1, Figs. 3 and 4) with an average value of 1.04 ± 0.33 µg/g dw. For larids, concentrations ranged from 0.81 ± 0.05 µg/g dw (ivory gull, Lucia et al., 2016) to 3.04 ± 1.41 µg/g dw (black-legged kittiwake, Mallory et al., 2018) (Table 1, Figs. 3 and 4) with an average value of 1.57 ± 0.48 µg/g dw. Finally, Hg concentrations in alcid blood samples ranged from 0.32 ± 0.80 µg/g dw to 4.58 µg/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 ± 1.58 µg/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 µg/g ww, equivalent to approximately 9.6 µg/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 ($\delta^{15}N$) 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 (*Uria lomvia*)	1992 & 1993	Hornøya	Norway	0.78 ± 0.18 (SD) (14)	Wenzel and Gabrielsen (1995)
	Common guillemot (*Uria aalge*)	1992 & 1993	Hornøya	Norway	0.88 ± 0.19 (SD) (10)	Wenzel and Gabrielsen (1995)
	Kittlitz's murrelet (*Brachyramphus brevirostris*) ADULT	August 2008–2011	Agattu	Aleutian Islands	1.66 ± 1.28 (SE) (29) a, b	Kaler et al. (2014)
	Kittlitz's murrelet (*Brachyramphus brevirostris*) CHICK	August 2008–2011	Agattu	Aleutian Islands	1.47 ± 0.67 (SE) (8) a, b	Kaler et al. (2014)
	Kittlitz's murrelet (*Brachyramphus brevirostris*)	2008–2011	Agattu	Aleutian Islands	2.06 ± 1.28 (SD) (37)	Kenney et al. (2018)
	Kittlitz's murrelet (*Brachyramphus brevirostris*)	2011	Adak Island	Aleutian Islands	5.15 ± 2.51 (SD) (7)	Kenney et al. (2018)
	Little auk (*Alle alle*)	2007	Kap Höegh	Greenland	1.00 ± 0.22 (SD) (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2008	Kap Höegh	Greenland	1.13 ± 0.24 (SD) (19)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2009	Kap Höegh	Greenland	1.04 ± 0.26 (SD) (19)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2010	Kap Höegh	Greenland	1.39 ± 0.73 (SD) (49)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2011	Kap Höegh	Greenland	1.84 ± 0.85 (SD) (18)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2012	Kap Höegh	Greenland	1.25 ± 0.51 (SD) (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2013	Kap Höegh	Greenland	1.69 ± 0.80 (SD) (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2014	Kap Höegh	Greenland	1.15 ± 0.33 (SD) (19)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2015	Kap Höegh	Greenland	1.25 ± 0.53 (SD) (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2016	Kap Höegh	Greenland	1.60 ± 0.62 (SD) (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2007	Kap Höegh	Greenland	1.00 ± 0.22 (SD) (20)	Fort et al. (2016)
	Little auk (*Alle alle*)	2008	Kap Höegh	Greenland	1.13 ± 0.24 (SD) (19)	Fort et al. (2016)
	Little auk (*Alle alle*)	2009	Kap Höegh	Greenland	1.04 ± 0.26 (SD) (19)	Fort et al. (2016)
	Little auk (*Alle alle*)	2010	Kap Höegh	Greenland	1.37 ± 0.70 (SD) (45)	Fort et al. (2016)
	Little auk (*Alle alle*)	2011	Kap Höegh	Greenland	1.70 ± 0.66 (SD) (17)	Fort et al. (2016)
	Little auk (*Alle alle*)	2012	Kap Höegh	Greenland	1.25 ± 0.51 (SD) (20)	Fort et al. (2016)
	Little auk (*Alle alle*)	2013	Kap Höegh	Greenland	1.69 ± 0.80 (SD) (20)	Fort et al. (2016)
	Little auk (*Alle alle*)	2014	Kap Höegh	Greenland	2.11 ± 1.49 (SD) (20)	Fort et al. (2016)
