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A risk assessment review of mercury exposure in Arctic marine and terrestrial mammals

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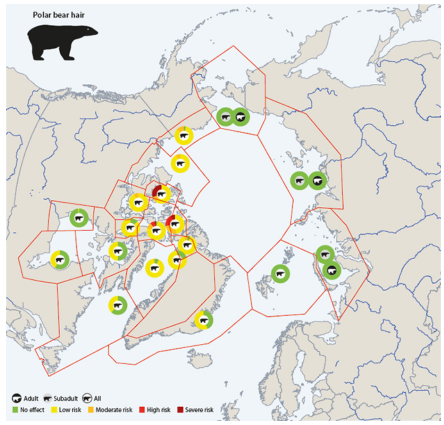
HIGHLIGHTS

- Most Arctic mammals are at low/no risk from mercury exposure.
- Terrestrial mammals are low and marine mammals high in mercury concentrations.
- Of 3500 marine mammal individuals, 6% are at high/severe risk from mercury.
- Knowledge gaps include improved effect thresholds and more recent data.
- High trophic biota hotspots in Canadian High Arctic seems linked to seawater MeHg.

GRAPHICAL ABSTRACT



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ABSTRACT

There has been a considerable number of reports on Hg concentrations in Arctic mammals since the last Arctic Monitoring and Assessment Programme (AMAP) effort to review biological effects of the exposure to mercury (Hg) in Arctic biota in 2010 and 2018. Here, we provide an update on the state of the knowledge of health risk associated with Hg concentrations in Arctic marine and terrestrial mammal species. Using available population-specific data post-2000, our ultimate goal is to provide an updated evidence-based estimate of the risk for adverse health effects from Hg exposure in Arctic mammal species at the individual and population level. Tissue residues of Hg in 13 species across the Arctic were classified into five risk categories (from No risk to Severe risk) based on critical tissue concentrations derived from experimental studies on harp seals and mink. Exposure to Hg lead to low or no risk for health effects in most populations of marine and terrestrial mammals, however, subpopulations of polar bears, pilot whales, narwhals, beluga and hooded seals are highly exposed in geographic hotspots raising concern for Hg-induced toxicological effects. About 6% of a total of 3500 individuals, across different marine mammal species, age groups and regions, are at high or severe risk of health effects from Hg exposure. The corresponding figure for the 12 terrestrial species, regions and age groups was as low as 0.3% of a total of 731 individuals analyzed for their Hg loads. Temporal analyses indicated that the proportion of polar bears at low or moderate risk has increased in East/West Greenland and Western Hudson Bay, respectively. However, there remain numerous knowledge gaps to improve risk assessments of Hg exposure in Arctic mammalian species, including the establishment of improved concentration thresholds and upscaling to the assessment of population-level effects.

1. Introduction

The circumpolar Arctic has been subject to minimal direct production, use and emission of industrial contaminants such as mercury (Hg). Hg is long-range transported to the Arctic by atmospheric and sea-currents (AMAP/UNEP, 2008, AMAP/UN Environment, 2019; AMAP, 2011). Methylmercury (MeHg) is readily bioavailable and biomagnifies in lipid-rich Arctic marine food webs, and thus raises heighted concern for the health of exposed wildlife and Indigenous human populations that largely depend on marine wildlife for their traditional diet (AMAP, 2011). Although Hg is a naturally occurring element, human activities and climate change have led to an average of 13.2-fold increase in Hg concentrations in the Arctic Inuit hair and selected Arctic marine mammal hard tissue samples. These include polar bear (*Ursus maritimus*) and ringed seal (*Pusa hispida*) hair, beluga (*Delphinapterus leucas*) and walrus (*Odobenus rosmarus*) teeth, and gyrfalcon (*Falco rusticulus*) and peregrine falcon (*Falco peregrinus*) feathers compared to preindustrial times (Dietz et al., 2009, 2011). As a result, an international treaty, UNEP's Minamata Convention on Mercury, was enacted in 2017 (Evers et al., 2016).

Here, we review the exposure risks of mercury (Hg) in Arctic biota as part of the AMAP (Arctic Monitoring and Assessment Programme) assessments on long-range transported contaminants in Arctic biota (AMAP, 2018, 2021; Dietz et al., 2013, 2019a, 2021a; AMAP, 1998, 2004, 2011, 2016; Letcher et al., 2010). It includes new information on the biological effects of Hg since 2018 and new data on Hg levels in Arctic mammals covering the period 2010 to 2020. Unlike the last AMAP assessment on combined effects of Hg and persistent organic pollutants (POPs), which provided a detailed literature review on adverse health effects across a range of physiological systems, this current study is more focused on predictive risk assessment. We address knowledge gaps identified in previous AMAP assessments, including sample size limitations and geographical data gaps in the Russian Arctic, to provide the most up-to-date risk assessment for health effects potentially resulting from Hg exposure in Arctic mammals, covering a plethora of species, tissues, and regions. The current work targets Arctic mammals only, whereas the assessments on seabirds and birds of prey and shorebirds are reported by Chastel et al. (2022/this issue) and results on fish are reported by Barst et al. (2022/this issue) as well as in a combined Assessment (AMAP, 2021/in press).

To our knowledge, there has been little effort to quantify population level effects of Hg exposure despite multiple health effects that have been reported in field studies of Arctic species (Dietz et al., 2019a; Routti et al., 2019). Establishing links between contaminant exposure and health outcomes is a difficult task (Rodríguez-Estival and Mateo, 2019). Such information is however extremely important to manage and conserve wildlife populations and provide evidence for regulation of contaminant emissions. It is critical to measure individual exposure impacts in order to estimate population-level effects using various modelling approaches, which consider physiological effects on reproduction, immune and endocrine functioning as well as energy demands (Svensson et al., 2011). This also requires a combination of controlled mechanistic studies (e.g. *in vitro* dose-response) and when possible in vivo studies on key species, as has been reported from hotspot areas like the Baltic primarily on POPs, which can be transferred to other pristine areas, including the Arctic. (Desforges et al., 2016, 2017, 2018a, 2018b, 2018c; Nyman et al., 2003; Routti et al., 2010). The approach taken here to address this issue is data-driven and combines toxicity data from relevant mammalian studies with an established risk modelling framework. We follow the methods of Ackerman et al. (2016) developed for North American birds, in which contaminant levels in tissues are used for Arctic species in a circumpolar risk analysis using critical body burdens and risk quotient analysis. In the present assessment, we have increased the liver data coverage from marine mammal species, regions and age groups from 70 to 112 groups (increase of 60%), increased the number of individuals from 2371 to 3772 (increase of 51%), and added polar bear hair data from 685 individuals from 22 regions and age groups compared to the Dietz et al. (2019a) risk analyses. The data from terrestrial mammals, although with much lower coverage, also increased from 8 to 16 species (100% increase), regions and age groups and the number of individuals increased from 211 to 814 (increase of 386%). We combined species and areas at risk to the observed temporal trends, which is reported in more detail for all available Arctic data series by Morris et al. (2022/this issue). Finally, we created heat maps from a generalized additive model (GAM) approach to examine the linkage between seawater MeHg concentrations in the upper 400 m water column with tissue concentrations of ringed seal (*Pusa hispida*) and polar bear liver and hair concentrations (see details in Section 2.4. Hotspot area detections for details on method and references).

2. Material and methods

2.1. Literature and data search

We did a search on PubMed, ScienceDirect, Google, Google Scholar, EBSCO, ProQuest, ScienceDirect, MEDLINE and grey literature combing the terms “Arctic” and/or “mercury” and/or “Hg” and/or “effects” and/or “marine mammals” and/or “terrestrial mammals” by ultimo 2020. In addition, Hg data from work in preparation from the appointed Key National Experts of the eight Arctic countries as well as from other scientific groups working in the Arctic was included. Only Hg data from accredited laboratories participating in the international QA/QC programs were used. All laboratories involved in the general AMAP monitoring participate in inter-calibration exercises and QA in the Quasimeme Program (Europe) and the Northern Contaminants Program (North America) and several of these are also accredited ISO 9000 laboratories. The study design is based on a review of the existing literature for post-2000 articles as well as unpublished data of Hg exposure in marine and terrestrial mammals from the Arctic and, where possible, the raw data were extracted. In addition, Hg data from Baltic marine mammals were obtained from the projects BONUS BALTHEALTH for comparative purposes.

2.2. Risk analysis

The risk analysis in the present assessment for potential Hg-associated health effects was based on no risk (NRC), low risk (LRC), moderate risk (MRC), high risk (HRC) and severe risk (SRC) categories (Table 1). These risk category thresholds reflect effects on reproduction and adverse effects on body condition and behavior. For marine mammals, the hepatic Hg threshold values were estimated using data from Ronald et al. (1977), Dietz et al. (2019a, 2021a) and AMAP (in press). As in Ronald et al. (1977), based on the measured liver THg concentrations in harp seal (*Pagophilus groenlandicus*) exposed to methylmercuric chloride, we assigned 5 risk categories where no risk refers to average concentrations in the control group (Dietz et al., 2021b,a); AMAP, in press). For terrestrial mammals, hepatic Hg threshold values were taken from studies in mink (*Mustela vison*) (Wobeser et al., 1976; Wren et al., 1987). To estimate health risks using Hg data in hair, it was necessary to convert liver threshold values to hair equivalents. This conversion was based on significant linear regression analyses ($p < 0.01$; $R^2 = 0.2824$; SE = 0.0592; $n = 174$) between East Greenland polar bear livers (Hg_H) and hair (Hg_L) according to AMAP (2022/in press).

$$\ln(Hg_H) = (\ln(Hg_L) - 1.18748)/0.79941$$

All liver data reported in the present assessment is provided on wet weight (ww) basis, whereas the hair concentrations are on dry weight (dw) basis.

2.3. Time trend analyses

For species and regions where Hg concentrations of concern fell undo the HRC and SRC (Table 1), time trend analysis was used to identify increasing or decreasing trends. The temporal trend analysis followed the methods of AMAP (in press) and Morris et al. (2022/this issue). In brief, the biota time series of Hg concentrations were assessed as changes in the log-transformed concentrations over time using linear mixed models. The type of temporal change considered was dependent on the number of years of data; for time-series including seven or more years of data the non-linearity of the trend was evaluated by use of smoothers (Fryer and Nicholson, 1999). For additional temporal trend data please see Morris et al. (2022/this issue).

2.4. Hotspot area detections

Since the last assessment, the Arctic Ocean has become one of the best observed oceans (Dastoor et al., in review). The new seawater Hg species observations cover the Irminger and Labrador Sea (Cossa et al., 2017, 2018), the Canadian Arctic Archipelago (Wang et al., 2018), the East Siberian Sea (Kim et al., 2020), the East Greenland Shelf, the Fram Strait and the Barents Sea (Petrova et al., 2020b) and the central Arctic Ocean (Agather et al., 2019, Charette et al., 2020, Heimbürger et al., 2015, Tesán Onrubia

Table 1
Estimated risk (i.e., Risk Categories, RC) to total mercury (THg) exposure on the health effects in marine and terrestrial mammals. WW: wet weight, DW: dry weight.

Species	Matrix	No risk NRC	Low risk LRC	Moderate risk MRC	High risk HRC	Severe risk SRC	Reference
Marine mammals	Liver (μg/g WW)	<16.0	16.0–64.0	64.0–83.0	83.0–123.0	≥123.0	Ronald et al., 1977
	Hair (μg/g DW)	<6.1	6.1–24.4	24.4–31.7	31.7–48.1	≥48.1	Risk intervals established from polar bear liver to hair correlation, this study
Terrestrial mammals	Liver (μg/g WW)	<4.2	4.2–7.3	7.3–22.7	22.7–30.5	≥30.5	Wobeser et al., 1976; Wren et al., 1987

et al., 2020). We combined MeHg seawater measurements from both the Arctic GEOTRACES programme and newly collected data from "Arven etter Nansen Seasonal Cruise Q1 2021" (Kohler et al. in prep) to produce heat maps from a GAM model. These maps cover the upper 400 m of the water column from the Barents Sea to the Canadian Archipelago, a region accessible to the seals and where the highest MeHg concentrations are usually detected in the Arctic Ocean (see Section 7). Comparable heat maps were produced for ringed seal liver and polar bear liver and hair (Rigét et al., 2005; Routti et al., 2011) to illustrate the patterns with the MeHg hotspot areas of the upper 400 m of the water column. The predicted GAM heat MeHg concentrations based on 68 water column station measurements were correlated with the biota concentrations for the respective sampling areas to estimate the linkage between the two variables.

