



HAL
open science

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

Rune Dietz, Robert Letcher, Jon Aars, Magnus Andersen, Andrei Boltunov, Erik Born, Tomasz Ciesielski, Krishna Das, Sam Dastnai, Andrew Derocher, et al.

► To cite this version:

Rune Dietz, Robert Letcher, Jon Aars, Magnus Andersen, Andrei Boltunov, et al.. A risk assessment review of mercury exposure in Arctic marine and terrestrial mammals. *Science of the Total Environment*, 2022, 829, pp.154445. 10.1016/j.scitotenv.2022.154445 . hal-03630638

HAL Id: hal-03630638

<https://hal.science/hal-03630638>

Submitted on 15 Nov 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

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

2
3 Rune Dietz^{a,*}, Robert J. Letcher^{b,*}, Jon Aars^c, Magnus Andersen^c, Aqqalu R. Asvid^d, Andrei
4 Boltunov^e, Erik W. Born^d, Tomasz M. Ciesielski^f, Krishna Das^g, Sam Dastnai^a, Andrew E. Derocher^h,
5 Jean-Pierre Desforges^{a,i}, Igor Eulaers^{a,c}, Steve Ferguson^{j,k}, Ingeborg G. Hallanger^c, Mads P. Heide-
6 Jørgensen^d, Lars-Eric Heimbürger-Boavida^{l,m}, Paul F. Hoekstraⁿ, Bjørn M. Jenssen^{a,f}, Stephen Gustav
7 Kohler^o, Martin M. Larsen^a, Ulf Lindstrøm^{p,q}, Anna Lippold^c, Adam Morris^r, Jacob Nabe-Nielsen^a,
8 Nynne H. Nielsen^d, Elizabeth Peacock^s, Marianna Pinzoneⁱ, Frank F. Rigét^a, Heli Routti^c, Ursula
9 Siebert^t, Garry Stenson^u, Gary Stern^v, Jakob Strand^a, Jens Søndergaard^a, Gabriele Treu^x, Gisli A.
10 Víkingsson^y, Feiyue Wang^v, Jeffrey M. Welker^{z,aa,ab}, Øystein Wiig^{ac}, Simon Wilson^{ad}, Christian
11 Sonne^a

12
13 ^aAarhus University, Arctic Research Centre (ARC), Department of Ecoscience, P.O. Box 358, DK-
14 4000 Roskilde, Denmark

15 ^bEcotoxicology and Wildlife Health Division, Environment and Climate Change Canada, National
16 Wildlife Research Centre, Carleton University, Ottawa, ON K1A 0H3, Canada

17 ^cNorwegian Polar Institute, Tromsø, NO-9296, Norway

18 ^dGreenland Institute of Natural Resources, P.O. Box 570, DK-3900 Nuuk, Greenland

19 ^eMarine Mammal Research and Expedition Centre, 36 Nahimovskiy pr., Moscow 117997, Russia

20 ^fDepartment of Biology, Norwegian University of Science and Technology, NO-7491 Trondheim,
21 Norway

22 ^gFreshwater and Oceanic sciences Unit of reSearch (FOCUS), University of Liege, 4000, Liege,
23 Belgium

24 ^hDepartment of Biological Sciences, University of Alberta, Edmonton, AB, Canada T6G 2E9

25 ⁱDepartment of Environmental Studies and Science, University of Winnipeg, Winnipeg, MB, Canada

26 ^jFisheries and Oceans Canada, 501 University Crescent, Winnipeg, MB R3T 2N6, Canada

27 ^kDepartment of Biological Sciences, University of Manitoba, Winnipeg, MB R3T 2N2, Canada

28 ^lGéosciences Environnement Toulouse, CNRS/IRD/Université Paul Sabatier Toulouse III, Toulouse,
29 France.

30 ^mAix Marseille Université, CNRS/INSU, Université de Toulon, IRD, Mediterranean Institute of
31 Oceanography (MIO) UM 110, Marseille, France.

32 ⁿGrain Farmers of Ontario Canada

33 ^oDepartment of Chemistry, Norwegian University of Science and Technology, Realfagbygget, E2-
34 128, Gløshaugen, NO-7491 Trondheim, Norway

35 ^pDepartment of Arctic and Marine Biology, UiT The Arctic University of Norway, NO-9037 Tromsø,
36 Norway

37 ^qDepartment of Arctic Technology, Institute of Marine Research, FRAM Centre, NO-9007 Tromsø,
38 Norway

39 ^rNorthern Contaminants Program, Crown-Indigenous Relations and Northern Affairs Canada, 15
40 Eddy Street, 14th floor, Gatineau, Quebec, Canada, K1A 0H4

41 ^sUSGS Alaska Science Center, 4210 University Dr., Anchorage, AK 99508-4626, USA

42 ^tInstitute for Terrestrial and Aquatic Wildlife Research, University of Veterinary Medicine Hannover,
43 Foundation, Werftstr. 6, DE-25761, Büsum, Germany

44 ^uNorthwest Atlantic Fisheries Centre, 80 East White Hills vie, Newfoundland and Labrador, St John's
45 A1C 5X1, Department DFO-MPO, Canada

46 ^vCentre for Earth Observation Sciences (CEOS), Clayton H. Riddell Faculty of Environment, Earth
47 and Resources, University of Manitoba, 586 Wallace Bld, 125 Dysart Rd., Winnipeg, Manitoba R3T
48 2N2, Canada

49 ^xLeibniz Institute for Zoo and Wildlife Research, Alfred-Kowalke-Str. 17, 10315 Berlin, Germany

50 ^yMarine and Freshwater Research Institute, Skúlagata 4, 101 Reykjavík, Iceland

51 ^zUniversity of Alaska Anchorage, Anchorage 99508, United States

52 ^{aa}University of Oulu, Oulu 90014, Finland

53 ^{ab}University of the Arctic, Rovaniemi 96460, Finland
54 ^{ac}Zoological Museum, University of Oslo, P.O. Box 1172, Blindern, N-0318 Oslo, Norway
55 ^{ad}Arctic Monitoring and Assessment Programme (AMAP) Secretariat, P.O. Box 8100 Dep., N-0032,
56 Oslo, Norway
57 *: Co-Corresponding authors. E-mail: rdi@ecos.au.dk. TEL: +45-30783172; robert.letcher@canada.ca TEL:
58 +1-613-998-6696

59 **Abstract**

60 There has been a considerable number of reports on Hg concentrations in Arctic mammals since the
61 last Arctic Monitoring and Assessment Programme (AMAP) effort to review biological effects of the
62 exposure to mercury (Hg) in Arctic biota in 2010 and 2018. Here, we provide an update on the state
63 of the knowledge of health risk associated with Hg concentrations in Arctic marine and terrestrial
64 mammal species. Using available population-specific data post-2000, our ultimate goal is to provide
65 an updated evidence-based estimate of the risk for adverse health effects from Hg exposure in Arctic
66 mammal species at the individual and population level. Tissue residues of Hg in 13 species across the
67 Arctic were classified into five risk categories (from No risk to Severe risk) based on critical tissue
68 concentrations derived from experimental studies on harp seals and minks. Exposure to Hg lead to
69 low or no risk for health effects in most populations of marine and terrestrial mammals, however,
70 subpopulations of polar bears, pilot whales, narwhals, beluga and hooded seals are highly exposed in
71 geographic hotspots raising concern for Hg-induced toxicological effects. About 6% of a total of
72 3,500 individuals, across different marine mammal species, age groups and regions, ar at high or
73 severe risk of health effects from Hg exposure. Temporal analyses indicated that the proportion of
74 polar bears at low or moderate risk has increased in East/West Greenland and Western Hudson Bay,
75 respectively. However, there remain numerous knowledge gaps to improve risk assessments of Hg
76 exposure in Arctic mammalian species, including the establishment of improved concentration
77 thresholds and upscaling to the assessment of population-level effects.

78 **Key words:** Biological effects, Circumpolar Arctic, Mercury, Wildlife, Marine mammals, Terrestrial
79 mammals

80	Table of Contents	
81	1. Introduction	5
82	2. Material and methods	6
83	2.1. Literature and data search	6
84	2.3. Risk analysis	6
85	2.4. Time trend analyses	7
86	2.5. Hotspot area detections	8
87	3. Marine mammals	8
88	3.1 Risk effects extrapolated from liver Hg concentrations in marine mammals	8
89	3.2 Risk effects extrapolated from hair Hg concentrations in polar bears	12
90	4. Terrestrial mammals	14
91	5. Estimating population effects from mercury loads in highly exposed wildlife	15
92	6. Temporal trends and risk for the highest Hg exposed species and regions	16
93	6.1 Polar bears	16
94	6.2 Ringed seals	17
95	6.3 Belugas	17
96	6.4 Pilot whales	18
97	6.5 Other marine mammals	18
98	6.6 Terrestrial mammals	18
99	7. Geographical hotspot areas in water and wildlife	18
100	8. Conclusions	21
101	9. Acknowledgements	22
102	10. References	22
103		
104		

105 **1. Introduction**

106 The circumpolar Arctic has been subject to minimal direct production, use and emission of industrial
107 contaminants such as mercury (Hg). Instead, Hg is long-range transported to the Arctic by
108 atmospheric and sea-currents (AMAP/ UNEP, 2009, 2019; AMAP, 2011). Methylmercury (MeHg)
109 is readily bioavailable and biomagnifies in lipid-rich Arctic marine food webs, and thus raises
110 heightened concern for the health of exposed wildlife and Indigenous human populations that largely
111 depend on marine wildlife for their traditional diet (AMAP, 2011). Although Hg is a naturally
112 occurring element, human activities and climate change have led to a 10-20-fold increase in Hg
113 concentrations in the Arctic environment compared to preindustrial times (Dietz et al., 2009, 2011).
114 As a result, an international treaty, UNEP's Minamata Convention on Mercury, was enacted in 2017
115 (Evers et al., 2016).