	Little auk (*Alle alle*)	2010–2011	Kap Höegh	Greenland	1.53 ± 0.84 (SD) (78)	Fort et al. (2014)
	Pigeon guillemot (*Cepphus columba*)	July 2004	Aleutian Islands	Aleutian Islands	7.11 ± 0.66 (SE) (38) a	Burger et al. (2009)
	Pigeon guillemot (*Cepphus columba*)	June 2004	Amchitka Island	Aleutian Islands	7.72 ± 0.84 (SE) (21) a	Burger et al. (2007)
	Pigeon guillemot (*Cepphus columba*)	June 2004	Kiska Island	Aleutian Islands	6.36 ± 1.03 (SE) (17) a	Burger et al. (2007)
	Pigeon guillemot (*Cepphus columba*)	2004	Prince William Sound	Alaska	2.81 ± 0.25 (SE) (40) a	Burger et al. (2007)
	Tufted puffin (*Fratercula cirrhata*)	July 2004	Aleutian Islands	Aleutian Islands	2.54 ± 0.19 (SE) (39) a	Burger et al. (2009)
	Tufted puffin (*Fratercula cirrhata*)	July 2004	Amchitka Island	Aleutian Islands	2.59 ± 0.29 (SE) (22)	Burger et al. (2009)
	Tufted puffin (*Fratercula cirrhata*)	July 2004	Kiska Island	Aleutian Islands	2.48 ± 0.25 (SE) (17)	Burger et al. (2009)
Anatid	Common eider (*Somateria mollissima*)	July 2007	Amchitka Island	Aleutian Islands	0.89 ± 0.16 (SE) (22) F a	Burger et al. (2008b)
	Common eider (*Somateria mollissima*)	July 2007	Kiska Island	Aleutian Islands	1.24 ± 0.32 (SE) (8) F a	Burger et al. (2008a)
	Common eider (*Somateria mollissima*)	July 2004	Aleutian Islands	Aleutian Islands	0.84 ± 0.08 (SE) (26) a	Burger et al. (2009)
	Common eider (*Somateria mollissima*)	July 2004	Amchitka Island	Aleutian Islands	0.85 ± 0.14 (SE) (13)	Burger et al. (2009)
	Common eider (*Somateria mollissima*)	July 2004	Kiska Island	Aleutian Islands	0.83 ± 0.08 (SE) (13)	Burger et al. (2009)
	Common eider (*Somateria mollissima*)	2011	Storholmen, Kongsfjorden	Svalbard	0.50–1.90 µg/g ww (29) F	Fenstad et al. (2017)
	King eider (*Somateria spectabilis*)	2014	Utqiagvik	Alaska	0.80 ± 0.37 (SD) (28) F	Miller et al. (2019)
	King eider (*Somateria spectabilis*)	Jun_July 1993	Chaun	Russia	0.57 (1)	Kim et al. (1996a, 1996b)
	Long-tailed duck (*Clangula hyemalis*)	Jun_July 1993	Chaun	Russia	1.96 ± 3.31 (SD) (8)	Kim et al. (1996a, 1996b)
	Spectacled eider (*Somateria fischeri*)	2010–2014	Utqiagvik	Alaska	1.35 ± 1.00 (SD) (42) F	Miller et al. (2019)
Hydrobatid	Fork-tailed storm-petrel (*Oceanodroma furcata*)	August 2008–2011	Agattu	Aleutian Islands	6.70 ± 1.63 (SE) (12) a, b	Kaler et al. (2014)
Larid	Arctic tern (*Sterna paradisae*)	Jun_July 1993	Chaun	Russia	0.89 ± 0.15 (SD) (10)	Kim et al. (1996a, 1996b)
	Black-legged kittiwakes (*Rissa tridactyla*)	2004	Prince William Sound	Alaska	2.91 ± 0.19 (SE) (61) a	Burger et al. (2008b)
	Black-legged kittiwakes (*Rissa tridactyla*) ADULT	1992 & 1993	Hornoya	Norway	2.03 ± 0.40 (SD) µg g-1 ww (27)	Wenzel and Gabrielsen (1995)
	Black-legged kittiwakes (*Rissa tridactyla*) FLEDGLING	1992 & 1993	Hornoya	Norway	0.55 ± 0.10 (SD) µg g-1 ww (10)	Wenzel and Gabrielsen (1995)
	Glaucous gull (*Larus hyperboreus*)	Jun_July 1993	Chaun	Russia	5.96 (1)	Kim et al. (1996a, 1996b)
	Glaucous-winged gulls (*Larus glaucescens*)	July 2004	Aleutian Islands	Aleutian Islands	3.68 ± 0.37 (SE) (63) a	Burger et al. (2009)