3. Marine mammals

3.1. Risk effects extrapolated from liver Hg concentrations in marine mammals

Overall, 30% of individuals from the 29 species, regions and age groups were within the two highest risk categories, of which 18 of the 112 presented region/age/sex-groups) had 19% individuals in the SRC (>126 μg/g ww) and an additional 11 groups (12% of the individuals) in the HRC (83–126 μg/g ww (Fig. S1; Table S1). Individuals from the 29 species, regions and age groups from the two highest risk categories, however, accounted only for 200 individuals representing only 5.8% of a total of 3445 individuals analyzed for their Hg loads. As for the SRC, this accounted for approximately 102 individuals (3.0%) and the HRC accounted for the remaining 98 individuals (2.8%) (Fig. S1, Table S1). The highest exposed animal groups (evaluated by percentages in the SRC) are in the following decreasing order: 1) adult hooded seals (*Cystophora cristata*) from the Denmark Strait (57%), 2) adult male hooded seals from Greenland Sea/Denmark Strait (45%), 3) adult polar bears from the Northern Beaufort Sea (41%), 4) juvenile polar bears from Qaanaaq North West (NW) Greenland (33%) (Fig. 1), 5) adult killer whales (*Orcinus orca*) from East (E.) Greenland, Iceland and Faroe Islands (33%), 6) adult long-finned pilot whales from the Faroe Islands (27%), 7) juvenile polar bears from Lancaster/Jones Sound (20%), 8) subadult long-finned pilot whale from the Faroe Islands (20%), 9) adult female ringed seals from Sachs Harbor (12%; Fig. 2) and 10) adult male ringed seals from Sachs Harbor (7.8%). Surprisingly, the hooded seals are the highest exposed group, however,

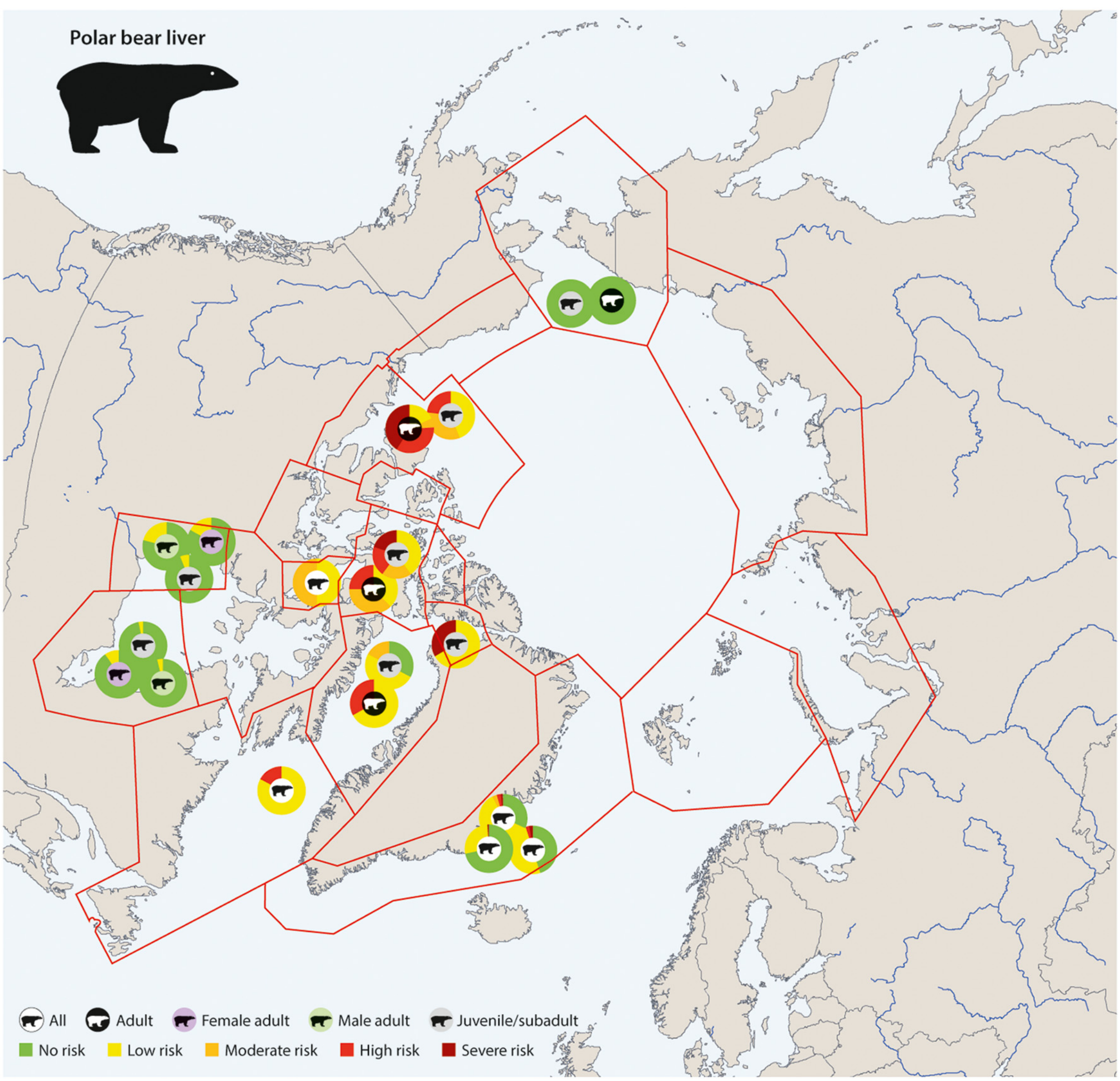

Fig. 1. The risk of Hg-mediated health effects in polar bear subpopulations and, based on post-2000 liver Hg concentration. The five risk categories are defined using effect threshold categories observed for harp seals (Ronald et al., 1977). SI Table 1 presents the detailed information as well as references upon which this summary graphic is based and SI Fig. 1 presents ranked histograms of bears together with the other marine mammals.

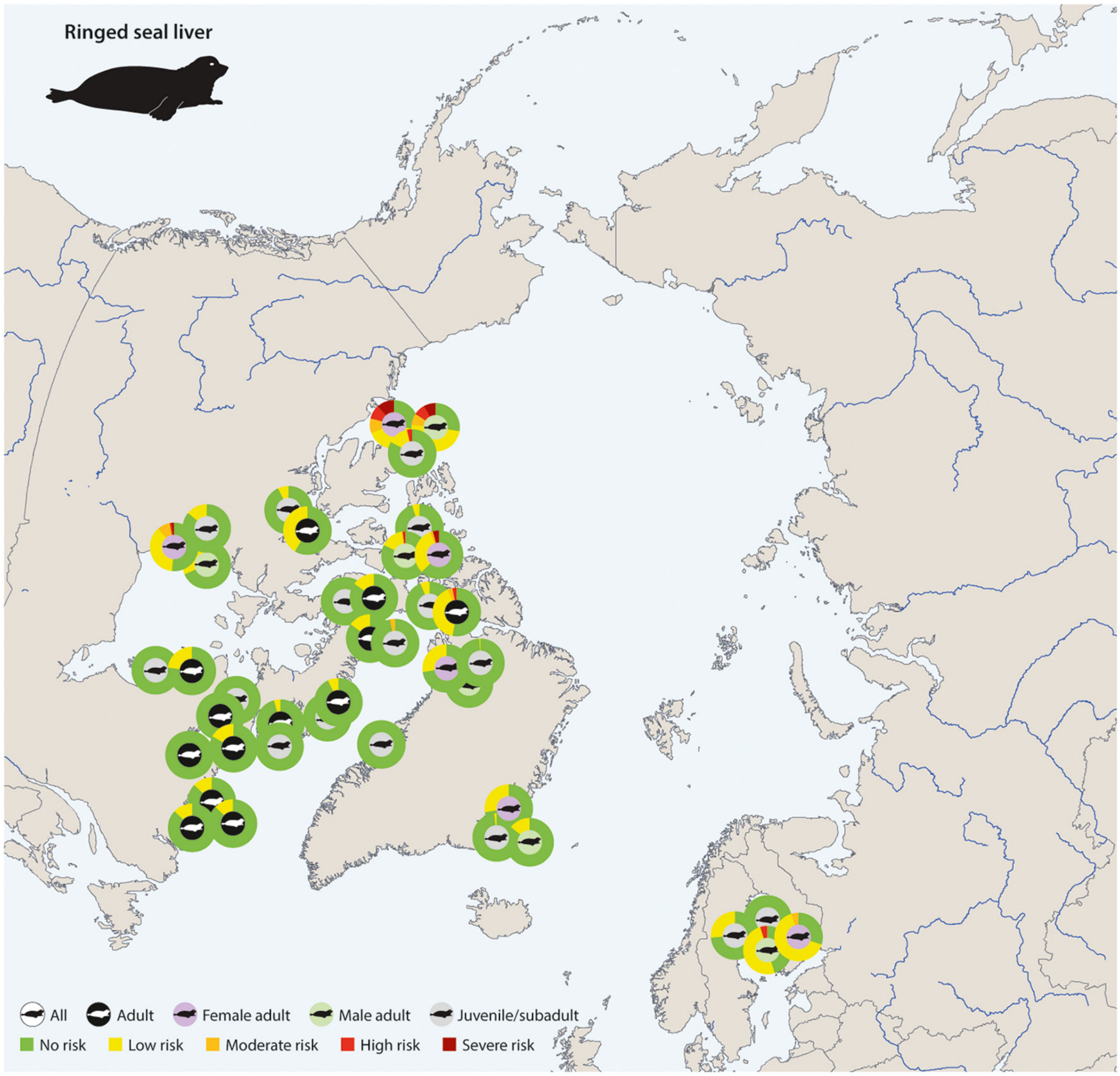

Fig. 2. The risk of Hg-mediated health effects in Arctic ringed seal subpopulations based on post-2000 monitoring data. The five risk categories are defined using effect threshold categories observed for harp seals (Ronald et al., 1977). Table S1 presents the detailed information upon which this summary graphic is based and Fig. S1 and S2 presents ranked histograms of the ringed seals together with the other marine mammals.

hooded seals have previously shown high Hg concentrations (Nielsen and Dietz, 1990; AMAP, 1998) due to their consumption of redfish. As for top predators such as polar bears from hot spot areas in northern Canada and Northwest Greenland (see Supplementary Information), previous studies report similar high concentrations in these areas (Braune et al., 1991; Dietz et al., 1998, 2000; Routti et al., 2011; Wang et al., 2018). The high concentrations of Hg in toothed whales such as killer whales, narwhals and pilot whales from the North Atlantic, is due to their lack of ability to excrete these substances as well as their high trophic position in the food chain (Sonne et al., 2010a, 2013, 2018a, 2018b; Dietz et al., 2019b).