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

130 To our knowledge, there has been little effort to quantify population level effects of Hg exposure
131 despite multiple health effects that have been reported in field studies of Arctic species (Dietz et al.,
132 2019a; Routti et al., 2019). Establishing links between contaminant exposure and health outcomes is
133 a difficult task (Rodriguez-Estival and Mateo, 2019). Such information is however extremely
134 important to manage and conserve wildlife populations and provide evidence for regulation of
135 contaminant emissions. It is critical to measure individual exposure impacts in order to estimate
136 population-level effects using various modelling approaches, which consider physiological effects on
137 reproduction, immune and endocrine functioning as well as energy demands (Svensson et al., 2011).
138 This also requires a combination of controlled mechanistic studies (e.g. *in vitro* dose-response) and

139 when possible *in vivo* studies on key species, as has been reported from hotspot areas like the Baltic
140 primarily on POPs, which can be transferred to other pristine areas, including the Arctic. (Desforges
141 et al., 2016, 2017, 2018a, 2018b, 2018c; Nyman et al., 2003; Routti et al., 2010). The approach taken
142 here to address this issue is data-driven and combines toxicity data from relevant mammalian studies
143 with an established risk modeling framework. We follow the methods of Ackerman et al. (2016)
144 developed for North American birds, in which contaminant levels in tissues are used for Arctic
145 species in a circumpolar risk analysis using critical body burdens and risk quotient analysis. In the
146 present assessment, we have increased the liver data coverage from marine mammal species, regions
147 and age groups from 70 to 112 groups (increase of 60 %), increased the number of individuals from
148 2371 to 3772 (increase of 51 %), and added polar bear hair data from 685 individuals from 22 regions
149 and age groups compared to the Dietz et al. (2019) risk analyses. The data from terrestrial mammals,
150 although with much lower coverage, also increased from 8 to 16 species (100% increase), regions
151 and age groups and the number of individuals increased from 211 to 814 (increase of 386 %). We
152 combined species and areas at risk to the observed temporal trends, which is reported in more detail
153 for all available Arctic data series by Morris et al. (2022/this issue). Finally we created heat maps
154 from a generalized additive model (GAM) approach to examine the linkage between seawater MeHg
155 concentrations in the upper 400 m water column with tissue concentrations of ringed seal (*Pusa*
156 *hispidus*) and polar bear liver and hair concentrations (see details in Section 2.5. Hotspot area
157 detections for details on method and references).

158

159 **2. Material and methods**

160 2.1. Literature and data search

161 We did a search on PubMed, ScienceDirect, Google, Google Scholar, EBSCO, ProQuest,
162 ScienceDirect, MEDLINE and grey literature combining the terms “Arctic” and/or “mercury” and/or
163 “Hg” and/or “effects” and/or “marine mammals” and/or “terrestrial mammals” by ultimo 2020. In
164 addition, Hg data from work in prep from the appointed Key National Experts of the eight Arctic
165 countries as well as from other scientific groups working in the Arctic was included. Only Hg data
166 from accredited laboratories participating in the international QA programs were used. The study
167 design is based on a review of the existing literature for post-2000 articles as well as unpublished data
168 of Hg exposure in marine and terrestrial mammals from the Arctic and, where possible, the raw data
169 were extracted. In addition, Hg data from Baltic marine mammals were obtained from the projects
170 BONUS BALTHEALTH for comparative purposes.

171

172 2.3. Risk analysis

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

186
$$\ln(Hg_H) = (\ln(Hg_L) - 1.18748) / 0.79941$$

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

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

Species	Matrix	No risk	Low risk	Moderate risk	High risk	Severe risk	Reference
		NRC	LRC	MRC	HRC	SRC	
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

191

192 2.4. Time trend analyses

193 For species and regions where Hg concentrations of concern fell under the HRC and SRC (Table 1),
 194 time trend analysis was used to identify increasing or decreasing trends. The temporal trend analysis
 195 followed the methods of AMAP (in press) and Morris et al. (2022/this issue). In brief, the biota time
 196 series of Hg concentrations were assessed as changes in the log-transformed concentrations over time
 197 using linear mixed models. The type of temporal change considered was dependent on the number of
 198 years of data; for time-series including 7 or more years of data the non-linearity of the trend was
 199 evaluated by use of smoothers (Fryer and Nicholson, 1999). The Akaike Informations Criterion for

200 small sample size (AICc) was applied for selection between different model formulations. More
201 complex (smoothed) patterns of change were modelled over time using the method of Fryer and
202 Nicholson (1999).

203

204 2.5. Hotspot area detections

205 Since the last assessment, the Arctic Ocean has become one of the best observed oceans (Dastoor et
206 al., in review). The new seawater Hg species observations cover the Irminger and Labrador Sea
207 (Cossa et al., 2017, 2018), the Canadian Arctic Archipelago (Wang et al., 2018), the East Siberian
208 Sea (Kim et al., 2020), the East Greenland Shelf, the Fram Strait and the Barents Sea (Petrova et al.
209 2020b) and the central Arctic Ocean (Agather et al., 2019, Charette et al., 2020, Heimbürger et al.,
210 2015, Tesan et al., 2020). We combined MeHg seawater measurements from both the Arctic
211 GEOTRACES programme and newly collected data from “Arven etter Nansen Seasonal Cruise Q1
212 2021” (Kohler et al. in prep) to produce heat maps from a GAM model. These maps cover the upper
213 400 m of the water column from the Barents Sea to the Canadian Archipelago, a region accessible to
214 the seals and where the highest MeHg concentrations are usually detected in the Arctic Ocean (see
215 section 3.4). Comparable heat maps were produced for ringed seal liver and polar bear liver and hair
216 (Rigét et al., 2005; Routti et al., 2011) to compare with the MeHg hotspot areas of the upper 400 m
217 of the water column (Dietz et al., this study) to assess overlap in spatial patterns.

218

219 3. Marine mammals

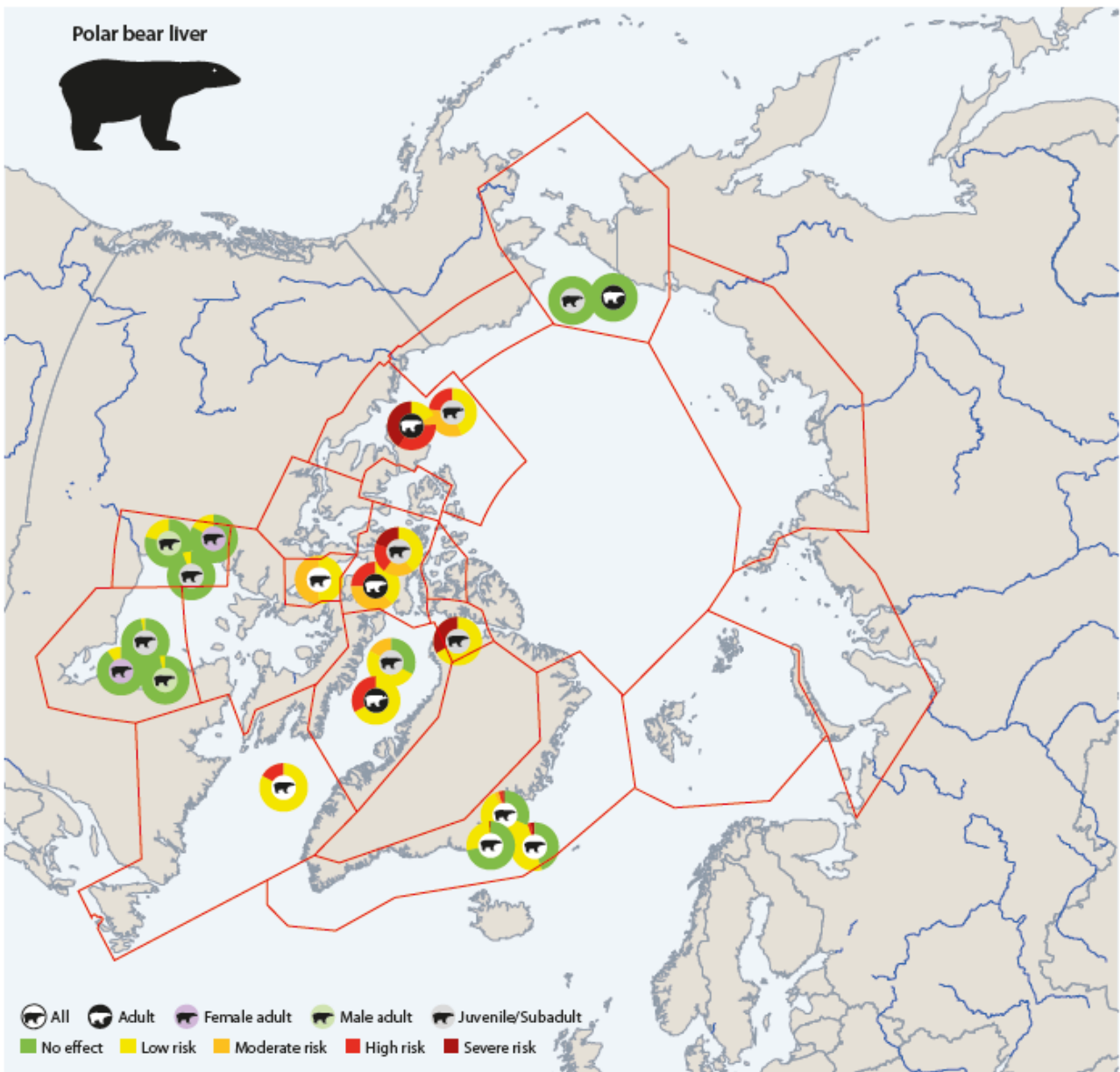
220 3.1 Risk effects extrapolated from liver Hg concentrations in marine mammals