	Glaucous-winged gulls (*Larus glaucescens*) ADULT	NA	Adak Island	Aleutian Islands	2.97 ± 0.57 (SE) (16) a	Burger et al. (2009)
	Glaucous-winged gulls (*Larus glaucescens*) ADULT	NA	Amchitka Island	Aleutian Islands	4.01 ± 0.54 (SE) (30) a	Burger et al. (2009)
	Glaucous-winged gulls (*Larus glaucescens*) ADULT	NA	Kiska Island	Aleutian Islands	3.76 ± 0.86 (SE) (17) a	Burger et al. (2009)
	Glaucous-winged gulls (*Larus glaucescens*) CHICKS	NA	Amchitka 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
	Glaucous-winged gulls (*Larus glaucescens*) CHICKS	NA	Kiska Island	Aleutian Islands	1.68 ± 0.23 (SE) (15) a	Burger et al. (2009)
	Herring gull (*Larus argentatus*)	June 1993	Chaun	Russia	6.10 ± 4.60 (SD) (5)	Kim et al. (1996a, 1996b)
	Herring gull (*Larus argentatus*)	Jun_July 1993	Chaun	Russia	6.06 ± 4.60 (SD) (6)	Kim et al. (1996a, 1996b)
	Ivory gull (*Pagophila eburnea*)	April 2011	Barentsøya	Svalbard	2.65 ± 0.42 (SD) (19)	Lucia et al. (2016)
	Ivory gull (*Pagophila eburnea*)	April 2012	Barentsøya	Svalbard	3.03 ± 0.41 (SD) (40)	Lucia et al. (2016)
	Ivory gull (*Pagophila eburnea*)	April 2013	Barentsøya	Svalbard	2.79 ± 0.24 (SD) (47)	Lucia et al. (2016)
	Ivory gull (*Pagophila eburnea*)	April 2014	Barentsøya	Svalbard	3.39 ± 0.30 (SD) (39)	Lucia et al. (2016)
	Ivory gull (*Pagophila eburnea*)	2010	Seymour Island	Canada	11.66 ± 6.52 (SD) (8)	Mallory et al. (2015)
	Sabine's gull (*Xema sabin*)	Jun_July 1993	Chaun	Russia	1.70 ± 0.47 (SD) (2)	Kim et al. (1996a, 1996b)
Stercorariid	Long-tailed jaeger (*Stercorarius longicaudus*)	Jun_July 1993	Chaun	Russia	1.95 ± 0.54 (SD) (5)	Kim et al. (1996a, 1996b)
	Parasitic jaeger (*Stercorarius parasiticus*)	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 bounds 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 ± SD µg/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 (*Uria lomvia*)	NA	Prince Leopold Island	Canada	1.94 ± 0.63 (10) a	Mallory et al. (2015)
	Kittlitz's murrelet (*Brachyramphus brevirostris*)	2011	Adak Island	Aleutian Islands	37.18 (1) b	Kenney et al. (2018)
Anatid	Long-tailed duck (*Clangula hyemalis*)	June 1993	Chaun	Russia	0.70 ± 0.20 (5) a	Kim et al., 1996a, 1996b
	Common eider (*Somateria mollissima*)	NA	Cape Dorset, Nunavut	Canada	0.59 ± 0.21 (10) a	Mallory et al. (2015)
Larid	Herring gull (*Larus argentatus*)	June 1993	Chaun	Russia	6.10 ± 4.60 (5) a	Kim et al., 1996a, 1996b
	Arctic tern (*Sterna paradisae*)	June 1993	Chaun	Russia	0.90 ± 0.10 (5) a	Kim et al., 1996a, 1996b
	Black-legged kittiwakes (*Rissa tridactyla*)	NA	Prince Leopold Island	Canada	3.58 ± 0.92 (2) a	Mallory et al. (2015)
	Ivory gull (*Pagophila eburnea*)	NA	Seymour Island	Canada	15.79 ± 14.13 (8) a	Mallory et al. (2015)
	Glaucous gull (*Larus hyperboreus*)	NA	Nasaruvaalik Island	Canada	2.31 ± 1.68 (4) a	Mallory et al. (2015)
Procellariid	Northern fulmar (*Fulmarus glacialis*)	NA	Prince Leopold Island	Canada	2.71 ± 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; Toochin 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 ± 0.19 µg/g dw), followed by alcids (2.20 ± 1.23 µg/g dw) and larids (3.53 ± 1.74 µg/g dw).