The least exposed animal groups (in liver and μg/g on a wet weight basis; Fig. S1, Table S1) in the Arctic regions (i.e. groups with all 100% of the individuals in the NRC are in the following increasing order: 1) yearling harp seals (*Pagophilus groenlandicus*) from the Greenland Sea (in ww 0.17 μg/g), 2) foetus killer whales from E. Greenland, Iceland and Faroe Islands (0.18 μg/g), 3) subadult harbor porpoises from the Barents Sea (0.49 μg/g), 4) adult male harbor porpoises from the Barents Sea (0.58 μg/g), 5) subadult harbor porpoises from the Norwegian Coast (0.69 μg/g), 6) subadult harp seals from the Greenland Sea/Denmark Strait (0.69 μg/g), 7) adult female harp seals from the Greenland Sea/Denmark Strait (0.76 μg/g), 8) adult harp seals from Ittoqqortoormiit (0.78 μg/g), 9) juvenile ringed seals from Qeqertarsuaq (0.92 μg/g); 10) juvenile ringed seals from Kangiqsujuaq (0.92 μg/g). Additional information from the other risk categories Fig. S1 and Table S1. In the present review regions outside the Arctic (North Atlantic, North Sea, Inner Danish Waters and Baltic) were include for comparative purpose for ringed seal and harbor porpoise (*Phocoena phocoena*).

There exists sufficient liver data for polar bears and ringed seals to provide an overview of the regional differences and consistencies in their Hg exposure and the related risks. Six of the 20 polar bear groups (regions, age and sex) from which we presented data had concentrations in the SRC. Of these adult polar bears from the Northern Beaufort Sea were the highest exposed group with 41% in the SRC (Fig. 1; Table S1; Fig. S1). Juvenile bears from Qaanaaq and juvenile bears from Lancaster Sound had 33 and 20%, respectively, in the SRC although these sample sizes were small (n = 6 and 5, respectively). All three exposure risk groups in Ittoqqortoormiit had between 1.0 and 2.5% percent of the populations in the SRC based on sample sizes from 40 to 96 individuals.

In four out of six groups, individuals were also in the HRC ranging from 36 to 2.5%. As for groups with the highest exposed individuals in the HRC, from 33 to 17% had individuals in this category. This included four groups, including adult bears from Baffin Bay and Lancaster Sound as well as juveniles from Northern Beaufort Sea and all age groups from the Davis Strait. Only two groups had individuals in the MRC, and with the highest exposed groups being for the overall population from Gulf of Boothia and juveniles from Baffin Bay. In the remaining eight groups, all individuals were in the two lowest risk categories, of which 100% of the juvenile and adult groups from the Chuckhi Sea were in the NEC. Information on polar bears from the Russian Arctic is lacking.

Four out of the 41 presented ringed seals region, age and sex groups had individuals in the SRC. Adult female and male ringed seals from Sachs Harbor were the highest exposed group with 11.5 and 7.8% of the individuals in the SRC, respectively (Fig. 2; Table S1; Fig. S1). Adult female ringed seals from Resolute and Arviat W. Hudson Bay were the other two groups with 3.9 and 1.4% of the individuals in the SRC respectively. Most og these four groups also had individuals in the HRC and MRC with 9.6 to 1.4% and 9.6 to 2.0% respectively. In addition, three groups including subadult ringed seals from Sachs harbor, adults from Grise Fjord, and adult males from Resolute had 3.4 to 2.1% of the individuals in the HRC. Additionally two groups, juvenile ringed seals from Pond Inlet and adult males from Arviat, had 3.4 and 2.3% of the individuals in the MRC respectively, and five out of seven of the two highest risk categories also had 9.6 to 2.0% of the individuals in the MRC. For 28 groups of 37 groups of Arctic ringed seals, all individuals ranged in the two lowest risk categories. As for the polar bears livers, information on ringed seals from the Russian Arctic is lacking. For comparative purpose, data from Baltic ringed seals are included in Fig. 2 (see Fig. S1 and Table S1).

3.2. Risk effects extrapolated from hair Hg concentrations in polar bears

Hair samples are often used to evaluate human exposure and risk to Hg (Wang et al., 2021; Petrova et al., 2020a). Similarly, here we conducted an assessment of the regional risk of polar bears based on polar bear hair samples collected from hunting as well as hair sampled from polar bears that were routinely collected during tagging studies. We use hair samples collected before 2000 to obtain a better spatial

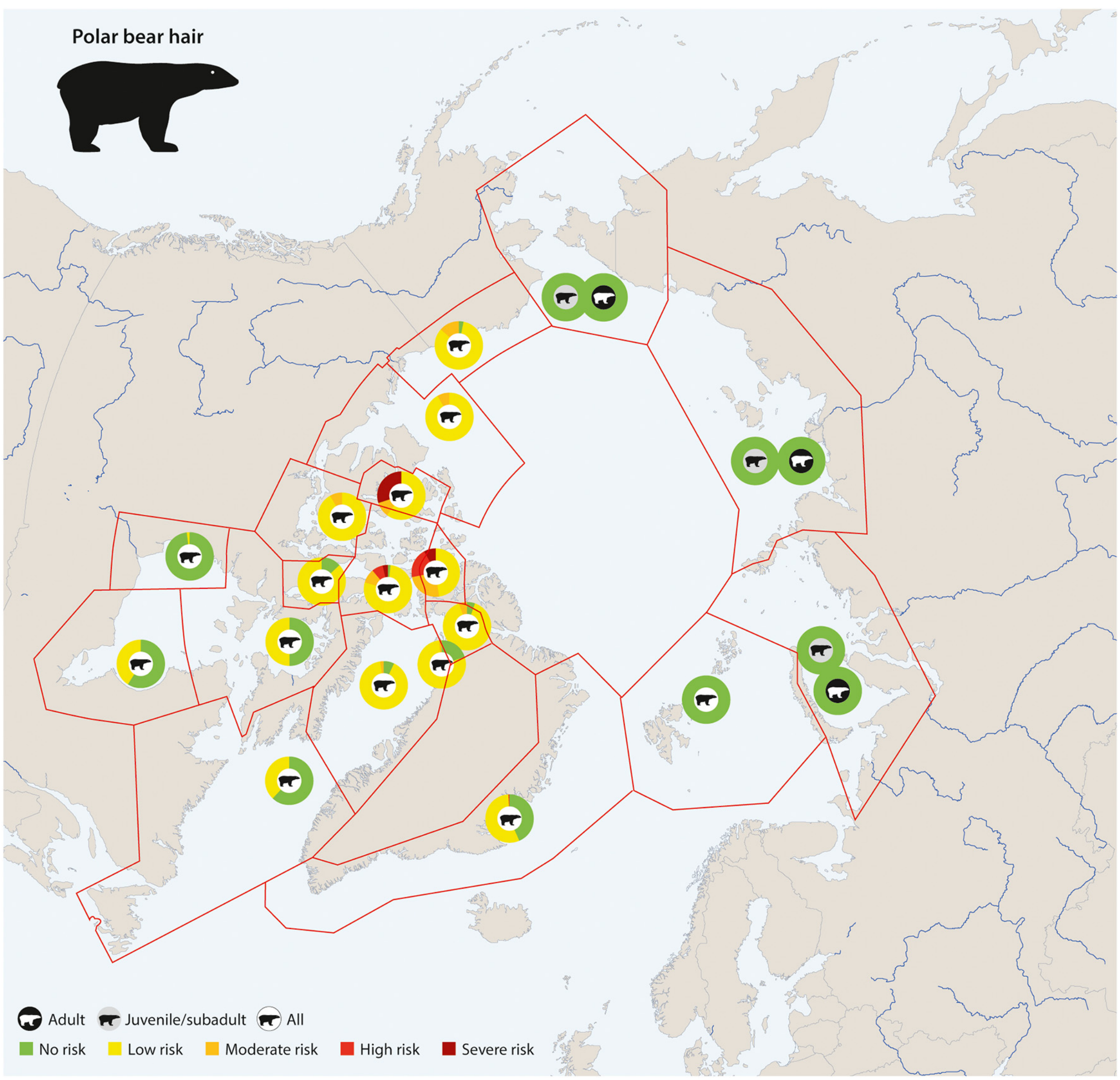

Fig. 3. The risk of Hg-mediated health effects in polar bear subpopulations based on pre- and post-2000 monitoring hair data. The five risk categories are defined using effect threshold categories observed for harp seals (Ronald et al., 1977) converted into hair Hg concentrations by the East Greenland correlation between these two matrices. See Table S2 for the detailed information upon which this summary graphic is based.

coverage. The analyses revealed that some bears from three populations in the central and northeastern Canadian Arctic, had concentrations in the SRC, namely Viscount Melville Sound (30%), Norwegian Bay (8.0%), and Lancaster Sound (3.6%) (Fig. 3; Fig. S2; Table S2). The corresponding percentages of bears from these three populations in the HRC were 0.0, 20 and 7.4%, respectively, and only a low percentage (0.7%) of bears from East Greenland had individuals in the HRC. As eight of the Canadian populations were sampled before year 2000, the present day risk patterns are uncertain. Unfortunately, the few time series from Morris et al. (2022/this issue), of which none is based on hair, allow us to update the data obtained from samples collected prior to 2000 Overall, the combined risk assessment using hair and liver Hg concentrations raises concern for polar bears in the Canadian High Arctic andg Northwestern Greenland. In contrast, hair Hg concentrations in Barents, Kara, Laptev and Chukchi Sea all fell within the NEC, which indicated that there are no Hg effect implications for polar bears in the regions of Svalbard and northern Russia (Lippold et al., 2020; resubmitted).

4. Terrestrial mammals

The majority of Hg concentrations in terrestrial mammals fell within the two lowest risk categories for Hg-mediated health effects (NRC and LRC, see Fig. 3). Individuals from the 12 species, regions and age groups from the two highest risk categories, however, accounted only for 2 individuals representing only 0.3% of a total of 731 individuals analyzed for their Hg loads. Arctic fox (*Vulpes lagopus*) from Iceland however, had 9.0% of the adult population being at HRC, 35% in the MRC, 22% in the LRC and 35% in the NEC. Juvenile Arctic foxes were, however, exposed to lower levels, as the majority of the foxes (67%) in this age group fell in the NEC. Juvenile Arctic foxes from Arviat and Svalbard had 98% and 100% in the NRC respectively, which raises the question on whether some local Hg sources are present on Iceland to cause elevated risk for the species there (Fig. 4 and Table S3). For sheep (*Ovis aries*) on the Faroe Islands, as much as 15% were found in the MRC, which is higher than expected and could be attributed to agricultural fertilization by fish remains or eutrophication by bird droppings (from the extensive seabird colonies on the islands) as suggested by AMAP (2018) and Dietz et al. (2019b). The remaining 85% of the sheep fell in the NRC. All (100%) seven Caribou/reindeer (*Rangifer tarandus*) populations and age groups were in the NRC with median Hg concentrations ranging from 0.12 to 1.24 μg/g ww (Fig. 4, Table S3). It should be noted that the risk categories are based on liver Hg threshold values from studies in mink, which is a carnivore (Wobeser et al., 1976; Wren et al., 1987; Dietz et al., 2019b; AMAP, in press). Sheep and caribou/reindeer are herbivores/ungulates, and it is possible that the threshold level of Hg may differ from that used for the carnivores herein.

5. Temporal trends and risk for the highest Hg exposed species and regions

5.1. Polar bears

Twenty region, age and sex groups from 10 polar bear management areas were assessed for trends in Hg risk based on their long-term liver concentration data (Figs. 1 and 3; Fig. S1; Table S1). Four polar bear management areas (Western Hudson Bay, Ittoqqortoormiit and Svalbard) and four out of 13 analyzed groups showed significant increasing Hg temporal trends, but no groups showed declining Hg loads (Fig. 5). For the regions with the highest Hg risks, namely Lancaster Sound/Jones Sound (SRC: 0–25%; HRC: 20–25%) as well as Northern Beaufort Sea (SRC: 0–41%; HRC: 22–35%) and Qaanaaq region (SRC: 33%), unfortunately no time trend information was available. However, yearly significant Hg increases of 1.6–1.7% ($p < 0.0001$) per year from 1892 to 2008 have previously been reported for polar bear hair from the Qaanaaq, NW Greenland (Fig. 5; Dietz et al., 2011). In Western Hudson Bay a significant increasing trend of 6.0% per year was likewise detected in the liver of adult males, whereas no trend was observed in juveniles and or adult females. Despite lower Hg concentrations in polar bears from Ittoqqortoormiit, CE Greenland (SRC: 1.0–33%; HRC: 3.0%), significant temporal increases in hair Hg levels for juveniles and adults suggest that a larger percentage of the population is likely to appear in the higher risk categories in the future. The hair risk analyses for polar bears were quite similar to the liver risk results (Figs. 1 and 3; Tables S1 and S2) as both Lancaster Sound, Norwegian Bay, Viscount Melville Sound, Northwest Greenland and East Greenland had animals in the SRC and in the HRC (SRC: 4.0–33%; HRC: 1.0–20%).