221 Overall, 30 % of individuals from the 29 species, regions and age groups were within the two highest
222 risk categories, of which 18 of the 112 presented region/age/sex-groups) had 19 % individuals in the
223 SRC ($> 126 \mu\text{g/g ww}$) and an additional 11 groups (12 % of the individuals) in the HRC (83-126 $\mu\text{g/g}$
224 ww (Fig. S1; Table S1). Individuals from the 29 species, regions and age groups from the two highest
225 risk categories, however, accounted only for 200 individuals representing only 5.8 % of a total of
226 3445 individuals analyzed for their Hg loads. As for the SRC, this accounted for approximately 102
227 individuals (3.0 %) and the HRC accounted for the remaining 98 individuals (2.8 %) (Fig. S1, Table
228 S1). The highest exposed animal groups (evaluated by percentages in the SRC) are in the following
229 decreasing order: 1) adult hooded seals (*Cystophora cristata*) from the Denmark Strait (57 %), 2)
230 adult male hooded seals from Greenland Sea/Denmark Strait (45 %), 3) adult polar bears from the
231 Northern Beaufort Sea (41 %), 4) juvenile polar bears from Qaanaaq North West (NW) Greenland
232 (33 %) (Fig. 1), 5) adult killer whales (*Orcinus orca*) from East (E.) Greenland, Iceland and Faroe
233 Islands (33%), 6) adult long-finned pilot whales from the Faroe Islands (27%), 7) juvenile polar bears

234 from Lancaster/Jones Sound (20 %), 8) subadult long-finned pilot whale from the Faroe Islands (20
235 %), 9) adult female ringed seals from Sachs Harbor (12 %; Fig. 2) and 10) adult male ringed seals
236 from Sachs Harbor (7.8 %).

237 The least exposed animal groups (in liver and $\mu\text{g/g}$ on a wet weight basis; Figure S1, Table S1) in the
238 Arctic regions (i.e. groups with all 100 % of the individuals in the NRC are in the following increasing
239 order: 1) yearling harp seals (*Pagophilus groenlandicus*) from the Greenland Sea (in ww 0.17 $\mu\text{g/g}$),
240 2) foetus killer whales from E. Greenland, Iceland and Faroe Islands (0.18 $\mu\text{g/g}$), 3) subadult harbour
241 porpoises from the Barents Sea (0.49 $\mu\text{g/g}$), 4) adult male harbour porpoises from the Barents Sea
242 (0.58 $\mu\text{g/g}$), 5) subadult harbour porpoises from the Norwegian Coast (0.69 $\mu\text{g/g}$), 6) subadult harp
243 seals from the Greenland Sea/Denmark Strait (0.69 $\mu\text{g/g}$), 7) adult female harp seals from the
244 Greenland Sea/Denmark Strait (0.76 $\mu\text{g/g}$), 8) adult harp seals from Ittoqqortoormiit (0.78 $\mu\text{g/g}$), 9)
245 juvenile ringed seals from Qeqertarsuaq (0.92 $\mu\text{g/g}$); 10) juvenile ringed seals from Kangiqsujuaq
246 (0.92 $\mu\text{g/g}$). Additional information from the other risk categories Figure S1 and Table S1. In the
247 present review regions outside the Arctic (North Atlantic, North Sea, Inner Danish Waters and Baltic)
248 were include for comparative purpose for ringed seal and harbour porpoise (*Phocoena phocoena*).

249 There exists sufficient liver data for polar bears and ringed seals to provide an overview of the
250 regional differences and consistencies in their Hg exposure and the related risks. Six of the 20 polar
251 bear groups (regions, age and sex) from which we presented data had concentrations in the SRC. Of
252 these adult polar bears from the Northern Beaufort Sea were the highest exposed group with 41 % in
253 the SRC (Fig. 1; Table S1; Fig. S1). Juvenile bears from Qaanaaq and juvenile bears from Lancaster
254 Sound had 33 and 20 %, respectively, in the SRC although these sample sizes were small ($n = 6$ and
255 5, respectively). All three exposure risk groups in Ittoqqortoormiit had between 1.0 and 2.5 % percent
256 of the populations in the SRC based on sample sizes from 40 to 96 individuals. In four out of six
257 groups, individuals were also in the HRC ranging from 36 to 2.5 %. As for groups with the highest
258 exposed individuals in the HRC, from 33 to 17 % had individuals in this category. This included four
259 groups, including adult bears from Baffin Bay and Lancaster Sound as well as juveniles from
260 Northern Beaufort Sea and all age groups from the Davis Strait. Only two groups had individuals in
261 the MRC, and with the highest exposed groups being for the overall population from Gulf of Boothia
262 and juveniles from Baffin Bay. In the remaining eight groups, all individuals were in the two lowest
263 risk categories, of which 100 % of the juvenile and adult groups from the Chuckhi Sea were in the
264 NEC. Information on polar bears from the Russian Arctic is lacking.



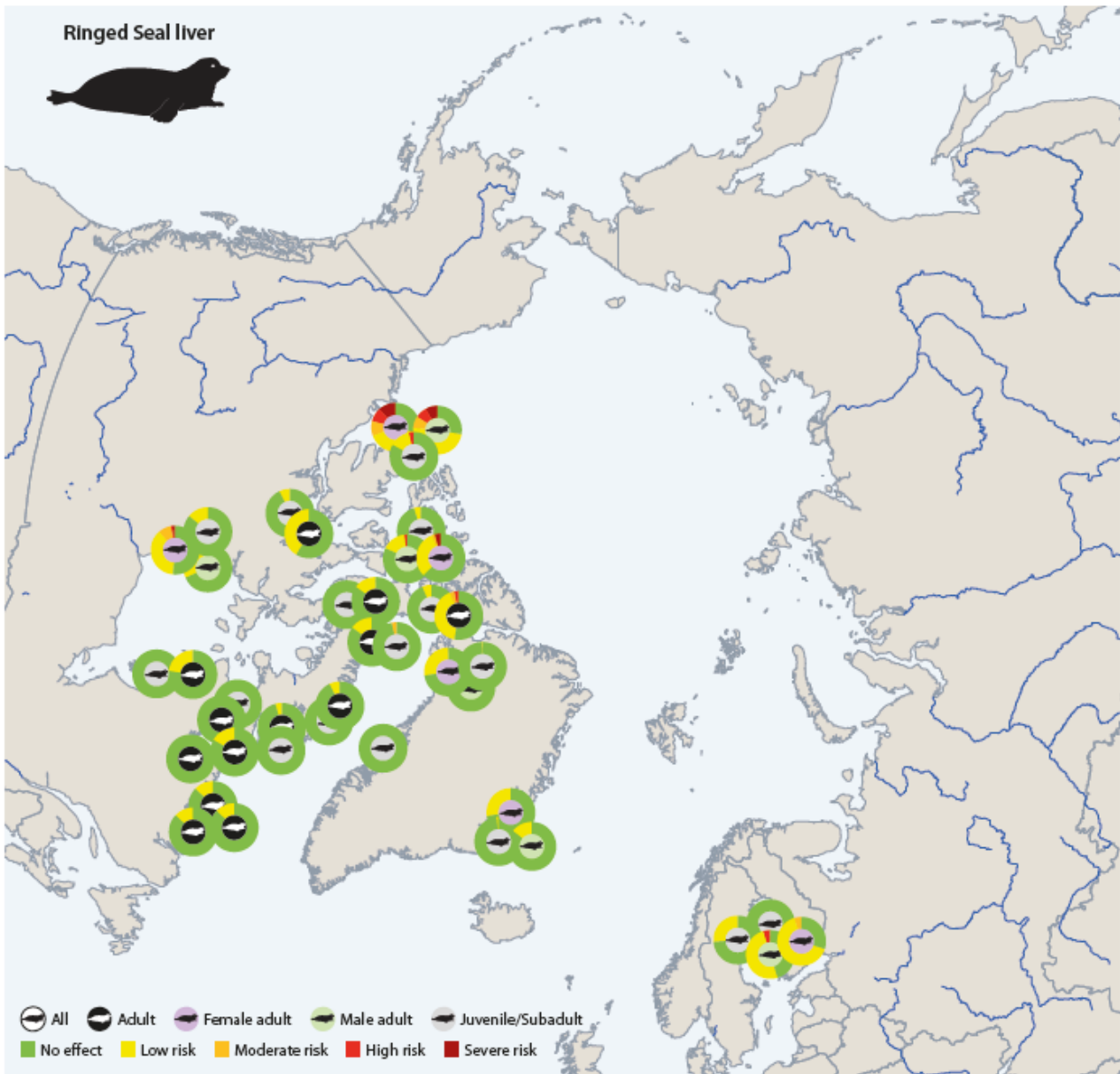
265

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

271

272 Four out of the 41 presented ringed seals region, age and sex groups had individuals in the SRC.
 273 Adult female and male ringed seals from Sachs Harbor were the highest exposed group with 11.5 and
 274 7.8 % of the individuals in the SRC, respectively (Fig. 2; Table S1; Fig. S1). Adult female ringed
 275 seals from Resolute and Arviat W. Hudson Bay were the other two groups with 3.9 and 1.4 % of the
 276 individuals in the SRC respectively. Most of these four groups also had individuals in the HRC and

277 MRC with 9.6 to 1.4 % and 9.6 to 2.0 % respectively. In addition, three groups including subadult
278 ringed seals from Sachs harbor, adults from Grise Fjord, and adult males from Resolute had 3.4 to
279 2.1 % of the individuals in the HRC. Additionally two groups, juvenile ringed seals from Pond Inlet
280 and adult males from Arviat, had 3.4 and 2.3 % of the individuals in the MRC respectively, and five
281 out of seven of the two highest risk categories also had 9.6 to 2.0 % of the individuals in the MRC.
282 For 28 groups of 37 groups of Arctic ringed seals, all individuals ranged in the two lowest risk
283 categories. As for the polar bears livers, information on ringed seals from the Russian Arctic is
284 lacking. For comparative purpose, data from Baltic ringed seals are included in Fig. 2 (see Fig. S1
285 and Table S1).