More specifically, while Hg concentrations measured in female anatids (for which body feathers represent yearly local exposure) did not exceed 2 µg/g dw and showed little intra- and interspecific variations, alcids (for which body feathers represent contamination during breeding period only), reached concentrations of 7 µg/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 ± 1.49 µg/g dw; Fort et al., 2016; Amélineau et al., 2019) feed mostly on zooplankton, while tufted puffins (2.48 ± 0.25 to 2.59 ± 0.29 µg/g dw; Burger and Gochfeld, 2009) and Kittlitz's murrelets (from 1.66 ± 1.28 (Kaler et al., 2014) to 5.15 ± 2.51 µg/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 ± 0.19 µg/g dw; Wenzel and Gabrielsen, 1995), Brünnich's (0.78 ± 0.18 µg/g dw; Wenzel and Gabrielsen, 1995), and pigeon guillemots (7.72 ± 0.84 µg/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

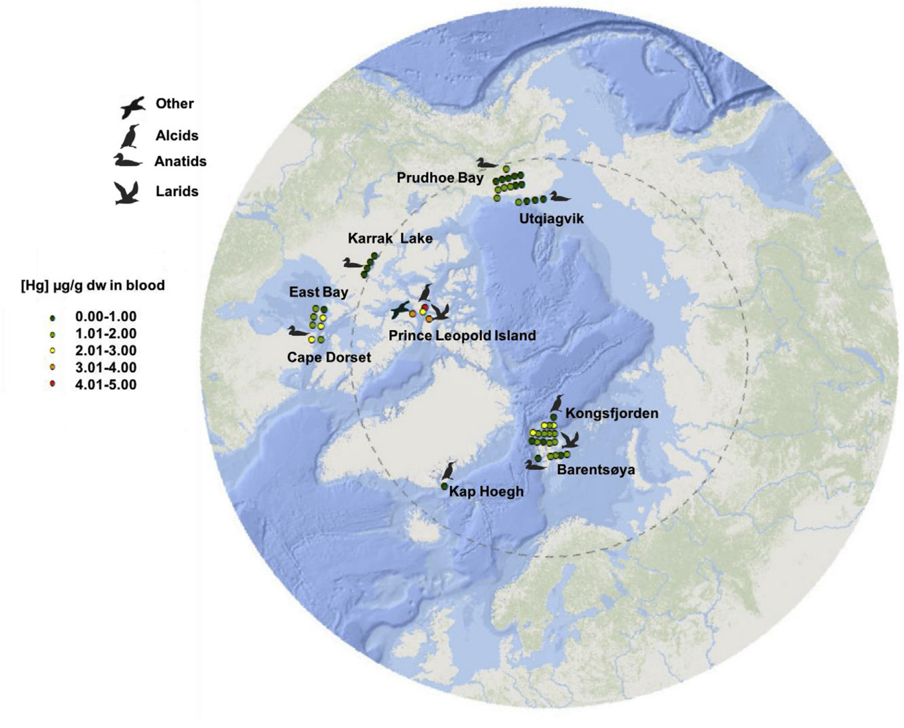

Fig. 3. Spatial distribution of Hg concentrations measured in blood.