5.2. Ringed seals

Thirty-seven groups from 18 Arctic regions were assessed for trends in Hg risk based on their long-term liver concentration data (Fig. 2; Fig. S1; Table S1). Only one of seven ringed seal regions (Labrador Sea) and one out of 22 analyzed groups (4.5%) showed significant increasing temporal trends of THg (Morris et al. (2022/this issue)). However, four (Eastern Beaufort Sea, Lancaster Sound (Resolute Passage), Labrador Sea and Western Hudson Bay) out of the seven ringed seal regions and six out of 22 analyzed groups (27%) showed significant annual declining Hg trends (−2.4% to −8.0%) in muscle tissue, which was

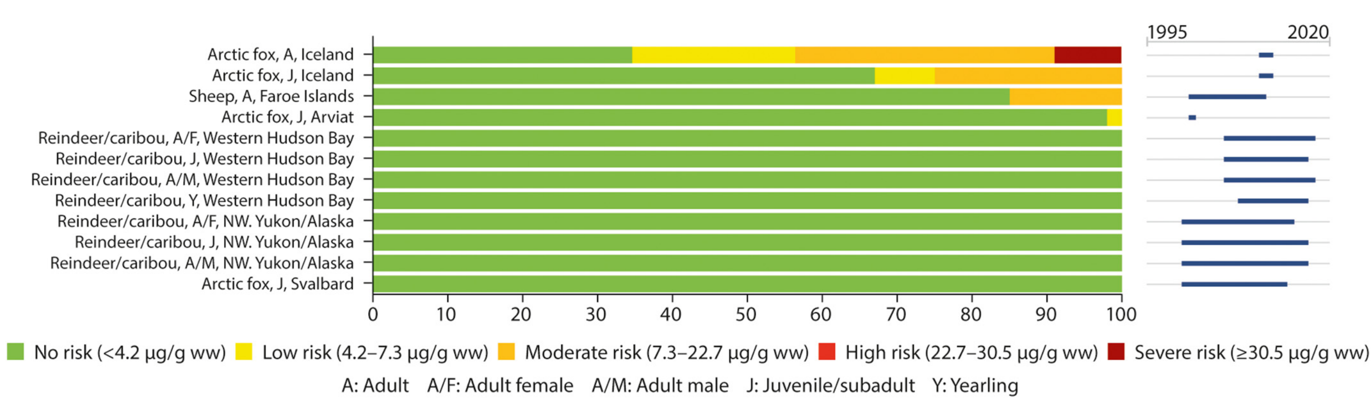

Fig. 4. The proportion of individuals ranked from highest to lowest of specific Arctic terrestrial mammal populations according to the risk of total Hg-mediated health effects. Based on liver Hg concentrations from 2000 to 2015, assignments were made to five risk categories and based upon effect threshold categories observed for mink (Wobeser et al., 1976; Wren et al., 1987). See Table S3 for the detailed information upon which this summary graphic is based.

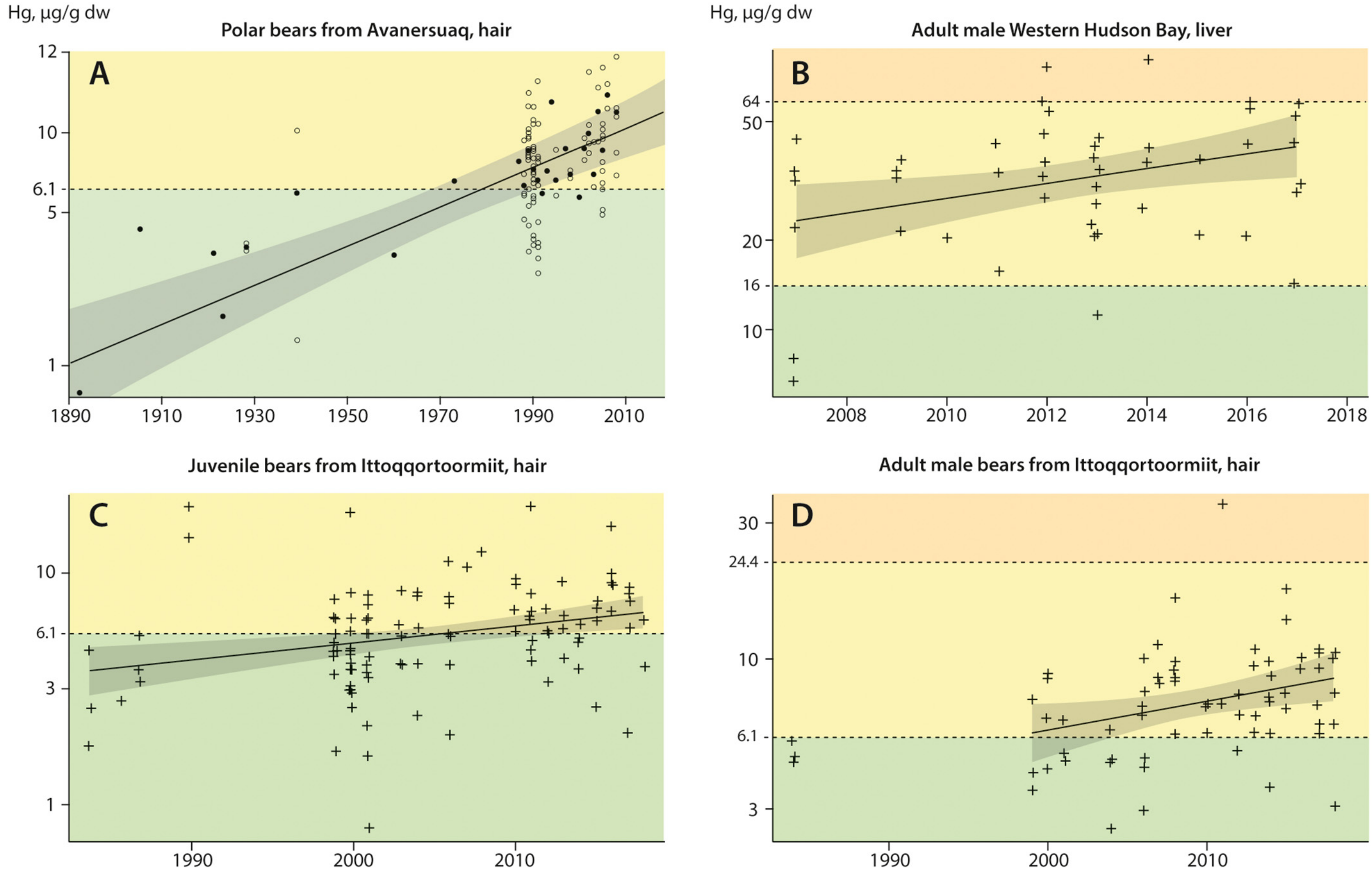

Fig. 5. Examples of significant Hg increases in polar bear hair and liver from Canadian and Greenlandic waters plotted on top of risk intervals (No risk (green), Low risk (yellow). Moderate risk (orange), High risk (red) and severe risk (dark red)) defined in Table 1. Panel A modified from Dietz et al. (2011) and panels BD from Morris et al., (2022/this issue).

documented to have a higher power than liver (Morris et al., 2022/this issue). Only adult seals (males and females) from the Labrador Sea showed significant increases in their livers. Overall, we conclude that ringed seals are not at significant risk with respect to increases in Hg exposure over time.

5.3. Belugas

Of the 10 recent (1999-present) temporal trend analyses conducted for epidermis, muscle and liver tissues of belugas from Southern Beaufort Sea and Southern Hudson Bay, three significant declines (30%; −2.5% to −8.6%) were detected and no significant increases. With the rather low percentages of belugas having individuals in the SRC and HRC it is encouraging that a large proportion of these whales are showing significant declines in their Hg loads.

5.4. Pilot whales

One (33%; 1.7%; muscle of juvenile males) out of the three pilot whale groups showed significant increasing trends and none showed significant declines (Morris et al., 2022/this issue). With the high percentage of pilot whales having individuals in the SRC, it is the juvenile pilot whales that showed significant increases in muscle Hg. With respect to human health exposure, it is encouraging that there is a decline in the human consumption of pilot whales (i.e. the meat) at the Faroe Islands due to the health advice from the Faroe Health Authorities (AMAP, 2015).

5.5. Other marine mammals

As no time trend analyses were available from other marine mammal groups, no overall evaluation can be made with respect to temporal patterns of these risks.

5.6. Terrestrial mammals

Except from one group of adult Arctic fox from Iceland, none of the terrestrial mammals had individuals in the SRC and HRC. Of the two caribou population time trends from Canada and one arctic fox from Svalbard no significant trends in Hg were detected, thus risk is likely to remain low for these animals in the near future.

6. Estimating population effects from mercury loads in highly exposed wildlife

It is challenging to assess the impact of Hg exposure and accumulation at the population-level for any species, however, especially so for species living in remote areas like the Arctic. These assessments require long term population monitoring on tissue Hg levels and relevant long-term individual fitness and population metrics such as adult survival, reproductive success and recruitment (i.e. offspring survival to reproductive age), and ultimately population growth rates. Actually the toxicity data derived from harp seals (Ronald et al., 1977) used to generate the risk categories used in our study documented the death of two harp seals out of six, exposed to 0.25 mg methylmercuric chloride/kg body weight/day. The two seals died on day 20 and 26 of exposure and blood parameters indicated toxic hepatitis, uremia and renal failure (Ronald et al., 1977). As the experiment was run for less than 90 days and mainly on immature animals no information on reproduction effects was generated. Also, there is a severe lack recent effect experiments on several Arctic marine mammal species relevant for the present assessment. We are confident that a permission such an experiment similar to the harp seal study would not be granted by Animal Care Committees today for seals, whales or polar bears and hence there are no data on effect concentrations targeted specifically these and other high trophic predatory marine mammals. The lack of effect studies has previously been concluded and further investigations recommended (e.g. AMAP/UN Environment, 2019, Dietz et al., 2019b, 2020). The closest recent example was POP effect studies using minke whale (*Balaenoptera acutorostrata*) blubber to expose sledge dogs (*Canis familiaris*) and Arctic fox (*Alopex lagopus*) as sentinels for the polar bear, which are better that seal data, but unfortunately this was never carried out with a focus on Hg (e.g. Bradley et al., 2018; Kirkegaard et al., 2010a, 2010b, 2011; Pedersen et al., 2015; Rogstad et al., 2017; Sonne, 2010; Sonne et al., 2005, 2006a, 2006b, 2006c, 2007a, 2007b, 2007c, 2008a, 2008b, 2008c, 2008d, 2008e, 2009a, 2009b, 2010b, 2014a, 2014b, 2017; Verreault et al., 2008). Other examples effect evaluation is the use of Risk Quotients comparing body burden and critical body residues estimated from PBPK modelling from rats and mice relative to immunologic, reproductive and carcinogenic effects, but such studies does not provide a better and more relevant picture relative to population effect of Arctic species in focus of this assessment (Sonne et al., 2009c, 2015, 2016; Dietz et al., 2015, 2018a). Observed effects in the HRC included potential for organ lesions (kidney, liver), anorexia, and reduced growth, while effects in the SRC included severe impacts on organ function (i.e. kidney failure), weight loss, and ultimately increased mortality. For toxicity in mink (i.e. terrestrial mammal assessment), observed effects in the HRC included reduced litter size and offspring growth rate, and SRC included brain lesions, reduced growth, anorexia, and increased adult mortality. Given the direct consequences on reproduction and survival for the HRC and SRC, two endpoints of key concern for potential population impacts, concern is warranted for select populations of hooded seals, killer whales, and pilot whales, as well as for Lancaster Sound and Northern Beaufort Sea polar bears. Here, >20% of sampled individuals within these populations (and up to 60–90%) had concerning tissue Hg levels; impacts in such a large proportion of the population has the potential to have a meaningful affect demographic rates and overall population fitness. For polar bears in highly exposed regions such as the Lancaster Sound and Jones Sound, the population size trends are data deficient (Vongraven and York, 2014; Dietz et al., 2015), making it difficult to track potential effects of Hg exposure. From areas like the S. Beaufort Sea and the Baffin Bay, populations of marine wildlife are declining, but the effects of climate change are likely to play a major role in these areas (Vongraven and York, 2014; Dietz et al., 2015). If the mink toxicity data translates to Arctic foxes, our assessment suggests that a large portion of the Arctic fox population in Iceland is at high risk for potential population relevant impacts from Hg exposure.