286

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

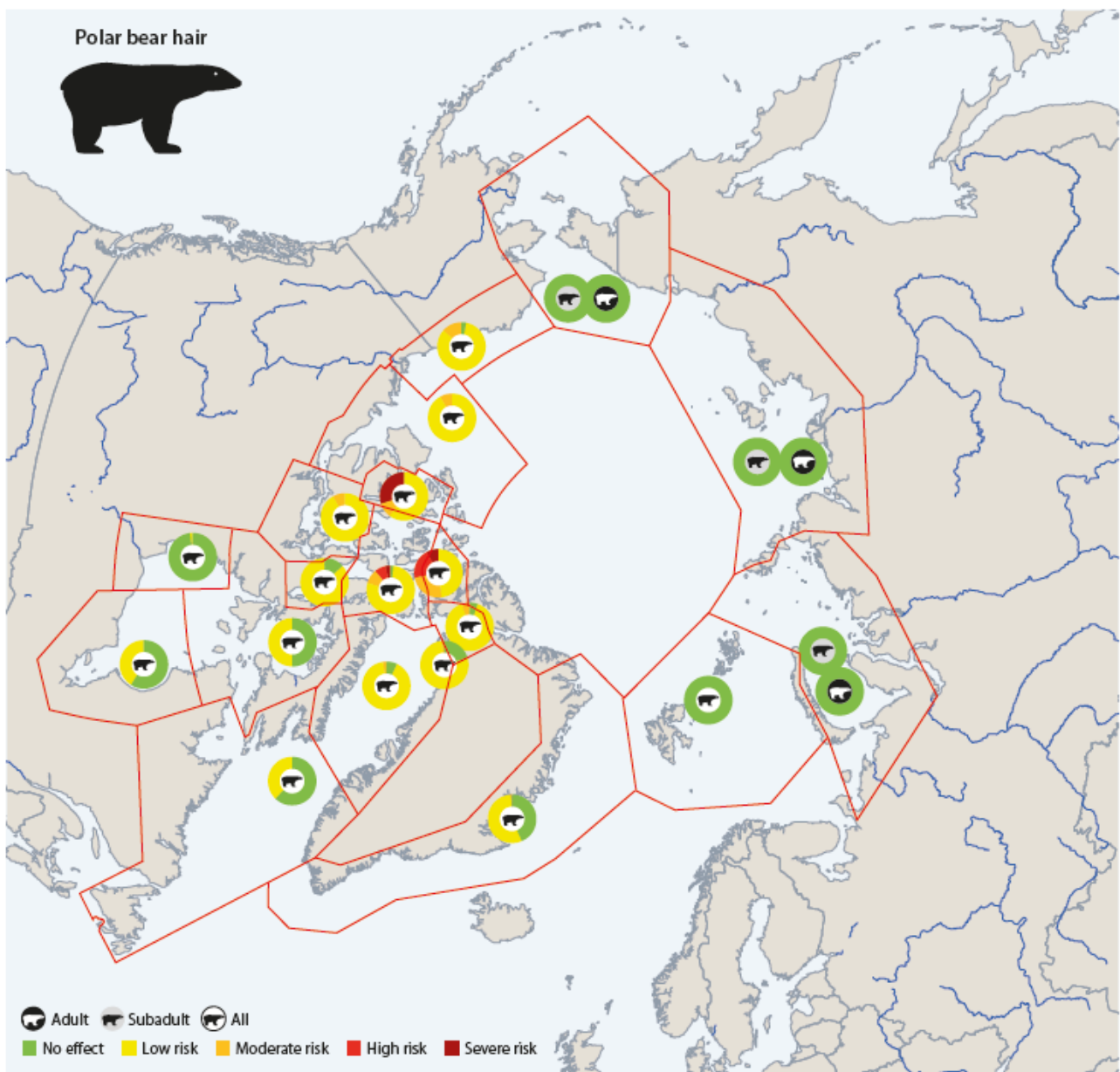
291

292 3.2 Risk effects extrapolated from hair Hg concentrations in polar bears

293 Hair samples are often used to evaluate human exposure and risk to Hg (Wang et al., 2021, Petrova
 294 et al. 2020a). Similarly, here we conducted an assessment of the regional risk of polar bears based on
 295 polar bear hair samples collected from hunting as well as hair sampled from polar bears that were
 296 routinely collected during tagging studies. We use hair samples collected before 2000 to obtain a
 297 better spatial coverage. The analyses revealed that some bears from three populations in the central
 298 and northeastern Canadian Arctic, had concentrations in the SRC, namely Viscount Melville Sound

299 (30 %), Norwegian Bay (8.0 %), and Lancaster Sound (3.6 %) (Fig. 3; Fig. S2; Table S2). The
300 corresponding percentages of bears from these three populations in the HRC were 0.0, 20 and 7.4 %,
301 respectively, and only a low percentage (0.7 %) of bears from East Greenland had individuals in the
302 HRC. As all the Canadian populations were sampled before year 2000, the present day risk patterns
303 are uncertain. Overall, the combined risk assessment using hair and liver Hg concentrations raises
304 concern for polar bears in the Canadian High Arctic and Northwestern Greenland. In contrast, hair
305 Hg concentrations in Barents, Kara, Laptev and Chukchi Sea all fell within the NEC, which indicated
306 that there are no Hg effect implications for polar bears in the regions of Svalbard and northern Russia
307 (Lippold et al. 2020; resubmitted).

308



309

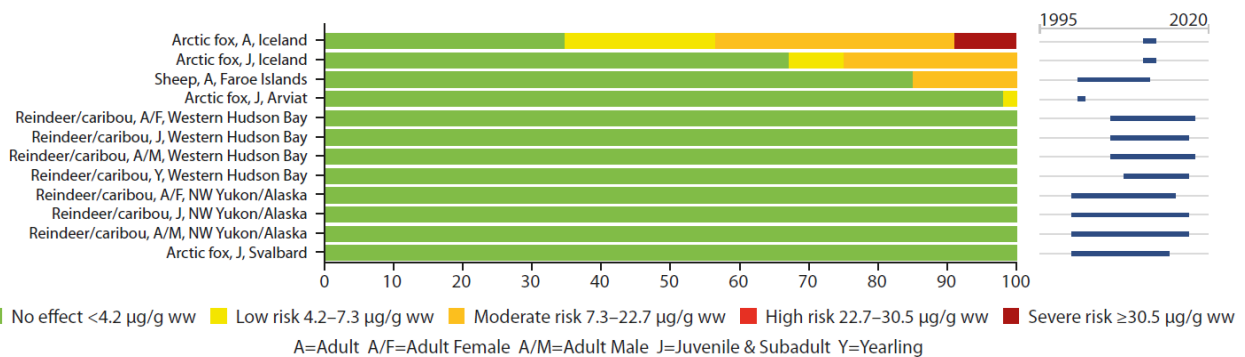
310 **Fig. 3.** The risk of Hg-mediated health effects in polar bear subpopulations based on pre- and post-2000
 311 monitoring hair data. The five risk categories are defined using effect threshold categories observed for harp
 312 seals (Ronald et al., 1977) converted into hair Hg concentrations by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

314

315 **4. Terrestrial mammals**

316 The majority of Hg concentrations in terrestrial mammals fell within the two lowest risk categories
 317 for Hg-mediated health effects (NRC and LRC, see Fig. 3). Arctic fox (*Vulpes lagopus*) from Iceland
 318 however, had 9.0 % of the adult population being at HRC, 35 % in the MRC, 22 % in the LRC and
 319 35 % in the NEC. Juvenile Arctic foxes were, however, exposed to lower levels, as the majority of
 320 the foxes (67 %) in this age group fell in the NEC. Juvenile Arctic foxes from Arviat and Svalbard
 321 had 98 % and 100 % in the NRC respectively, which raises the question on whether some local Hg
 322 sources are present on Iceland to cause elevated risk for the species there (Fig. 4 and Table S3). For
 323 sheep (*Ovis aries*) on the Faroe Islands, as much as 15 % were found in the MRC, which is higher
 324 than expected and could be attributed to agricultural fertilization by fish remains or eutrophication by
 325 bird droppings (from the extensive seabird colonies on the islands) as suggested by AMAP (2018)
 326 and Dietz et al. (2019). The remaining 85 % of the sheep fell in the NRC. All (100 %) seven
 327 Caribou/reindeer (*Rangifer tarandus*) populations and age groups were in the NRC with median Hg
 328 concentrations ranging from 0.12 to 1.24 µg/g ww (Fig. 4, Table S3). It should be noted that the risk
 329 categories are based on liver Hg threshold values from studies in mink, which is a carnivore (Wobeser
 330 et al., 1976; Wren et al., 1987, Dietz et al., 2019; AMAP, in press). Sheep and caribou/reindeer are
 331 herbivores/ungulates, and it is possible that the threshold level of Hg may differ from that used for
 332 the carnivores herein

333



334

335 **Fig. 4.** The proportion of individuals ranked from highest to lowest of specific Arctic terrestrial mammal
 336 populations according to the risk of total Hg-mediated health effects. Based on liver Hg concentrations from
 337 2000 to 2015, assignments were made to five risk categories and based upon effect threshold categories

338 observed for mink (Wobeser et al., 1976; Wren et al., 1987). See Table S3 for the detailed information upon
339 which this summary graphic is based.