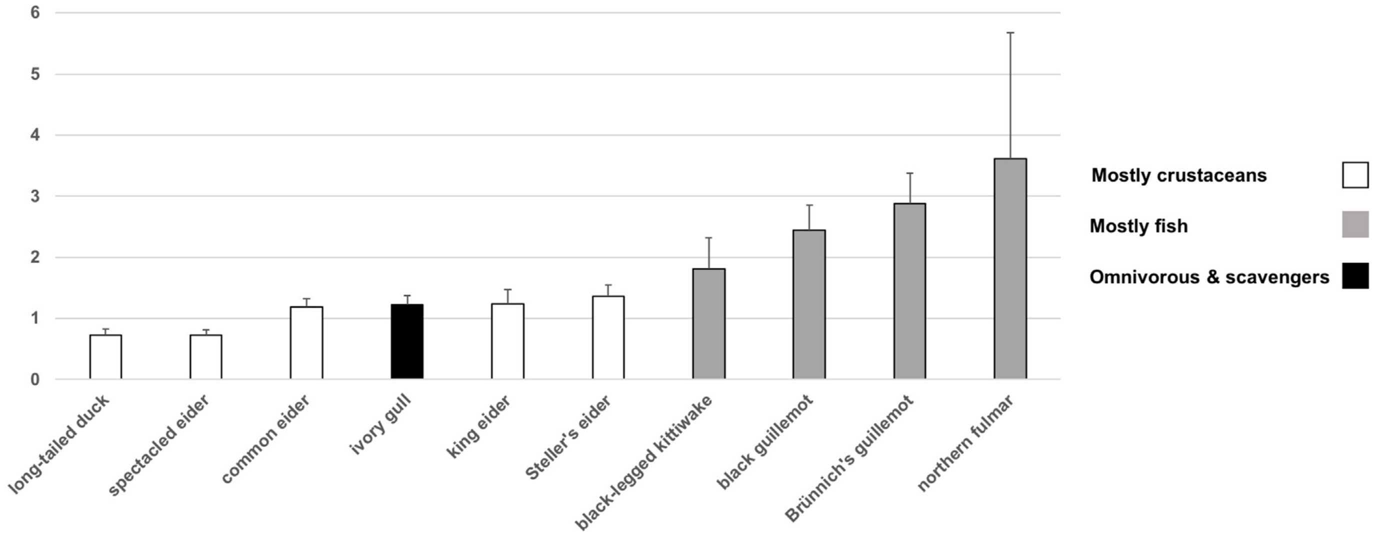

Fig. 4. Mean Hg concentrations (mean ± SD μg/g dw) measured in blood by species, in relation to bird diet during the breeding season.

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 ± 0.15 μg/g dw (Arctic tern, Kim et al., 1996a, 1996b) to 11.66 ± 6.52 μg/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