It is important to note that the above population assessment is based on toxicity data from only one relevant species for each group (e.g. harp seal and mink). Care must be taken to extrapolate effects across species because of potential inter-species differences in Hg toxicokinetics (e.g. uptake and distribution) and toxicodynamics (e.g. species sensitivity to effects). Because such differences, including differences between carnivores and herbivores, are unknown at this time and difficult to assess, the current risk exercise provides the best available evidence-based assessment of potential impacts across Arctic species. Furthermore, Arctic species may also be impacted at the population-level through similar effects on reproduction and adult survival due to exposure to other contaminants (POPs including PFASs) and stress related to climate change and hunting (e.g. Laidre et al., 2015; Dietz et al., 2015, 2019a). Teasing out the effects of Hg from other concurrent stressors remains a challenge, though it is expected that these stressors act in concert to increase the overall stress of individuals and populations. Overall, more work on exact risk benchmark values for different species and regions is recommended as well as population effect studies in relation to Hg and other contaminant loads as conducted for killer whales by Desforges et al. (2018c).

7. Geographical hotspot areas in water and wildlife

Previous publications have considered geographical hotspots with respect to Hg biomagnification and adverse biological effects in mammals such as ringed seals and polar bears (e.g. AMAP, 1998; Brown et al., 2016; Dietz et al., 1998, 2000, 2013, Rigét et al., 2005; Routti et al., 2011, 2012). Most of these surveys have shown that the Canadian Arctic Archipelago and northwestern Greenland are hotspots for Hg exposure in biota. Heimbürger et al. (2015) put the idea forth that the shallow seawater MeHg maximum in the Arctic Ocean, typically found near 200 m depth (vs other oceans 500–1000 m, Bowman et al., 2020), may be responsible for the high Hg levels of arctic biota. Wang et al. (2018) recently reported high-resolution vertical profiles of total Hg and MeHg in seawater during GEOTRACES ship transect surveys conducted in 2015 from the Labrador Sea and across Baffin Bay to the Canadian Arctic Archipelago and Canada Basin. They showed the highest Hg concentrations in the Beaufort with a distinctive subsurface maximum of MeHg in seawater in the upper 400 m depths peak concentration decrease from Canada Basin eastwards (Fig. 6). It was hypothesized that Hg concentration in seawater was linked to Hg exposure in biota. The reason for focusing on MeHg is that this fraction of the food is more readily taken up by biota (ca. 95%) whereas the corresponding proportion for inorganic Hg is thought to be lower than 15% (Berlin, 1986; WHO, 1993; Mori et al., 2012; Dietz et al., 2013). In addition MeHg, not inorganic Hg, is the main fraction of which accumulates and biomagnifies in marine biota (AMAP, 2011). Therefore, we produced a heat map from a generalized additive model (GAM) approach covering the upper 400 m of the water column for available seawater with the 2016 GEOTRACES GRIFF transect data obtained between Northeast Greenland and Svalbard (Petrova et al., 2020b). Similar heat maps were produced for ringed seals liver and polar bear liver and hair based on data from Rigét et al. (2005), Routti et al. (2011) and Dietz et al. (this study) to illustrate the relative geographical high trophic biota hotspots as well as. Hotspot areas in the water column and biota was detected in the Beaufort Sea and central Canadian Arctic Archipelago In addition high concentrations in Northwest Greenland was detected for biota but not in the sea water and the high concentration in sea water in Northeast Greenland was not reflected in biota. The ringed seal and polar bear concentrations along Svalbard are somehow lower in concordance with lower concentrations of MeHg in the water columns towards Svalbard compared to Northeast Greenland. The predicted GAM heat MeHg concentrations based on 68 water station column measurements showed as expected only limited significant correlations and only for the ringed seal ($p = 0.024$; $r^2 = 0.66750$; $f = 10.037$; $n = 11$) liver Hg concentrations, whereas the correlation with the polar bear hair Hg concentrations ($p = 0.132$; $r^2 = 0.23288$; $f = 2.732$; $n = 16$) and polar bear liver Hg concentrations ($p = 0.142$; $r^2 = 0.37793$; $f = 3.038$; $n = 10$) were not significant. To further elucidate these relations more modelling work from larger parts of the Arctic with more transects, more species, corresponding years and seasons, as well as adjustment for age, feeding proxy adjustments and the right tissues. Here muscle tissue would during summer time would be preferable as the ship based transects are carried out during open water seasons. It may however be complicated to get such samples as Polar bears quotas following the calendar year has been obtain before the summer and ringed seal are seldom hunted during the open water season from June to August as the seals are thinner and tend to sink if hunted Seabirds were included in a similar modelling exercise in the AMAP Mercury Assessment (2022/in press) and showed corresponding hotspots in the Canadian High Arctic. Further details on the MeHg entrance into the food chain and trophic transport between phytoplankton, zooplankton and higher trophic levels is provided by Wang et al. (2018). In addition, Wang et al. (2018) concludes that detailed investigations will be required

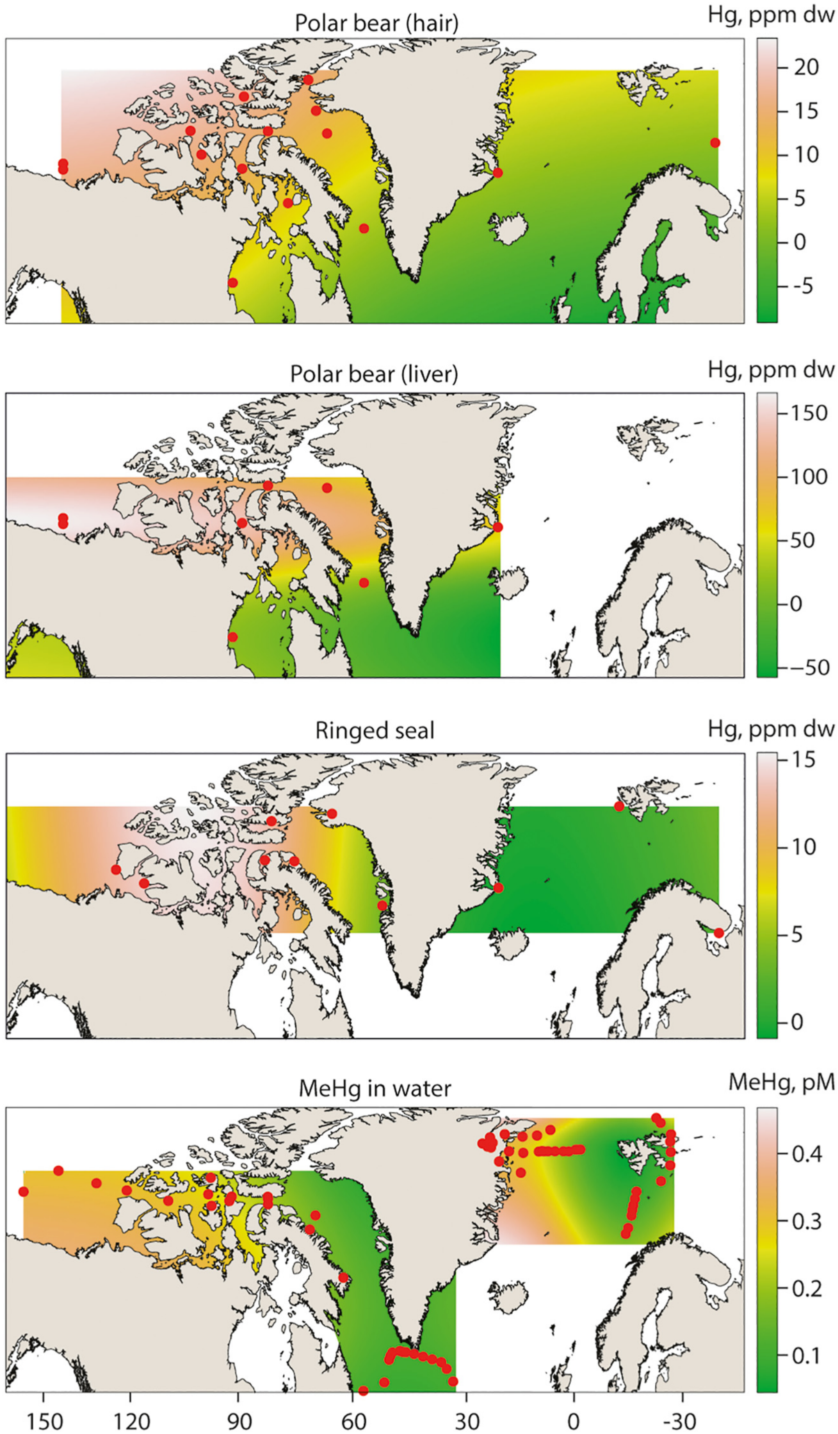

Fig. 6. Heat maps based on a generalized additive model (GAM) of geographical patterns of mercury. From the bottom: MeHg in the upper 400 m of the ocean, mercury in juvenile ringed seal liver, mercury in polar bear liver and polar bear hair (Cossa et al., 2018; Petrova et al., 2020b; Rigét et al., 2005; Routti et al., 2011; Wang et al., 2018; Dietz et al., this article).

to identify processes controlling the production and loss of MeHg associated with the upper halocline waters of the western Arctic Ocean as well as how these processes respond to the changing climate. As these processes are not known in detail it is also a challenge to understand to what extent the geographical differences are driven by anthropogenic or natural processes. It is hence at present uncertain whether the hotpot areas are linked to natural or anthropogenic processes and these preliminary comparisons needs further work.

8. Conclusions

In general, based on the most recently published information, most marine and terrestrial mammal species are at low risk of health effects from Hg exposure. Nonetheless, Hg continues to pose a justifiable concern for some long-lived and high-trophic level Arctic marine mammals, such as polar bear, pilot whale, narwhal, beluga and hooded seal. For these keystone species, a notable proportion of the population is at high or severe risk of health effects from Hg exposure. Terrestrial mammals, with the exception of Arctic fox on Iceland, were not at risk for Hg exposure mediated health effects assessed based on the limited recent Hg data available. Hotspot areas of Hg have been detected in Northwestern Arctic Canada and Northwest Greenland. These hotspots are likely to be driven by MeHg in the epipelagic layer. There is a need for an increased understanding of the adverse effects of Hg exposure on Arctic wildlife, and particularly in the face of a changing climate and how such changes are altering abiotic and biotic exposure pathways and exposure-effect relationships. We recommend more basic and applied research efforts to focus on defining and refining risk threshold values. There may also be a need for advances in multidisciplinary studies to further identify cumulative and interactive effects of Hg and other environmental stressors (e.g., other chemical contaminants, climate change, food-web structure, pathogens) on Arctic biota. For most Arctic species where their Hg concentrations indicate potential effects included in this report, little to no studies have been conducted to verify Hg impacts. Overall, we recommend more research efforts on linking relevant Arctic Hg hotspot species and regions to potential effects and even studies on population effects.