340

341 **5. Estimating population effects from mercury loads in highly exposed wildlife**

342 It is challenging to assess the impact of Hg exposure and accumulation at the population-level for any
343 species, however, especially so for species living in remote areas like the Arctic. These assessments
344 require long term population monitoring on tissue Hg levels and relevant long-term individual fitness
345 and population metrics such as adult survival, reproductive success and recruitment (i.e. offspring
346 survival to reproductive age), and ultimately population growth rates. Unfortunately, the toxicity data
347 derived from harp seals (Ronald et al., 1977) and used to generate the risk categories used in our study
348 did not report effects on reproductive performance. Observed effects in the HRC included potential
349 for organ lesions (kidney, liver), anorexia, and reduced growth, while effects in the SRC included
350 severe impacts on organ function (i.e. kidney failure), weight loss, and ultimately increased mortality.
351 For toxicity in mink (i.e. terrestrial mammal assessment), observed effects in the HRC included
352 reduced litter size and offspring growth rate, and SRC included brain lesions, reduced growth,
353 anorexia, and increased adult mortality. Given the direct consequences on reproduction and survival
354 for the HRC and SRC, two endpoints of key concern for potential population impacts, concern is
355 warranted for select populations of hooded seals, killer whales, and pilot whales, as well as for
356 Lancaster Sound and Northern Beaufort Sea polar bears and harbour porpoises in the Danish Straits.
357 Here, >20 % of sampled individuals within these populations (and up to 60-90 %) had concerning
358 tissue Hg levels; impacts in such a large proportion of the population has the potential to have a
359 meaningful affect demographic rates and overall population fitness. For polar bears in highly exposed
360 regions like the Lancaster Sound and Jones Sound the population size trends are data deficient
361 (Vongraven and York, 2014; Dietz et al., 2015), making it difficult to track potential effects of Hg
362 exposure. From areas like the S. Beaufort Sea and the Baffin Bay, populations of marine wildlife are
363 declining, but the effects of climate change are likely to play a major role in these areas (*Ibid.*).. If the
364 mink toxicity data translates to Arctic foxes, our assessment suggests that a large portion of the Arctic
365 fox population in Iceland is at high risk for potential population relevant impacts from Hg exposure.
366 It is important to note that the above population assessment is based on toxicity data from only one
367 relevant species for each group (e.g. harp seal and mink). Care must be taken to extrapolate effects
368 across species because of potential inter-species differences in Hg toxicokinetics (e.g. uptake and
369 distribution) and toxicodynamics (e.g. species sensitivity to effects). Because such differences,
370 including differences between carnivores and herbivores, are unknown at this time and difficult to
371 assess, the current risk exercise provides the best available evidence-based assessment of potential
372 impacts across Arctic species. Furthermore, Arctic species may also be impacted at the population-

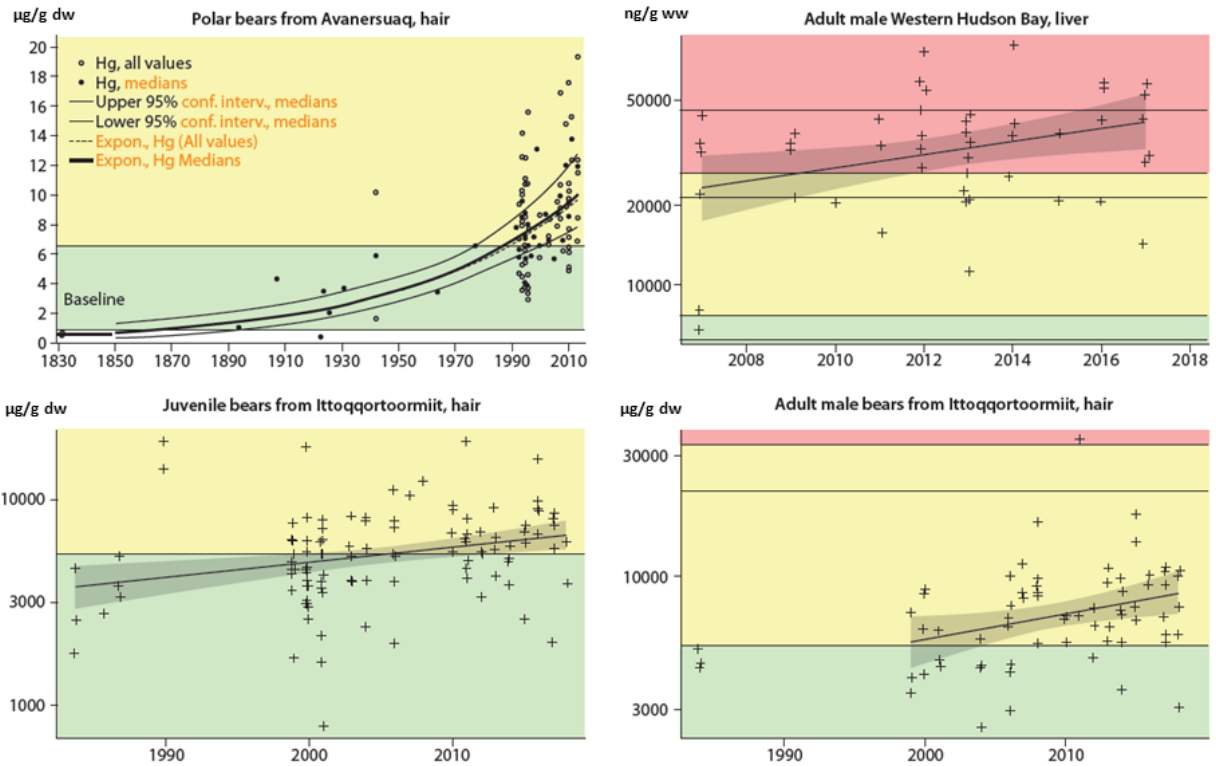
373 level through similar effects on reproduction and adult survival due to exposure to other contaminants
374 (POPs including PFASs) and stress related to climate change and hunting (e.g. Laidre et al., 2015;
375 Dietz et al., 2015, 2019a). Teasing out the effects of Hg from other concurrent stressors remains a
376 challenge, though it is expected that these stressors act in concert to increase the overall stress of
377 individuals and populations. Overall, more work on exact risk benchmark values for different species
378 and regions is recommended as well as population effect studies in relation to Hg and other
379 contaminant loads as conducted for killer whales by Desforges et al. (2018).

380

381 **6. Temporal trends and risk for the highest Hg exposed species and regions**

382 **6.1 Polar bears**

383 Twenty region, age and sex groups from 10 polar bear management areas were assessed for trends in
384 Hg risk based on their long-term liver concentration data (Figs. 1 and 3; Fig S1; Table S1). Four polar
385 bear management areas (Western Hudson Bay, Ittoqqortoormiit and Svalbard) and four out of 13
386 analyzed groups showed significant increasing Hg temporal trends, but no groups showed declining
387 Hg loads (Fig. 5). For the regions with the highest Hg risks, namely Lancaster Sound/Jones Sound
388 (SRC: 0-25 %; HRC: 20-25 %) as well as Northern Beaufort Sea (SRC: 0-41 %; HRC: 22-35 %) and
389 Qaanaaq region (SRC: 33 %), unfortunately no time trend information was available. However,
390 yearly significant Hg increases of 1.6-1.7 % ($p < 0.0001$) per year from 1892 to 2008 have previously
391 been reported for polar bear hair from the Qaanaaq, NW Greenland (Fig. 5; Dietz et al., 2011). In
392 Western Hudson Bay a significant increasing trend of 6.0 % per year was likewise detected in the
393 liver of adult males, whereas no trend was observed in juveniles and or adult females. Despite lower
394 Hg concentrations in polar bears from Ittoqqortoormiit, CE Greenland (SRC: 1.0-33 %; HRC: 3.0
395 %), significant temporal increases in hair Hg levels for juveniles and adults suggest that a larger
396 percentage of the population is likely to appear in the higher risk categories in the future. The hair
397 risk analyses for polar bears were quite similar to the liver risk results (Figs. 1 and 3; Tables S1 and
398 S2) as both Lancaster Sound, Norwegian Bay, Viscount Melville Sound, Northwest Greenland and
399 East Greenland had animals in the SRC and in the HRC (SRC: 4.0-33 %; HRC: 1.0-20 %).



400

401 **Fig. 5.** Examples of significant Hg increases in polar bear hair and liver from Canadian and Greenlandic waters
 402 plotted on top of risk intervals (No risk (green), Low risk (yellow). Moderate risk (orange), High risk (red) and
 403 severe risk (dark red)) defined in Table 1.

404

405 **6.2 Ringed seals**

406 Thirty-seven groups from 18 Arctic regions were assessed for trends in Hg risk based on their long-
 407 term liver concentration data (Fig. 2; Fig. S1; Table S1). Only one of seven ringed seal regions
 408 (Labrador Sea) and one out of 22 analyzed groups (4.5 %) showed significant increasing temporal
 409 trends of THg (Morris et al. (this issue)). However, four (Eastern Beaufort Sea, Lancaster Sound
 410 (Resolute Passage), Labrador Sea and Western Hudson Bay) out of the seven ringed seal regions and
 411 six out of 22 analyzed groups (27 %) showed significant declining Hg trends in muscle tissue which
 412 has a higher power than liver (Morris et al., 2022/this issue). Only adult seals (males and females)
 413 from the Labrador Sea showed significant increases in their livers. Overall, we conclude that ringed
 414 seals are not at significant risk with respect to increases in Hg exposure over time.

415

416 **6.3 Belugas**

417 Of the 10 temporal trend analyses conducted for muscle and liver tissues of belugas from Southern
418 Beaufort Sea and Southern Hudson Bay, three significant declines (30 %) were detected and no
419 significant increases. With the rather low percentages of belugas having individuals in the SRC and
420 HRC it is encouraging that a large proportion of these whales are showing significant declines in their
421 Hg loads.

422

423 **6.4 Pilot whales**

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

430

431 **6.5 Other marine mammals**

432 However, as no time trend analyses were available from these groups no overall evaluation can be
433 made with respect to temporal patterns of these risks.