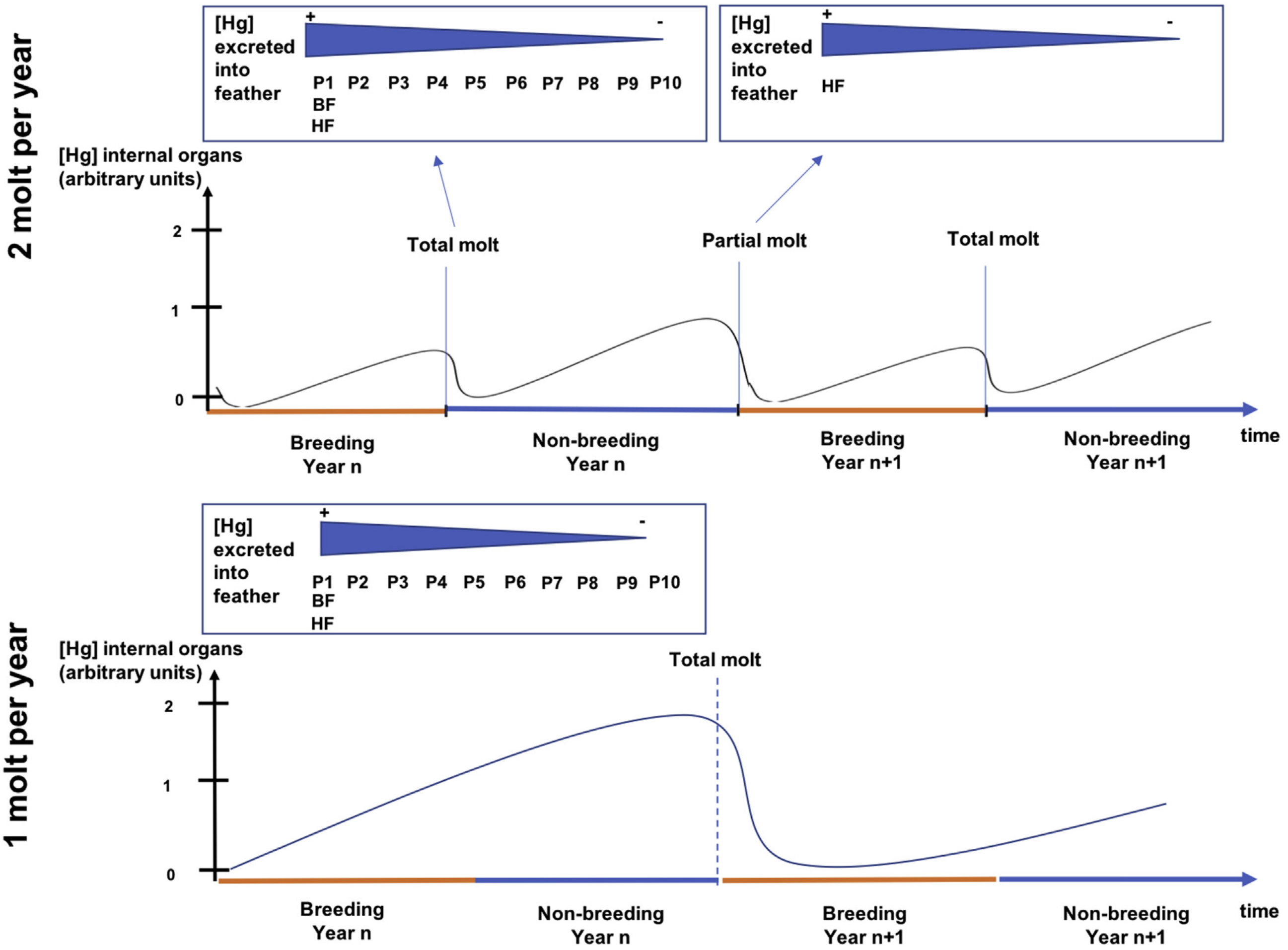

Fig. 5. Schematic representation of Hg excretion from internal tissues and organs to body, head and primary feathers for seabirds with one and two molts per year.