CRediT authorship contribution statement

Rune Dietz: Project administration, conceptualization, methodology, funding acquisition, graphics, tabelling, writing original draft, reviewing, editing, data curation. **Robert J. Letcher**: Project administration, conceptualization, methodology, writing contribution to original draft, reviewing, editing. **Sam Dastnai**: Writing minor sections of original draft, graphics, tabelling, reviewing, editing. **Jean-Pierre Desforges**: Writing section of original draft, reviewing, editing. **Igor Eulaers**: Programming, tabelling, contributions to original draft, reviewing. **Frank F. Rigét**, **Adam Morris, Jacob Nabe-Nielsen**: extracting data, running stats and graphics, contributions to original and final draft, reviewing **Christian Sonne**: Contributions to original draft, reviewing, funding acquisition, contributing to project administration. **Simon Wilson**: Data collection, graphics, writing minor sections of original manus, editing, contributing to project administration.

Jon Aars, Magnus Andersen, Aqqalu R. Asvid, Andrei Boltunov, Erik W. Born, Tomasz M. Ciesielski, Krishna Das, Andrew E. Derocher, Steve Ferguson, Ingeborg G. Hallanger, Mads P. Heide-Jørgensen, Lars-Eric Heimbürger-Boavida, Paul F. Hoekstra, Bjørn M. Jenssen, Stephen Gustav Kohler, Martin M. Larsen, Ulf Lindstrøm, Anna Lippold, Nynne H. Nielsen, Elizabeth Peacock, Marianna Pinzone, Heli Routti, Ursula Siebert, Garry Stenson, Gary Stern, Jakob Strand, Jens Søndergaard, Gabriele Treu, Gisli A. Víkingsson, Feiyue Wang, Jeffrey M. Welker, Øystein Wiig: Data and sample providers, writing, review and editing.

Declaration of competing interest

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

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Appendix A. Supplementary Material

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References

Ackerman, J.T., Eagles-Smith, C.A., Herzog, M.P., Hartman, C.A., Peterson, S.H., Evers, D.C., Jackson, A.K., Elliott, J.E., Vander Pol, S.S., Bryan, C.E., 2016. Avian mercury exposure and toxicological risk across western North America: a synthesis. Sci. Total Environ. 568, 749–769.

Agather, A.M., Bowman, K.L., Lamborg, C.H., Hammerschmidt, C.R., 2019. Distribution of mercury species in the Western Arctic Ocean (U.S. GEOTRACES GN01). Mar. Chem. 216 (103686).

AMAP, 1998. Arctic Pollution Issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2004. AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2011. AMAP Assessment 2011: Mercury in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2015. Human Health in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway (vii + 165 pp).

AMAP, 2016. AMAP Assessment 2015: Temporal Trends in Persistent Organic Pollutants in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway (vi +71pp).

AMAP, 2018. AMAP Assessment 2017: Biological Effects of Contaminants on Arctic Wildlife and Fish, Arctic Monitoring and Assessment Programme (AMAP), Co-Leads: R. Dietz and R.J. Letcher, Oslo, Norway (vii+84pp).

AMAP, 2021. AMAP Assessment 2021: Mercury in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Tromsø, Norway. 326 pp.AMAP/UNEP. Technical Background Report to the Global Atmospheric Mercury Assessment; Arctic Monitoring and Assessment Programme/UNEP Chemicals Branch, 2008; 159 pp. http://www.chem.unep.ch/mercury/Atmospheric_Emissions/Technical_background_report.pdf.

AMAP Assessment 2021: Mercury in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Tromsø, Norway. viii+287pp.

AMAP/UN Environment, 2019. Technical Background Report for the Global Mercury Assessment 2018. Arctic Monitoring and Assessment Programme, Oslo, Norway/UN Environment Programme, Chemicals and Health Branch, Geneva, Switzerland (viii + 426 pp including E-Annexes).

AMAP/UNEP, 2008. Technical Background Report to the Global Atmospheric Mercury Assessment. Arctic Monitoring and Assessment Programme, Oslo, Norway / UNEP Chemicals Branch, Geneva, Switzerland.

Barst, B.D., Basu, N., Chételat, J., 2022. Toxicological risk of mercury for fish and invertebrate prey in the Arctic. Sci. Total Environ. (2022/this issue).

Berlin, M., 1986. Mercury. In: Friberg, L., Nordberg, G.F., Vouk, V.B. (Eds.), Handbook on the toxicology of metals. Elsevier Science Publishers, Amsterdam, pp. 187–445 [Vol. II].

Bowman, K.L., Lamborg, C.H., Agather, A.M., 2020. A global perspective on mercury cycling in the ocean. Sci. Tot. Environ. 710, 136166.

Bradley, M.M., Perra, M., Ahlstrøm, Ø., Jenssen, B.M., Jørgensen, E.H., Fuglei, E., Muir, D.C.G., Sonne, C., 2018. Mandibular shape in farmed Arctic foxes (*Vulpes lagopus*) exposed to persistent organic pollutants. Sci. Total Environ. 646, 1063–1068.

Braune, B.M., Norstrom, R.J., Wong, M.P., Collins, B.T., Lee, J., 1991. Geographical distribution of metals in livers of polar bears from the Northwest Territories, Canada. Sci. Total Environ. 100, 283–299.

Brown, T.M., Fisk, A.T., Wang, X., Ferguson, S.H., Young, B.G., Reimer, K.J., Muir, D.C.G., 2016. Mercury and cadmium in ringed seals in the Canadian Arctic: influence of location and diet. Sci. Tot. Environ. 545-546, 503–511.

Charette, M.A., Kipp, L.E., Jensen, L.T., Dabrowski, J.S., Whitmore, L.M., Fitzsimmons, J.N., Williford, T., Ulfsbo, A., Jones, E., Bundy, R.M., Vivancos, S.M., Pahnke, K., John, S.G., Xiang, Y., Hatta, M., Petrova, M.V., Heimbürger-Boavida, L.-E., Bauch, D., Newton, R., Pasqualini, A., Agather, A.M., Amon, R.M.W., Anderson, R.F., Andersson, P.S., Benner, R., Bowman, K.L., Edwards, R.L., Gdaniec, S., Gerringa, L.J.A., González, A.G., Granskog, M., Haley, B., Hammerschmidt, C.R., Hansell, D.A., Henderson, P.B., Kadko, D.C., Kaiser, K., Laan, P., Lam, P.J., Lamborg, C.H., Levier, M., Li, X., Margolin, A.R., Measures, C., Middag, R., Millero, F.J., Moore, W.S., Paffrath, R., Planquette, H., Rabe, B., Reader, H., Rember, R., Rijkenberg, M.J.A., Roy-Barman, M., Rutgers van der Loeff, M., Saito, M., Schauer, U., Schlosser, P., Sherrell, R.M., Shiller, A.M., Slagter, H., Sonke, J.E., Stedmon, C., Woosley, R.J., Valk, O., van Ooijen, J., Zhang, R., 2020. The transpolar drift as a source of riverine and shelf-derived trace elements to the central arctic ocean. J. Geophys. Res.: Oceans 125 (5), e2019JC015920.

Chastel, O., Fort, J., Albert, C., Angelier, F., Barst D., B., Basu, N, Blévin, P., Boertmann, D, Bustnes, J.-O., Bustamante, P., Dietz, R., Eagles-Smith A., C., Eulaers, I., Gabrielsen W., G., Goutte, A., Letcher J., R., Gilchrist, G., Merkel R., F., Moe, B., Mosbech, A., Søndergaard, J., Tartu, S., Wilson, S., Ackerman T., J., 2022. Mercury exposure and potential health risk for Arctic seabirds and shorebirds. Sci. Tot. Environ. This Issue, xxx–yyy.

Cossa, D., Heimbürger, L.E., Sonke, J.E., Planquette, H., Lherminier, P., García-Ibáñez, M.I., Pérez, F.F., Sarthou, G., 2017. Sources, cycling and transfer of mercury in the Labrador Sea (Geotraces-Geovide cruise). Mar. Chem. 198, 64–69.

Cossa, D., Heimbürger, L.E., Pérez, F.F., García-Ibáñez, M.I., Sonke, J.E., Planquette, H., Lherminier, P., Boutorh, J., Cheize, M., Lukas, Menzel Barraqueta, J., Shelley, R., Sarthou, G., 2018. Mercury distribution and transport in the North Atlantic Ocean along the GEOTRACES-GA01 transect. Biogeosciences 15 (8), 2309–2323.

Desforges, J.P.W., Sonne, C., Levin, M., Siebert, U., De Guise, S., Dietz, R., 2016. Immunotoxic effects of environmental pollutants in marine mammals. Environ. Int. 86, 126–139.

Desforges, J.P., Levin, M., Jasperse, L., De Guise, S., Eulaers, I., Letcher, R.J., Acquarone, M., Nordøy, E., Folkow, L.P., Jensen, T.H., Grøndahl, C., Bertelsen, M.F., St. Leger J, Almunia J, Sonne C, Dietz R., 2017. The in vitro effects of polar bear and killer whale blubber-derived contaminant 'cocktail' mixtures on marine mammal immune cells. Environ. Sci. Technol. 51, 11431–11439.

Desforges, J.P., Bandoro, C., Shehata, L., Sonne, C., Dietz, R., Puryear, W.B., Runstadler, J.A., 2018a. Environmental contaminant mixtures modulate in vitro influenza infection. Sci. Total Environ. 634, 20–28.

Desforges, J.P., Levin, M., Jasperse, L., De Guise, S., Jensen, T.H., Grøndahl, C., Bertelsen, M.F., Sonne, C., Dietz, R., 2018b. Immune function in Arctic mammals: natural killer (NK) cell activity in polar bear, muskox and reindeer. Vet. Immunol. Immunopathol. 195, 72–75.

Desforges, J.P., Hall, A., McConnell, B., Rosing Asvid, A., Barber, J.L., Brownlow, A., De Guise, S., Eulaers, I., Jepson, P.D., Letcher, R.J., Levin, M., Ross, P.S., Samarra, F., Víkingson, G., Sonne, C., Dietz, R., 2018c. Predicting global killer whale population collapse from PCB pollution. Science 361, 1373–1376.

Dietz, R., Pacyna, J., Thomas, D.J., Asmund, G., Gordeev, V., Johansen, P., Kimstach, V., Lockhart, L., Pfirman, S.L., Riget, F.F., Shaw, G., Wagemann, R., White, M., 1998. Chapter 7: heavy metals. AMAP Assessment Report: Arctic Pollution Issues. Arctic Monitoring and Assessment Programme. Oslo, Norway, pp. 373–524.

Dietz, R., Riget, F., Born, E.W., 2000. Geographical differences of zinc, cadmium, mercury and selenium in polar bears (*Ursus maritimus*) from Greenland. Sci. Total Environ. 245, 25–48.

Dietz, R., Outridge, P.M., Hobson, K.A., 2009. Anthropogenic contribution to mercury levels in present day Arctic animals – A review. Sci. Tot. Environ. 407, 6120–6131.

Dietz, R, Born, E.W., Rigét, F.F., Aubail, A., Sonne, C., Drimmei, R.C., Basu, N., 2011. Temporal Trends and Future Predictions of Mercury Concentrations in Northwest Greenland Polar Bear (*Ursus maritimus*) Hair. Environ. Sci. Technol. 45, 1458–1465.

Dietz, R., Basu, N., Braune, B., O'Hara, T., Scheuhammer, T.M., Sonne, C., Andersen, M., Andreasen, C., Andriashek, D., Asmund, G., Aubail, A., Baagøe, H., Born, E.W., Chan, H.M., Derocher, A.E., Grandjean, P., Knott, K., Kirkegaard, M., Lunn, N., Messier, F., Obbard, M., Olsen, M.T., Peacock, E., Renzoni, A., Rigét, F., Skaare, J.U., Stern, G., Stirling, I., Taylor, M., Wiig, Ø., Aars, J., 2013. What are the toxicological effects of mercury in Arctic biota? Sci. Total Environ. 443, 775–790.