434

435 **6.6 Terrestrial mammals**

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

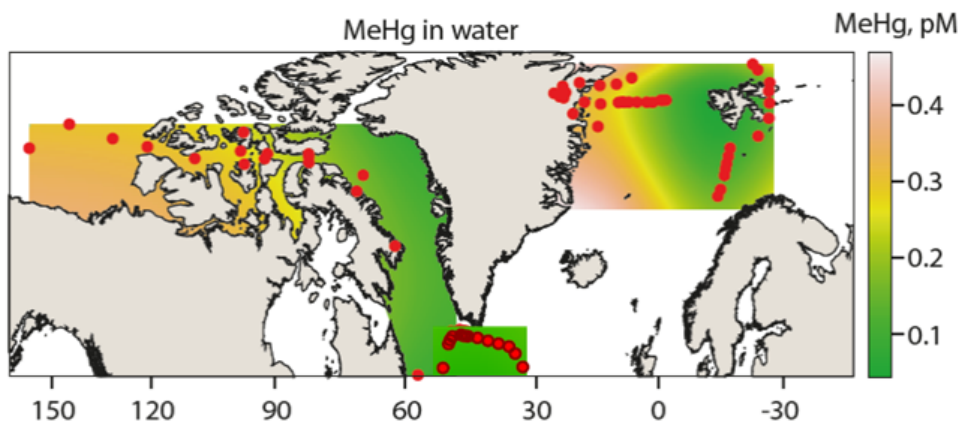
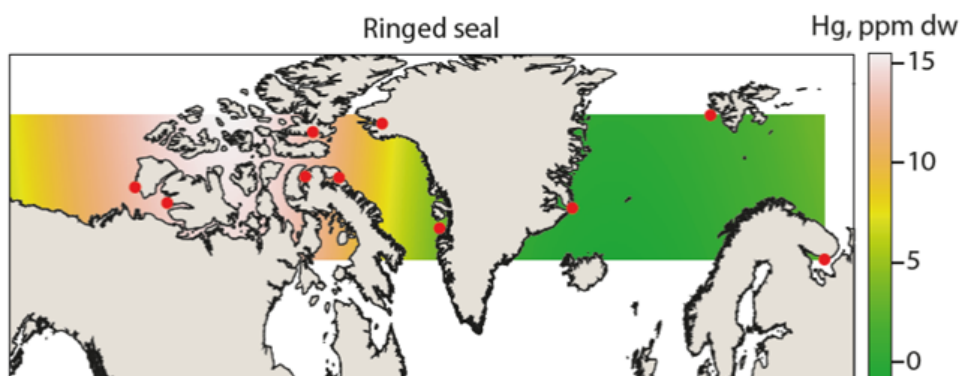
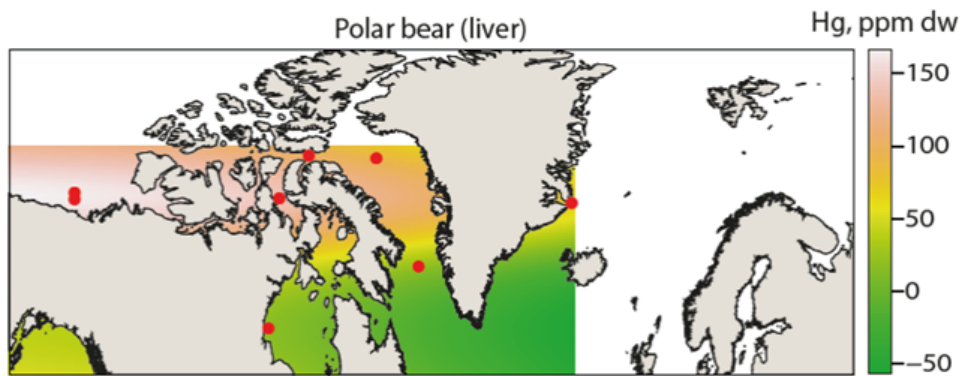
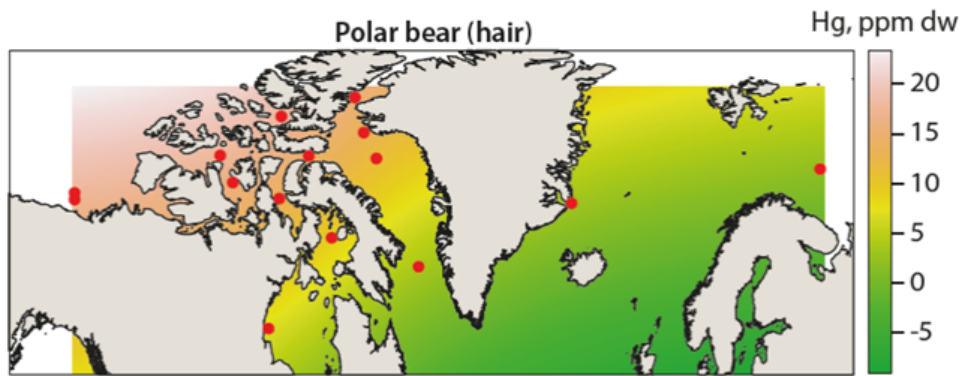
440

441 **7. Geographical hotspot areas in water and wildlife**

442 It is clear from the risk categorizations presented above that there are “hotspot” populations and
443 regions. The marine mammal species with the highest Hg concentrations for seals are the hooded and
444 ringed seals. For hooded seals, populations from the Denmark Strait and from Greenland
445 Sea/Denmark Strait, (adult males) show the highest levels, and where this is also the case for ringed
446 seals from Sachs Harbour (adults) (Fig. S1; Table S1; Fig. 2). For polar bears, adults from the
447 Northern Beaufort Sea as well as juveniles from Qaanaaq (NW Greenland) and Lancaster/Jones
448 Sound are those that are most exposed. For toothed whales, populations with the highest

449 concentrations are found in killer whales from E. Greenland, Iceland and Faroe Islands (adults), and
450 long-finned pilot whales from the Faroe Islands (adult and subadult).

451 Previous publications have considered geographical hotspots with respect to Hg biomagnification and
452 adverse biological effects in mammals such as ringed seals and polar bears (e.g. AMAP, 1998; Brown
453 et al., 2016; Dietz et al., 1998, 2000b, 2013a, this assessment; Rigét et al., 2005; Routti et al., 2011,
454 2012). Most of these surveys have shown that the Canadian Arctic Archipelago and northwestern
455 Greenland are hotspots for Hg exposure in biota. Heimbürger et al. (2015) put the idea forth that the
456 shallow seawater MeHg maximum in the Arctic Ocean, typically found near 200 m depth (vs other
457 oceans 500-1000 m, Bowman et al. 2020), may be responsible for the high Hg levels of arctic biota.
458 Wang et al. (2018) recently reported high-resolution vertical profiles of total Hg and MeHg in
459 seawater during GEOTRACES ship transect surveys conducted in 2015 from the Labrador Sea and
460 across Baffin Bay to the Canadian Arctic Archipelago and Canada Basin. They showed the highest
461 Hg concentrations in the Beaufort with a distinctive subsurface maximum of MeHg in seawater in
462 the upper 400 m depths peak concentration decrease from Canada Basin eastwards (Fig. 5). It was
463 hypothesized that Hg concentration in seawater was linked to Hg exposure in biota. Therefore, we
464 produced a heat map from a generalized additive model (GAM) approach covering the upper 400 m
465 of the water column for available seawater with the 2016 GEOTRACES GRIFF transect data obtained
466 between Northeast Greenland and Svalbard (Petrova et al., 2020b). Similar heat maps were produced
467 for ringed seals liver and polar bear liver and hair based on data from Rigét et al. (2005), Routti et al.
468 (2011) and Dietz et al. (this study). here between the hotspot areas in the water column and in
469 Northern Canada and Northwest Greenland (Rigét et al., 2005; Routti et al., 2011; Wang et al., 2018;
470 Dietz et al., this study). The high MeHg concentrations in the water column off Greenland could not
471 be detected in the biota due a lack of samples from this region. The ringed seal and polar bear
472 concentrations along Svalbard are somehow lower in concordance with lower concentrations of
473 MeHg in the water columns towards Svalbard compared to Northeast Greenland. Further modelling
474 work from larger parts of the Arctic with more transects and more species, corresponding years and
475 seasons, would be for future comparisons. Seabirds were included in a similar modelling exercise in
476 the AMAP Mercury Assessment (2022/in press) and showed corresponding hotspots in the Canadian
477 High Arctic. Comparisons would also benefit from normalization of data across time periods, age and
478 sex groups, and dietary patterns.



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

484

485 **8. Conclusions**

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

505

506 **9. Acknowledgements**

507 Support was provided for participants in the core group of the AMAP Assessment by the DANCEA
508 (Danish Cooperation for Environment in the Arctic) program for employees at Aarhus University
509 while support for Canadian participants was received from the Northern Contaminants Program
510 (NCP; Crown-Indigenous Relations and Northern Affairs Canada (CIRNAC)), Pangnirtung Hunters
511 and Trappers Association, Environment and Climate Change Canada (ECCC), Chemicals
512 Management Plan (CMP; ECCC and Health Canada), Department of Fisheries and Oceans (DFO)
513 Canada and the Natural Sciences and Engineering Research Council of Canada. Research Council of
514 Norway (RCN#276730) is also acknowledged for support to the Nansen Legacy Project, as well as
515 Anders Jahres Fund and the scientists and crew of the 2021 AeN Q1 Cruise. Funding sources for the
516 large number of additional writing, data and sample contributors are likewise acknowledged. We
517 gratefully acknowledge the reviewers including those that peer-reviewed the AMAP Effects
518 Assessment, which is the basis of the present journal review. Likewise, thanks to the numerous people
519 including colleagues and Northern and Indigenous hunters in various circumpolar jurisdictions and
520 communities who provided their expertise and assistance with sample collections, analytical work
521 and age determination for the many published studies as well as any new data that were used in the
522 present study. The following persons generously provided data or samples, but did not find their
523 contribution important enough to justify a co-authorship: Birgitta Andreasen, Dennis Andriashek,
524 Aurora Aubail, Eva Fuglei, Mary Gamberg, Magali Houde, Katrin Hoydal, Lisa Loseto, Nick Lunn,
525 Francoise Messier, Derek C.G. Muir, Martyn Obbard, Filipa Samarra, Ian Stirling, Mitch Taylor and
526 Cortney Watt.