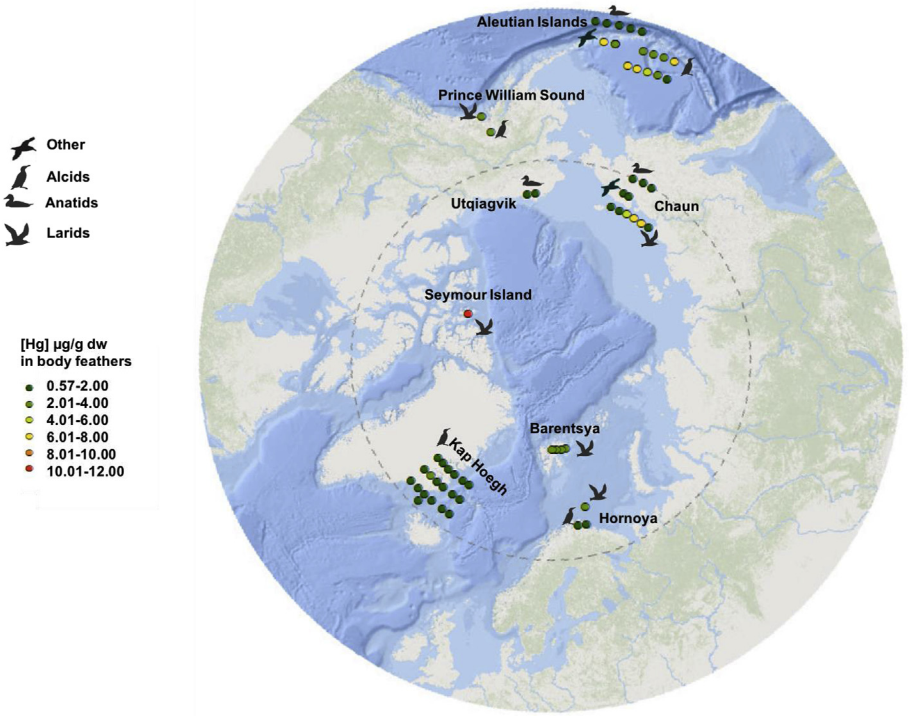

Fig. 6. Spatial distribution of Hg concentrations in body feathers.

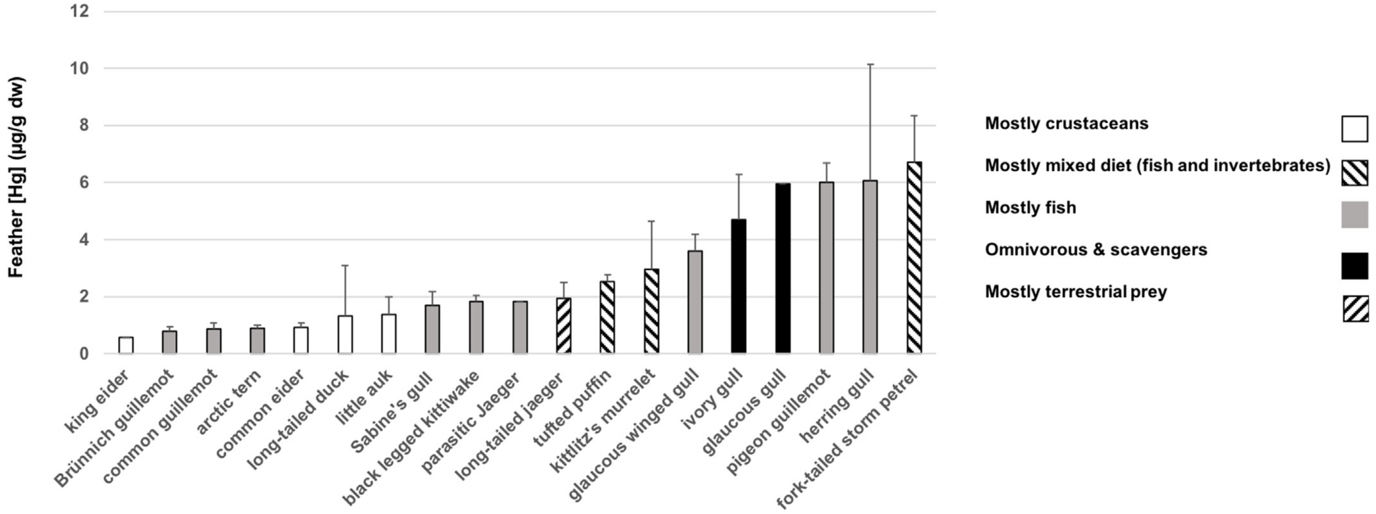

Fig. 7. Mean Hg concentrations (mean ± SD µg/g dw) measured in body feathers, by species, in relation to bird diet during the breeding season.