Dietz, R., Gustavson, K., Sonne, C., Desforges, J.-P., Rigét, F.F., Pavlova, V., McKinney, M.A., Letcher, R.J., 2015. Physiologically-based pharmacokinetic modelling of immune, reproductive and carcinogenic effects from contaminant exposure in polar bears (*Ursus maritimus*) across the Arctic. Environ. Res. 140, 45–55.

Dietz, R., Desforges, J.-P., Gustavson, K., Sonne, C., Rigét, F.F., Born, E.W., Letcher, R.J., 2018a. Immunologic, reproductive, and carcinogenic risk assessment in East Greenland polar bears (*Ursus maritimus*) during the period 1983-2013. Environ. Int. 118, 169–178.

Dietz, R., Mosbech, A., Flora, J., Eulaers, I., 2018b. Interactions of climate, socio-economics and global mercury pollution in the North Water region. AMBIO 47 (Suppl. 2), 281–295. https://doi.org/10.1007/s13280-018-1033z.

Dietz, R., Letcher, R., Desforges, J.-P., Eulaers, I., Sonne, C., Wilson, S., Andersen-Ranberg, E., Basu, N., Barst, B.D., Bustnes, J.O., Bytingsvik, J., Ciesielski, T.M., Drevnick, P.E., Gabrielsen, G.W., Haarr, A., Hylland, K., Jenssen, B.M., Levin, M., McKinney, M.A., Nørregaard, R.D., Pedersen, K.E., Provencher, J., Styrishave, B., Tartu, S., Aars, J., Ackerman, J.T., Rosing-Asvid, A., Barrett, R., Bignert, A., Born, E.W., Branigan, M., Braune, B., Bryant, C.E., Dam, M., Eagles-Smith, C.A., Evans, M., Evans, T.J., Fisk, A.T., Gamberg, M., Gustavson, K., Hartman, C.A., Helander, B., Herzog, M.P., Hoekstra, P.F., Houde, M., Hoydal, K., Jackson, A.K., Kucklickt, J., Lie, E., Loseto, L., Mallory, M.L., Miljeteig, C., Mosbech, A., Muir, D.C.G., Nielsen, S.T., Peacock, E., Pedro, S., Peterson, S.H., Polder, A., Rigét, F.F., Roach, P., Saunes, H., M-HS. Sinding, M., Skaare, J.U., Søndergaard, J., Stenson, G., Stern, G., Treu, G., Schuur, S.S., Víkingsson, G., 2019a. Current state of knowledge on biological effects from contaminants on Arctic wildlife and fish. Sci. Total Environ. 696 (133792), 1–39.

Dietz, R., Sonne, C., Albert, C., Bustess, J.O., Christensen, T.K., Ciesielski, T.M., Danielsen, J., Dastnai, S., Eens, M., Erikstad, K.E., Fort, J., Galatius, A., Garbus, S.-E., Gilg, O., Hanssen, S.A., Helander, B., Helberg, M., Jaspers, V.L.B., Jenssen, B.M., Kauhala, K., Kyhn, L.A., Labansen, A.L., Larsen, M.M., Lindstøm, U., Reiertsen, T.K., Rigét, F.F., Roos, A., Strand, J., Strøm, H., Sveegaard, S., Søndergaard, J., Sun, J., Teilmann, J., Therkildsen, O.R., Tjørnløv, R.S., Wilson, S., Eulaers, I., 2021a. A risk assessment of the effects of mercury on Baltic Sea, Greater North Sea and North Atlantic wildlife, fish and bivalves. Environ. Int. 146, 106178.

Dietz, R., Rigét, F.F., Eulaers, I., Desforges, J.-P., Vorkamp, K., Bossi, R., Søndergaard, J., Ambus, P., McKinney, M., Letcher, R.L., Sonne, C., 2021b. Unexpected Increases of Persistent Organic Pollutant and Mercury levels in East Greenland Polar Bears (UNEXPECTED). Aarhus University, DCE – Danish Centre for Environment and Energy, 44 pp. Technical Report No. 214. http://dce2.au.dk/pub/TR214.pdf.

Dietz, R., Sonne, C., Desforges, J.-P., Eulaers, I., Rosing Asvid, A., Letcher, R., Jepson, P., 2019b. Killer whales calls for further protection. Environ. Int. 126, 443–444. https://doi.org/10.1016/j.envint.2019.02.060.

Evers, D.C., Keane, S., Buck, D., Basu, N., 2016. Evaluating the effectiveness of the Minamata convention on mercury: principles and recommendations for next steps. Sci. Total Environ. 569, 888–903.

Fryer, R.J., Nicholson, M.D., 1999. Using smoothers for comprehensive assessments of contaminant time series in marine biota. ICES J. Mar. Sci. 56, 779–790.

Heimbürger, L.E., Sonke, J.E., Cossa, D., Point, D., Lagane, C., Laffont, L., Galfond, B., Nicolaus, M., Rabe, B., Rutgers van der Loeff, M., Rutgers van der Loeff, M., 2015. Shallow methylmercury production in the marginal sea ice zone of the central Arctic Ocean. Sci. Rep. 5 (1), 10318.

Kim, J., Soerensen, A.L., Kim, M.S., Eom, S., Rhee, T.S., Jin, Y.K., et al., 2020. Methylmercury in the east Siberian Sea: the importance of sediment sources. Environ. Sci. Technol. 54 (16), 9949–9957.

Kirkegaard, M., Sonne, C., Dietz, R., Jenssen, B.M., Leifsson, P.S., Jensen, J.E.B., Letcher, R.J., 2010a. Testosterone concentrations and male genital organ morphology in Greenland sled dogs (*Canis familiaris*) dietary exposed to organohalogen contaminants. Toxicol. Environ. Chem. 92, 955–967.

Kirkegaard, M., Sonne, C., Dietz, R., Jenssen, B.M., Letcher, R.J., 2010b. Organohalogens in a whale blubber supplemented diet reduce hepatic retinol and renal tocopherol concentrations in Greenland sled dogs. J. Toxicol. Environ. Health A 73, 773–786.

Kirkegaard, M., Sonne, C., Dietz, R., Letcher, R.J., Jensen, A.L., Jenssen, B.M., Grandjean, P., 2011. Alterations in thyroid hormone status in Greenland sledge dogs exposed to naturally OHC contaminated whale blubber. Ecotoxicol. Environ. Saf. 74, 157–163.

Laidre, K.L., Stern, H., Kovacs, K.M., Lowry, L., Moore, S.E., Regehr, E.V., Ferguson, S.H., et al., 2015. Arctic marine mammal population status, sea ice habitat loss, and conservation recommendations for the 21st century. Conserv. Biol. 00, 1–14.

Letcher, R.J., Bustnes, J.O., Dietz, R., Jenssen, B.M., Jørgensen, E.H., Sonne, C., Verreault, J., Vijayan, M.M., Gabrielsen, G.W., 2010. Effects assessment of persistent organohalogen contaminants in Arctic wildlife and fish. Sci. Total Environ. 408, 2995–3043.

Lippold, A., Aars, J., Andersen, M., Aubail, A., Derocher, A., Dietz, R., Eulaers, I., Sonne, C., Welker, J., Wiig, Ø., Routti, H., 2020. Two decades of mercury concentrations in Barents Sea polar bears (*Ursus maritimus*) in relation to dietary carbon, sulfur, and nitrogen. Environ. Sci. Technol. 54, 7388–7397.

Mori, N., Yamamoto, M., Tsukada, E., Yokooji, T., Matsumura, N., Sasaki, M., et al., 2012. Comparison of in vivo with in vitro pharmacokinetics of mercury between methylmercury chloride and methylmercury cysteine using rats and caco2 cells. Arch. Environ. Contam. Toxicol. 63, 628–636.

Morris, A.D., Wilson, S.J., Fryer, R.J., Thomas, P.J., Hudelson, K., Andreasen, B., Blévin, P., Bustamante, P., Chastel, O., Christensen, G., Dietz, R., Evans, M., Evenset, A., Ferguson, S.H., Fort, J., Gamberg, M., Grémillet, D., Houde, M., Letcher, R.J., Loseto, L., Muir, D., Pinzone, M., Poste, A., Routti, H., Sonne, C., Stern, G., Rigét, F.F., 2022. Temporal trends of mercury in Arctic biota: 10 more years of progress in Arctic monitoring. Sci. Total Environ. (this issue).

Nielsen, C.O., Dietz, R., 1990. Distributional pattern of zinc, cadmium, mercury, and selenium in livers of hooded seal (*Cystophora cristata*). Biol. Trace Elem. Res. 24, 61–71.

Nyman, M., Bergknut, M., Fant, M.L., Raunio, H., Jestoi, M., Bengs, C., Murk, A., Koistinen, J., Backman, C., Pelkonen, O., Tysklind, M., Hirvi, T., Helle, E., 2003. Contaminant exposure and effects in Baltic ringed and grey seals as assessed by biomarkers. Mar. Environ. Res. 55, 73–99.

Pedersen, K.E., Styrishave, B., Sonne, C., Dietz, R., Jenssen, B.M., 2015. Accumulation and potential health effects of organohalogenated compounds in the Arctic fox (*Vulpes lagopus*) - a review. Sci. Total Environ. 502, 510–516.

Petrova, M.V., Ourgaud, M., JRH, Boavida, Dufour, A., Tesán Onrubia, J.A., Lozingot, A., Heimbürger-Boavida, L.-E., 2020a. Human mercury exposure levels and fish consumption at the French Riviera. Chemosphere 258, 127232.

Petrova, M.V., Krisch, S., Lodeiro, P., Valk, O., Dufoura, A., Rijkenberg, M.J.A., Achterberg, E.P., Rabe, B., van der Loeff, M.R., Hamelin, B., Sonke, J.E., Garnier, C., Heimbürger-Boavida, L.-E., 2020b. Mercury species export from the Arctic to the Atlantic Ocean. Mar. Chem. 225, 103855.

Rigét, F.F., Muir, D., Kwan, M., Savinova, T., Nyman, M., Woshner, V., O'Hara, T., 2005. Circumpolar pattern of mercury and cadmium in ringed seals. Sci. Total Environ. 351–352 (2005), 312–322.

Rodríguez-Estival, J., Mateo, R., 2019. Exposure to anthropogenic chemicals in wild carnivores: a silent conservation threat demanding long-term surveillance. Curr. Op. Environ. Sci. Health 11, 21–25.

Rogstad, T.W., Sonne, C., Villanger, G.D., Ahlstrøm, Ø., Fuglei, E., Muir, D., Jørgensen, E., Jenssen, B.M., 2017. Concentrations of vitamin A, E, thyroid and testosterone hormones in blood plasma and tissues from emaciated adult male Arctic foxes (*Vulpes lagopus*) dietary exposed to persistent organic pollutants (POPs). Environ. Res. 154C, 284–290.

Ronald, K., Tessaro, S.V., Uthe, J.F., Freeman, H.C., Frank, R., 1977. Methylmercury poisoning in the harp seal (*Pagophilus groenlandicus*). Sci. Total Environ. 8, 1–11.

Routti, H., Arukwe, A., Jenssen, B.M., Letcher, R.J., Nyman, M., Bäckman, C., Gabrielsen, G.W., 2010. Comparative endocrine disruptive effects of contaminants in ringed seals (Phoca hispida) from Svalbard and the Baltic Sea. Comp. Biochem. Physiol. C 152, 306–312.

Routti, H., Letcher, R.J., Born, E.W., Branigan, M., Dietz, R., Evans, T.J., Fisk, A.T., Peacock, E., Sonne, C., 2011. Spatio-temporal trends of selected trace elements in liver tissue from polar bears (*Ursus maritimus*) from Alaska, Canada and Greenland. J. Environ. Monit. 13, 2260–2267.