527

528 **10. References**

529 Ackerman, J.T., Eagles-Smith, C.A., Herzog, M.P., Hartman, C.A., Peterson, S.H., Evers, D.C.,
530 Jackson, A.K., Elliott, J.E., Vander Pol, S.S., Bryan, C.E., 2016. Avian mercury exposure and
531 toxicological risk across western North America: a synthesis. *Sci. Total Environ.* 568, 749–769.
532 Agather A.M., Bowman K.L., Lamborg C.H., Hammerschmidt C.R., 2019. Distribution of mercury
533 species in the Western Arctic Ocean (U.S. GEOTRACES GN01). *Mar. Chem.* 216:103686.
534 AMAP, 1998. Arctic Pollution Issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo,
535 Norway.
536 AMAP, 2004. AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic. Arctic
537 Monitoring and Assessment Programme (AMAP), Oslo, Norway.
538 AMAP, 2011. AMAP Assessment 2011: Mercury in the Arctic. Arctic Monitoring and Assessment
539 Programme (AMAP), Oslo, Norway.
540 AMAP, 2015: Human Health in the Arctic. Arctic Monitoring and Assessment Programme (AMAP),
541 Oslo, Norway. vii + 165 pp.
542 AMAP, 2016. AMAP Assessment 2015: Temporal Trends in Persistent Organic Pollutants in the
543 Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway vi+71pp.

544 AMAP, 2017. AMAP Assessment 2016: Chemicals of Emerging Arctic Concern. AMAP, Oslo,
545 Norway. xvi+353pp.

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

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

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

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

559 Andreassen, B., Hoydal, K., Mortensen, R., Erenbjerg S.V., and Dam, M. 2019. AMAP Faroe Islands
560 2013 – 2016: Heavy Metals and POPs Core Programme, Umhvørvisstovan, Argir, Faroe Islands,
561 x + 103 pp (DOI: 10.13140/RG.2.2.36147.37924)

562 Barst BD, Basu N, Chételat J, Dietz R, Eulaers I, Wilson S 2022/This issue. What are the toxicological
563 effects of mercury in Arctic biota? Part 3 Fish and invertebrates. *Science of the Total Environment*
564 (2022/this issue).

565 Bradley M.A., B.D. Barst, N. Basu, 2017. A Review of Mercury Bioavailability in Humans and Fish.
566 *Int J Environ Res Public Health*. 2017;14(2):169. Published 2017 Feb 10.
567 doi:10.3390/ijerph14020169

568 Bowman K.L., Lamborg C.H., Agather A.M. 2020. A global
569 perspective on mercury cycling in the ocean. *Sci Total Environ* 710.
570 <https://doi.org/10.1016/j.scitotenv.2019.136166>

571 Charette, M.A., Kipp, L.E., Jensen, L.T., Dabrowski, J.S., Whitmore, L.M., Fitzsimmons, J.N.,
572 Williford, T., Ulfso, A., Jones, E., Bundy, R.M., Vivancos, S.M., Pahnke, K., John, S.G., Xiang,
573 Y., Hatta, M., Petrova, M.V., Heimbürger-Boavida, L.-E., Bauch, D., Newton, R., Pasqualini, A.,
574 Agather, A.M., Amon, R.M.W., Anderson, R.F., Andersson, P.S., Benner, R., Bowman, K.L.,
575 Edwards, R.L., Gdaniec, S., Gerringa, L.J.A., González, A.G., Granskog, M., Haley, B.,
576 Hammerschmidt, C.R., Hansell, D.A., Henderson, P.B., Kadko, D.C., Kaiser, K., Laan, P., Lam,
577 P.J., Lamborg, C.H., Levier, M., Li, X., Margolin, A.R., Measures, C., Middag, R., Millero, F.J.,
578 Moore, W.S., Paffrath, R., Planquette, H., Rabe, B., Reader, H., Rember, R., Rijkenberg, M.J.A.,
579 Roy-Barman, M., Rutgers van der Loeff, M., Saito, M., Schauer, U., Schlosser, P., Sherrell, R.M.,
580 Shiller, A.M., Slagter, H., Sonke, J.E., Stedmon, C., Woosley, R.J., Valk, O., van Ooijen, J.,
581 Zhang, R., 2020. The Transpolar Drift as a Source of Riverine and Shelf-Derived Trace Elements
582 to the Central Arctic Ocean. *J. Geophys. Res. C: Oceans* e2019JC015920.

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

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

590 Desforges JP, Bandoro C, Shehata L, Sonne C, Dietz R, Puryear WB, Runstadler JA. 2018a.
591 Environmental contaminant mixtures modulate in vitro influenza infection. *Sci Total Environ*
592 634:20-28.

593 Desforges JP, Hall A, McConnell B, Rosing Asvid A, Barber JL, Brownlow A, De Guise S, Eulaers
594 I, Jepson PD, Letcher RJ, Levin M, Ross PS, Samarra F, Víkingsson G, Sonne C, Dietz R. 2018c.
Predicting global killer whale population collapse from PCB pollution. *Science* 361:1373-1376.

- 595 Desforbes JP, Levin M, Jasperse L, De Guise S, Eulaers I, Letcher RJ, Acquarone M, Nordøy E,
596 Folkow LP, Jensen TH, Grøndahl C, Bertelsen MF, St. Leger J, Almunia J, Sonne C, Dietz R.
597 2017. The in vitro effects of polar bear and killer whale blubber-derived contaminant ‘cocktail’
598 mixtures on marine mammal immune cells. *Environ Sci Technol* 51:11431-11439.
- 599 Desforbes JP, Levin M, Jasperse L, De Guise S, Jensen TH, Grøndahl C, Bertelsen MF, Sonne C,
600 Dietz R. 2018b. Immune function in Arctic mammals: Natural killer (NK) cell activity in polar
601 bear, muskox and reindeer. *Vet Immun Immunopathol* 195:72-75.
- 602 Desforbes JPW, Sonne C, Levin M, Siebert U, De Guise S, Dietz R. 2016. Immunotoxic effects of
603 environmental pollutants in marine mammals. *Environ Int* 86:126-139.
- 604 Dietz R, Basu N, Braune B, O’Hara T, Scheuhammer TM, Sonne C, Andersen M, Andreasen C,
605 Andriashek D, Asmund G, Aubail A, Baagøe H, Born EW, Chan HM, Derocher AE, Grandjean
606 P, Knott K, Kirkegaard M, Lunn N, Messier F, Obbard M, Olsen MT, Peacock E, Renzoni A,
607 Rigét F, Skaare JU, Stern G, Stirling I, Taylor M, Wiig Ø, Aars J. 2013. What are the toxicological
608 effects of mercury in Arctic biota? *Sci Total Environ* 443:775-790.
- 609 Dietz R, C Sonne, J-P Desforbes, I Eulaers, A Rosing Asvid, R. Letcher, P. Jepson 2019. Killer
610 whales calls for further protection. *Environment International* 126: 443-444.
611 <https://doi.org/10.1016/j.envint.2019.02.060>
- 612 Dietz R, K Gustavson, C Sonne, J-P Desforbes, FF Rigét, V Pavlova, MA McKinney, RJ. Letcher
613 2015. Physiologically-based pharmacokinetic modelling of immune, reproductive and
614 carcinogenic effects from contaminant exposure in polar bears (*Ursus maritimus*) across the
615 Arctic. *Environmental Research* 140: 45–55.
- 616 Dietz R, Sonne C, Albert C, Bustess JO, Christensen TK, Ciesielski TM, Danielsen J, Dastnai S, Eens
617 M, Erikstad KE, Fort J, Galatius A, Garbus S-E, Gilg O, Hanssen SA, Helander, B, Helberg M,
618 Jaspers VLB, Jenssen BM, Kauhala K, Kyhn, LA, Labansen, AL, Larsen, MM, Lindstøm U,
619 Reiertsen TK, Rigét FF, Roos A, Strand J, Strøm H, Sveegaard S, Søndergaard J, Sun J, Teilmann
620 J, Therkildsen OR, Tjørnløv RS, Wilson S, Eulaers I, 2021. A risk assessment of the effects of
621 mercury on Baltic Sea, Greater North Sea and North Atlantic wildlife, fish and bivalves.
622 *Environmental International* 146, 106178.
- 623 Dietz R, R Letcher, J-P Desforbes, I Eulaers, C Sonne, S Wilson, E Andersen-Ranberg, N Basu, BD
624 Barst, JO Bustnes, J Bytingsvik, TM Ciesielski, PE Drevnick, GW Gabrielsen, A Haarr, K
625 Hylland, BM Jenssen, M Levin, MA McKinney, RD Nørregaard, KE Pedersen, J Provencher, B
626 Styrihave, S Tartu, J Aars, JT Ackerman, A Rosing-Asvid, R Barrett, A Bignert, EW Born, M
627 Branigan, B Braune, CE Bryant, M Dam, CA Eagles-Smith, M Evans, TJ Evans, AT Fisk, M
628 Gamberg, K Gustavson, CA Hartman, B Helander, MP Herzog, PF Hoekstra, M Houde, K Hoydal,
629 AK Jackson, J Kucklickt , E Lie, L Loseto, ML Mallory, C Miljeteig, A Mosbech, DCG Muir, ST
630 Nielsen, E Peacock, S Pedro, SH Peterson, A Polder, FF Rigét, P Roach, H Saunes, M-HS.
631 Sinding, M, JU. Skaare, J Søndergaard, G Stenson, G Stern, G Treu, SS Schuur, G Víkingsson
632 2019a. Current State of Knowledge on Biological Effects from Contaminants on Arctic Wildlife
633 and Fish. *Science of the Total Environment* 696: 133792, 1-39.
- 634 Dietz, R., Sonne, C., Desforbes, J.-P., Eulaers, I., Letcher, R.J. and Jepson, P.D., 2019b. Killer whales
635 call for further protection. *Environment International*, 126:443-444.
- 636 Dietz, R., Riget, F., Born, E.W. 2000a. An assessment of selenium to mercury in Greenland marine
637 animals. *The Science of the Total Environment* 245: 15 – 24.
- 638 Dietz, R. F. Riget and E.W. Born 2000b. Geographical differences of zinc, cadmium, mercury and
639 selenium in polar bears (*Ursus maritimus*) from Greenland. *The Science of the Total Environment*
640 245: 25-48.
- 641 Dietz, R., J. Pacyna, D.J. Thomas, G. Asmund, V. Gordeev, P. Johansen, V. Kimstach, L. Lockhart,
642 S.L. Pfirman, F. F. Riget, G. Shaw, R. Wagemann, M.White 1998. Chapter 7: Heavy metals. In:
643 AMAP Assessment Report: Arctic Pollution Issues. Arctic Monitoring and Assessment
644 Programme. Oslo, Norway: 373-524.
- 645 Evers, D.C., S. Keane, D. Buck and N. Basu. 2016. Evaluating the Effectiveness of the Minamata
646 Convention on Mercury: Principles and Recommendations for Next Steps. *Science of the Total*