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 ± 1.63 µg/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 µg/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 ± SD μg/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 (*Alle alle*)	2007	Kap Hoegh (East Greenland)	Greenland	3.73 ± 1.33 (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2008	Kap Hoegh (East Greenland)	Greenland	2.86 ± 0.71 (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2009	Kap Hoegh (East Greenland)	Greenland	3.06 ± 0.99 (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2010	Kap Hoegh (East Greenland)	Greenland	3.17 ± 0.82 (40)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2011	Kap Hoegh (East Greenland)	Greenland	3.21 ± 0.99 (25)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2012	Kap Hoegh (East Greenland)	Greenland	2.27 ± 0.41 (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2013	Kap Hoegh (East Greenland)	Greenland	2.75 ± 0.87 (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2014	Kap Hoegh (East Greenland)	Greenland	3.99 ± 1.96 (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2015	Kap Hoegh (East Greenland)	Greenland	3.06 ± 0.86 (20)	Amélineau et al. (2019)
	Little auk (*Alle alle*)	2007	Kap Hoegh (East Greenland)	Greenland	3.73 ± 1.33 (20)	Fort et al. (2016)
	Little auk (*Alle alle*)	2008	Kap Hoegh (East Greenland)	Greenland	2.86 ± 0.71 (20)	Fort et al. (2016)
	Little auk (*Alle alle*)	2009	Kap Hoegh (East Greenland)	Greenland	3.06 ± 0.99 (20)	Fort et al. (2016)
	Little auk (*Alle alle*)	2010	Kap Hoegh (East Greenland)	Greenland	3.17 ± 0.82 (40)	Fort et al. (2016)
	Little auk (*Alle alle*)	2011	Kap Hoegh (East Greenland)	Greenland	3.21 ± 0.99 (25)	Fort et al. (2016)
	Little auk (*Alle alle*)	2012	Kap Hoegh (East Greenland)	Greenland	2.27 ± 0.41 (20)	Fort et al. (2016)
	Little auk (*Alle alle*)	2013	Kap Hoegh (East Greenland)	Greenland	2.60 ± 0.56 (19)	Fort et al. (2016)
	Little auk (*Alle alle*)	2014	Kap Hoegh (East Greenland)	Greenland	3.02 ± 1.14 (16)	Fort et al. (2016)
	Little auk (*Alle alle*)	2010–2011	Kap Hoegh (East Greenland)	Greenland	3.17 ± 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 μg/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 ± 0.21 μg/g dw and 0.70 ± 0.20 μg/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 ± 14.13 μg/g dw; Mallory et al., 2015). Brünnich's guillemots and northern fulmars had intermediate Hg concentrations (1.94 ± 0.63 and 2.71 ± 0.72 μg/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 μg/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 ± 0.41 to 3.99 ± 1.96 μg/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 μg/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 δ^{202}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 Δ^{199}Hg and Δ^{201}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 hotspots of Hg contamination, and to allow international actions (see the Minamata Convention, the Arctic Monitoring Assessment Program or the Global Mercury Assessment).

Acknowledgments

This study is part of several research programs supported by the French Polar Institute (IPEV - Pgr. 388 ADACLIM), the French Agency for National Research (ILETOP project ANR-16-CE34-0005, MAMBA project ANR-16-TERC-0004), the French Arctic Initiative - CNRS (PARCS project), and the Mission pour l'Interdisciplinarité - CNRS (Changements en Sibérie project). C.A is supported by a PhD fellowship from the French Ministry of higher education and research. The IUF (Institut Universitaire de France) is also acknowledged for its support to P.B. as a Senior Member.

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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