Routti, H., Letcher, R.J., Born, E.W., Branigan, M., Dietz, R., Evans, T.J., McKinney, M.A., Peacock, E., Sonne, C., 2012. Influence of carbon and lipid sources on variation of mercury and other trace elements in polar bears (*Ursus maritimus*). Environ. Toxicol. Chem. 31 (12), 2739–2747.

Routti, H., Atwood, T., Bechshøft, T., Boltunov, A., Ciesielski, T., Desforges, J.P., Dietz, R., Gabrielsen, G.W., Jenssen, B.M., Letcher, R.J., McKinney, M., Morris, A., Rigét, F.F., Sonne, C., Styrishave, B., Tartu, S., 2019. State of knowledge on current exposure, fate and potential health effects of contaminants in polar bears from the circumpolar Arctic. Sci. Total Environ. 664, 1063–1083.

Sonne, C., 2010. Health effects from long-range transported contaminants in Arctic top predators: an integrated review based on studies of polar bears and relevant model species. Environ. Int. Environ. Int. 36, 461–491.

Sonne, C., Dietz, R., Leifsson, P.S., Born, E.W., Kirkegaard, M., Rigét, F.F., Letcher, R.J., Muir, D.C.G., Hyldstrup, L., 2005. Do organohalogen contaminants contribute to liver histopathology in East Greenland polar bears (Ursus maritimus)? Environ. Health Perspect. 113, 1569–1574.

Sonne, C., Dietz, R., Larsen, H.J.S., Loft, K.E., Kirkegaard, M., Letcher, R.J., Shahmiri, S., Møller, P., 2006a. Impairment of cellular immunity in West Greenland sledge dogs (Canis familiaris) dietary exposed to polluted minke whale (Balaenoptera acutorostrata) blubber. Environ. Sci. Technol. 40, 2056–2062.

Sonne, C., Leifsson, P.S., Dietz, R., Born, E.W., Letcher, R.J., Hyldstrup, L., Rigét, F.F., Kirkegaard, M., Muir, D.C.G., 2006b. Xenoendocrine pollutants may reduce size of sexual organs in East Greenland polar bears (Ursus maritimus). Environ. Sci. Technol. 40, 5668–5674.

Sonne, C., Dietz, R., Leifsson, P.S., Born, E.W., Kirkegaard, M., Letcher, R.J., Muir, D.C.G., Rigét, F.F., Hyldstrup, L., 2006c. Are organohalogen contaminants a co-factor in the development of renal lesions in East Greenland polar bears (Ursus maritimus)? Environ. Toxicol. Chem. 25, 1551–1557.

Sonne, C., Leifsson, P.S., Dietz, R., Kirkegaard, M., Møller, P., Jensen, A.L., Letcher, R.J., Shahmiri, S., 2007a. Renal lesions in Greenland sledge dogs (*Canis familiaris*) exposed to a natural dietary cocktail of persistent organic pollutants. Toxicol. Environ. Chem. 89, 563–576.

Sonne, C., Fonfara, S., Dietz, R., Kirkegaard, M., Letcher, R.J., Shahmiri, S., Andersen, S., Møller, P., 2007b. Multiple cytokine and acute phase protein gene transcription in West Greenland sledge dogs (*Canis familiaris*) dietary exposed to organic environmental pollutants. Arch. Environ. Contam. Toxicol. 53, 110–118.

Sonne, C., Dietz, R., Leifsson, P.S., Asmund, G., Born, E.W., Kirkegaard, M., 2007c. Are liver and renal lesions in East Greenland Polar Bears (Ursus maritimus) associated with high mercury levels? Environ. Health 6, 11.

Sonne, C., Dietz, R., Born, E.W., Leifsson, P.S., Andersen, S., 2008a. Is there a link between *Hypospadias* and organochlorine exposure in East Greenland Sledge Dogs (*Canis familiaris*)? Ecotoxicol. Environ. Saf. 69, 391–395.

Sonne, C., Dietz, R., Kirkegaard, M., Letcher, R.J., Shahmiri, S., Andersen, S., Møller, P., Olsen, A.K., Jensen, A.L., 2008b. Effects of organohalogen pollutants on haematological and urine clinical-chemical parameters in Greenland sledge dogs (*Canis familiaris*). Ecotoxicol. Environ. Saf. 69, 381–390.

Sonne, C., Leifsson, P.S., Dietz, R., Kirkegaard, M., Møller, P., Jensen, A.L., Letcher, R.J., 2008c. Greenland sledge dogs (*Canis familiaris*) exhibit liver lesions when exposed to low-dose dietary organohalogen contaminated minke whale (*Balaenoptera acutorostrata*) blubber. Environ. Res. 106, 72–80.

Sonne, C., Leifsson, P.S., Wolkers, H., Jenssen, B.M., Fuglei, E., Ahlstrøm, Ø., Dietz, R., Kirkegaard, M., Muir, D.C.G., Jørgensen, E., 2008d. Organochlorine-induced histopathology in kidney and liver tissue from Arctic fox (*Alopex lagopus*). Chemosphere 71, 1214–1224.

Sonne, C., Rigét, F.F., Jensen, J.E.B., Hyldstrup, L., Teilmann, J., Dietz, R., Kirkegaard, M., Andersen, S., Letcher, R.J., Jakobsen, J., 2008e. Does the nutrition profile of vitamins, fatty acids and microelements counteract the negative impact from organohalogen pollutants on bone mineral density in Greenland sledge dogs (*Canis familiaris*)? Environ. Int. 34, 811–820.

Sonne, C., Wolkers, H., Leifsson, P.S., Iburg, T., Munro Jenssen, B., Fuglei, E., Ahlstrøm, Ø., Dietz, R., Kirkegaard, M., Muir, D.C.G., Jørgensen, E., 2009a. Chronic dietary exposure to environmental organochlorine contaminants induces thyroid gland lesions in Arctic foxes (*Vulpes lagopus*). Environ. Res. 109, 702–711.

Sonne, C., Wolkers, H., Rigét, F.F., Beck Jensen, J.E., Teilmann, J., Jenssen, B.M., Fuglei, E., Ahlstrøm, Ø., Dietz, R., Muir, D.C.G., Jørgensen, E., 2009b. Mineral density and biomechanical properties of bone tissue from male arctic foxes (*Vulpes lagopus*) exposed to organochlorines and emaciation. Comp. Biochem. Physiol. C 149, 97–103.

Sonne, C., Gustavson, K., Rigét, F.F., Dietz, R., Birkved, M., Letcher, R.J., Bossi, R., Vorkamp, K., Born, E.W., Petersen, G., 2009c. Reproductive performance in East Greenland polar bears (*Ursus maritimus*) may be affected by organohalogen contaminants as shown by physiologically-based pharmacokinetic (PBPK) modelling. Chemosphere 77, 1558–1568.

Sonne, C., Dam, M., Leifsson, P.L., Dietz, R., 2010a. Liver and renal histopathology of North Atlantic long-finned pilot whales (*Globicephala melas*) with high concentrations of heavy metals and organochlorines. Toxicol. Environ. Chem. 92, 969–985.

Sonne, C., Larsen, H.J.S., Kirkegaard, M., Letcher, R.J., Dietz, R., 2010b. Trans-generational and neonatal immune suppression in West Greenland sledge dogs (*Canis familiaris*) exposed to organohalogenated environmental contaminants. Sci. Total Environ. 408, 5801–5807.

Sonne, C., Leifsson, P.S., Dietz, R., 2013. Liver and renal lesions in mercury contaminated narwhals (Monodon monoceros) from North West Greenland. Toxicol. Environ. Chem. 95, 515–528.

Sonne, C., Dietz, R., Rigét, F.F., Letcher, R.J., Munk Pedersen, K., Styrishave, B., 2014a. Steroid hormones in blood plasma from Greenland sledge dogs (*Canis familiaris*) dietary exposed to organohalogen polluted minke whale (*Balaenoptera acuterostrata*) blubber. Toxicol. Environ. Chem. 96, 273–286.

Sonne, C., Kirkegaard, M., Jacobsen, J., Jenssen, B.M., Letcher, R.J., Dietz, R., 2014b. Altered 25-hydroxyvitamin D3 in liver tissue from Greenland sledge dogs (*Canis familiaris*) dietary exposed to organohalogen polluted minke whale (*Balaenoptera acuterostrata*) blubber. Ecotoxicol. Environ. Saf. 104, 403–408.

Sonne, C., Gustavson, K., Dietz, R., Letcher, R.J., 2015. Physiologically-based pharmacokinetic modelling and verification of distribution, elimination and bioaccumulation of persistent organic pollutants in Greenland sledge dogs (*Canis familiaris*). Environ. Res. 140, 45–55.

Sonne, C., Gustavson, K., Dietz, R., Letcher, R.J., 2016. Risk evaluation of the Arctic environmental POP exposure based on critical body residue and critical daily dose using captive Greenland sledge dogs (Canis familiaris) as surrogate species. Environ. Int. 88, 221–227.

Sonne, C., Torjesen, P.A., Berg, K.A., Fuglei, E., Muir, D.C.G., Jenssen, B.M., Jørgensen, E., Dietz, R., Ahlstøm, Ø., 2017. Exposure to persistent organic pollutants reduces testosterone concentrations and affects sperm viability and morphology during the mating peak-period in a controlled experiment on farmed Arctic foxes (*Vulpes lagopus*). Environ. Sci. Technol. 51, 4673–4680.

Sonne, C., Leifsson, P.S., Søndergaard, J., Dietz, R., 2018a. Hepatic and renal histology and mercury concentrations of North West and North East Greenland narwhals (Monodon monoceros). J. Toxic. Environ. Health A https://doi.org/10.1080/15287394.2018.1435601.

Sonne, C., Jepson, P.D., Desforges, J.-P., Alstrup, A.K.O., Olsen, M.T., Eulaers, I., Hansen, M., Letcher, R.J., McKinney, M.A., Dietz, R., 2018b. Pollution threatens toothed whales. Science 361 (6498), 1208 (https://doi).

Svensson, C.J., Eriksson, A., Härkönen, T., Harding, K.C., 2011. Detecting density dependence in recovering seal populations. Ambio 40, 52–59.

Tesán Onrubia, J.A., Petrova, M.V., Puigcorbé, V., Black, E.E., Valk, O., Dufour, A., Hamelin, B., Buesseler, K.O., Masqué, P., Le Moigne, F.A.C., Sonke, J.E., Rutgers van der Loeff, M., Heimbürger-Boavida, L.-E., 2020. Mercury Export Flux in the Arctic Ocean Estimated from 234 Th/ 238 U Disequilibria. ACS Earth and Space Chemistry acsearthspacechem.0c00055.

Verreault, J., Dietz, R., Sonne, C., Gebbink, W.A., Shahmiri, S., Letcher, R.J., 2008. Comparative toxicokinetics of organohalogen contaminants in two top Greenland carnivores: captive sledge dogs (*Canis familiaris*) and wild polar bears (*Ursus maritimus*). Comp. Biochem. Physiol. C 147, 306–315.

Wang, K., Munson, K.M., Beaupré-Laperrière, A., Mucci, A., Macdonald, R.W., Wang, F., 2018. Subsurface seawater methylmercury maximum explains biotic mercury concentrations in the Canadian Arctic. Sci. Rep. 8.

Wang, Y.M., Li, L.J., Yao, C., Tian, X.S., Wu, Y.R., Xie, Q., Wang, D.Y., 2021. Mercury in human hair and its implications for health investigation. Curr. Opin. Environ. Sci. Health 22, 100271.

WHO 1993. Environmental Health Criteria 101: Methylmercury. Geneva, Switzerland: World Health Organization; 1993 [144 pp.].

Wobeser, G., Nielsen, N.O., Schiefer, B., 1976. Mercury and mink II. Experimental methyl mercury intoxication. Can. J. Comp. Med. 40, 34–45.

Wren, C.D., Hunter, D.B., Leatherland, J.F., Stokes, P.M., 1987. The effects of polychlorinated biphenyls and methylmercury, singly and in combination, on mink. I: uptake and toxic responses. Arch. Environ. Contam. Toxicol. 16, 441–447.