647 Environment 569, 888-903.

648 Kim J, Soerensen AL, Kim MS, Eom S, Rhee TS, Jin YK, et al. 2020. Methylmercury in the East
649 Siberian Sea: The importance of sediment sources. *Environ Sci Technol.* 54(16):9949-57.

650 Laidre, K. L., H. Stern, K. M. Kovacs, L. Lowry, S. E. Moore, E. V Regehr, S. H. Ferguson, et al.
651 2015. Arctic marine mammal population status, sea ice habitat loss, and conservation
652 recommendations for the 21st century. *Conservation Biology* 00:1–14.

653 Letcher RJ, Bustnes JO, Dietz R, Jenssen BM, Jørgensen EH, Sonne C, Verreault J, Vijayan MM,
654 Gabrielsen GW. 2010. Effects Assessment of Persistent Organohalogen Contaminants in Arctic
655 Wildlife and Fish. *Sci Total Environ* 408:2995-3043.

656 Lippold, A, Aars, J, Andersen, M, Aubail, A, Derocher, A, Dietz, R, Eulaers, I, Sonne, C, Welker, J,
657 Wiig, Ø; Routti, H 2020. Two Decades of Mercury Concentrations in Barents Sea Polar Bears
658 (*Ursus maritimus*) in Relation to Dietary Carbon, Sulfur, and Nitrogen. *Environmental Science &*
659 *Technology* 54, 7388-7397.

660 Lippold, A, Aars, J, Andersen, M, Blanchet M-A, A, Dietz, R, Eulaers, I, Welker, J, Routti, H
661 submitted. Spatial Variation in Mercury Concentrations in Polar Bear (*Ursus maritimus*) Hair from
662 the Norwegian and Russian Arctic. *Science of the Total Environment* re-submitted.

663 Morris, AD., Riget, F.R., Wilson, S., Fryer, R., et al. 2022/This issue. Temporal trends of mercury in
664 Arctic biota: 10 more years of progress in Arctic monitoring. *Science of the Total Environment*
665 (this issue).

666 Nyman M., Bergknut M., Fant M.L., Raunio H., Jestoi M., Bengs C., Murk A., Koistinen J., Backman
667 C., Pelkonen O., Tysklind M., Hirvi T., Helle E. 2003. Contaminant exposure and effects in Baltic
668 ringed and grey seals as assessed by biomarkers. *Marine Environmental Research* 55:73-99.

669 Outridge PM, Dietz R, Amyot M, Barkay T, Basu N, Berg T, Braune B, Carrie J, Chételat J, Cole A,
670 Constant P, Dastoor A, Dommergue A, Donaldson SG, Douglas T, Durnford D, Evans M, Ferrari
671 C, Gaden A, Gantner AK, Gantner N, Goodsite M, Hedman J, Hintelmann H, Hobson K, Johnson
672 M, Kirk J, Kroer N, Krümmel E, Larose C, Lean D, Leech T, Letcher RJ, Loseto L, Macdonald
673 RW, Muir DCG, J. Munthe J, Nielsen TG, O’Hara T, Pacyna J, Poissant L, Poulain A, Rigét F,
674 Rognerud S, Ryzhkov A, Scheuhammer T, Skov H, Sonne C, Sørensen S, Steenhuisen F, Steffen
675 A, Stern G, Stow J, Sundseth K, Travnikov O, Verta M, Wang F, Wängberg I, Wilson SJ,
676 Zdanowicz C, 2011a. Mercury in the Arctic. Arctic Monitoring and Assessment Programme
677 (AMAP), Oslo, Norway. xiv + 193 pp.

678 Pagano AM, Durner GM, Rode KD, Atwood TC, Atkinson SN, Peacock E, Costa DP, Owen MA,
679 Williams TM. 2018. High-energy, high-fat lifestyle challenges an Arctic apex predator, the polar
680 bear. *Science.* 2018 Feb 2;359 (6375):568-572. Petrova MV, Ourgaud M, Boavida JRH, Dufour
681 A, Tesán Onrubia JA, Lozingot A, Heimbürger-Boavida L-E 2020a. Human mercury exposure
682 levels and fish consumption at the French Riviera. *Chemosphere* 258:127232.

683 Petrova MV, Krisch S, Lodeiro P, Valk O, Dufoura A, Rijkenberg MJA, Achterberg EP, Rabe B, van
684 der Loeff MR, Hamelin B, Sonke JE, Garnier C, Heimbürger-Boavida L-E 2020b. Mercury species
685 export from the Arctic to the Atlantic Ocean. *Marine Chemistry* 225, 103855.

686 Rigét FF, Muir D, Kwan M, Savinova T, Nyman M, Woshner V, O’Hara T 2005. Circumpolar pattern
687 of mercury and cadmium in ringed seals. *Science of the Total Environment* 351–352 (2005) 312–
688 322.

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

691 Routti H, Atwood T, Bechshøft T, Boltunov A, Ciesielski T, Desforges JP, Dietz R, Gabrielsen GW,
692 Jenssen BM, Letcher RJ, McKinney M, Morris A, Rigét FF, Sonne C, Styrihave B, Tartu S 2019
693 State of knowledge on current exposure, fate and potential health effects of contaminants in polar
694 bears from the circumpolar Arctic. *Sci Total Environ* 664:1063-1083.

695 Routti H, Letcher RJ, Born EW, Branigan M, Dietz R, Evans TJ, Fisk AT, Peacock E, Sonne C 2011.
696 Spatio-temporal trends of selected trace elements in liver tissue from polar bears (*Ursus*
697 *maritimus*) from Alaska, Canada and Greenland. *J Environ Monitor* 13: 2260-2267.

- 698 Routti H, Lille-Langøy R, Berg MK, Fink T, Harju M, Kristiansen K, Rostkowski P, Rusten M, Sylte
699 I, Øygarden L, Goksøyr A. 2016. Environmental chemicals modulate polar bear (*Ursus maritimus*)
700 peroxisome proliferator-activated receptor gamma (PPARG) and adipogenesis in vitro. Environ
701 Sci Technol 50:10708-10720.
- 702 Routti, H, Letcher, RJ, Born, EW, Branigan, M; Dietz R, Evans TJ, McKinney MA, Peacock, E,
703 Sonne C 2012. Influence of carbon and lipid sources on variation of mercury and other trace
704 elements in polar bears (*Ursus maritimus*). Environmental Toxicology and Chemistry 31, 12,
705 2739–2747.
- 706 Routti H., Arukwe A., Jenssen B.M., Letcher R.J., Nyman M., Bäckman C., Gabrielsen G.W. 2010.
707 Comparative endocrine disruptive effects of contaminants in ringed seals (*Phoca hispida*) from
708 Svalbard and the Baltic Sea. Comparative biochemistry and physiology Part C 152:306–312.
- 709 Tesán Onrubia, J.A., Petrova, M.V., Puigcorbé, V., Black, E.E., Valk, O., Dufour, A., Hamelin, B.,
710 Buessler, K.O., Masqué, P., Le Moigne, F.A.C., Sonke, J.E., Rutgers van der Loeff, M.,
711 Heimbürger-Boavida, L.-E., 2020. Mercury Export Flux in the Arctic Ocean Estimated from 234
712 Th/ 238 U Disequilibria. ACS Earth and Space Chemistry acsearthspacechem.0c00055.
- 713 Wang, K., Munson, K.M., Beaupré-Laperrière, A., Mucci, A., Macdonald, R.W., Wang, F., 2018.
714 Subsurface seawater methylmercury maximum explains biotic mercury concentrations in the
715 Canadian Arctic. Scientific Reports 8.
- 716 Wang YM, Li LJ, Yao C, Tian XS, Wu YR, Xie Q, Wang DY. 2021. Mercury in human hair and its
717 implications for health investigation. Current Opinion in Environmental Science and Health
718 22:100271.
- 719 Wobeser, G., Nielsen, N.O., Schiefer, B., 1976. Mercury and mink II. Experimental methyl mercury
720 intoxication. Can. J. Comp. Med. 40, 34–45.
- 721 Wren, C.D., Hunter, D.B., Leatherland, J.F., Stokes, P.M., 1987. The effects of polychlorinated
722 biphenyls and methylmercury, singly and in combination, on mink. I: uptake and toxic responses.
723 Arch. Environ. Contam. Toxicol. 16, 441–447.