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A risk assessment of the effects of mercury on Baltic Sea, Greater North Sea and North Atlantic wildlife, fish and bivalves

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ABSTRACT

A wide range of species, including marine mammals, seabirds, birds of prey, fish and bivalves, were investigated for potential population health risks resulting from contemporary (post 2000) mercury (Hg) exposure, using novel risk thresholds based on literature and *de novo* contamination data. The main geographic focus is on the Baltic Sea, while data from the same species in adjacent waters, such as the Greater North Sea and North Atlantic, were included for comparative purposes. For marine mammals, 23% of the groups, each composing individuals of a specific sex and maturity from the same species in a specific study region, showed Hg-concentrations within the High Risk Category (HRC) and Severe Risk Category (SRC). The corresponding percentages for seabirds, fish and bivalves were 2.7%, 25% and 8.0%, respectively, although fish and bivalves were not represented in the SRC.

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Juveniles from all species showed to be at no or low risk. In comparison to the same species in the adjacent waters, i.e. the Greater North Sea and the North Atlantic, the estimated risk for Baltic populations is not considerably higher. These findings suggest that over the past few decades the Baltic Sea has improved considerably with respect to presenting Hg exposure to its local species, while it does still carry a legacy of elevated Hg levels resulting from high neighbouring industrial and agricultural activity and slow water turnover regime.

1. Introduction

Contaminant studies have been conducted across the world in many different ecosystems and species, and understanding the health risk associated to the observed contaminant bioaccumulation remains a warranted task. Only few studies have undertaken large-scale evaluations using Risk Categories for bioaccumulation of contaminants, including mercury (Hg). These studies include North American birds (Ackerman et al., 2016), white-tailed eagles (*Haliaeetus albicilla*) in the Baltic, Norway and West Greenland (Sun et al., 2019) as well as other wildlife in the Arctic (Dietz et al., 2013, 2018, 2019). However, there is a lack of efforts simultaneously addressing multiple functional groups and trophic levels, composing marine mammals, seabirds, birds of prey, fish and invertebrates, and a holistic food web evaluation of health risks associated with Hg contamination remains to be endeavoured.

The Baltic Sea is among the most polluted ecosystems in the world, known for presenting its food web to very high concentrations of Hg and organic contaminants. This high contamination has been associated with detrimental effects on seals in terms of impaired reproduction and histopathological damage, leading to severe population impacts (Bergman, 2007; Bergman and Olsson, 1985; Blomkvist et al., 1992; Harding et al., 2007; Helle et al., 1976a,b; Olsson et al., 1975; Routti et al., 2005, 2008, 2009). In addition, in seabirds, birds of prey and fish, a plethora of harmful health effects has been reported (Gercken et al., 2006; Skarphedinsdottir et al., 2010; Bignert & Helander 2015, Helander et al., 1982, 2002, 2008). Although efforts have been made to quantify population level effects following reports of multiple health effects on Baltic sentinel species, there is a grave lack in efforts to quantify risks of population effects in fish and invertebrates, such as bivalves (Korsman et al. 2012; Roos et al., 2012; Helander et al., 2008; Siebert et al., 2006, 2007; HELCOM, 2010, 2018). Establishing links between contaminant bioaccumulation and health outcome is a difficult task, but an important one to manage and conserve fish stocks and wildlife populations, and the marine ecosystems they build up (Rodriguez-Estival and Mateo, 2019).

The aim of the present study is to use contemporary (post-2000) data on Hg concentrations in a large diversity of species groups, , and conduct a holistic risk assessment of Hg bioaccumulation on the Baltic Sea food web groupings using established and novel risk thresholds. Doing so we also provide a comparison to the same species in adjacent waters, i.e. the Greater North Sea and North Atlantic. This is the first time such a large-scale effort on this region and for species ranging from marine mammals, birds down to fish and bivalves is being performed. We also discuss the limitations of the current risk assessment and potential for improving future risk assessments.

2. Materials and methods

2.1. Study design

We reviewed the existing literature for contemporary (post-2000) Hg concentrations in marine mammals, seabirds, birds of prey, fish and bivalves and adding recent unpublished data from BALTHEALTH and ARCTOX from the Baltic Sea, Greater North Sea and North Atlantic and made a risk evaluation based on existing effect thresholds (SI Tables 1-4). An exhaustive formal risk assessment would ideally have included MeHg, inorganic mercury and selenium, However, as these data were not available from most of the datasets included here, a geographical or species related comparison could not be conducted. When possible, we extracted raw data or obtained data by contacting the authors. In addition, we conducted Hg analyses on key knowledge gaps (see below for further details). Furthermore, we retrieved data on fish and bivalve exposure from the ICES (ICES Data Centre, 2019) and Swedish EPA databases (Swedish EPA, 2019) for the following ICES ecoregions: Greenland Sea, Norwegian Sea, Barents Sea, Icelandic Waters, Faroes Waters, Greater North Sea and Baltic Sea. With the focus of the present study on the Baltic Sea, we defined the region in four study basins: Gulf of Bothnia, Gulf of Finland, Baltic Proper and Danish Straits (SI Fig. 5). The obtained raw data was harmonised to wet weight concentrations (μg g^{-1}) using the reported concurrent dry matter (DM) percentages or using a reported one for the same or similar species. For seabird blood, we used DM = 21.9% (Eulaers et al. Pers. Comm.), for Common guillemot and European herring gull egg, we used DM = 20.8% as reported by Eagles-Smith et al. (2008).

A range of marine mammal, seabird, birds of prey fish, and bivalve species from different study basins were analysed for hepatic, blood, body feathers and eggs, muscle and soft tissue Hg content, respectively (SI Tables 1-4). Samples from harbour (*Phoca vitulina*) and grey (*Halichoerus grypus*) seals from the Danish Straits were obtained from seals regulated in relation to stationary fishing gear, from seals by-caught in fishing gear, and from seals found newly stranded along the Danish coastline. Samples from grey and ringed (*Pusa hispida*) seals from the Gulf of Bothnia were collected during regular hunt or from seals by-caught in fishing gear. Samples from harbour porpoises (*Phocoena phocoena*) from the Danish Straits were collected from porpoises by-caught in fishing gear or found newly stranded along the Danish coastline.

2.2. Mercury analysis and quality control

We refer to the peer-reviewed articles (SI Tables 1-4) for the

Table 1
Estimated Risk Categories for health effects in wildlife and human consumption (bivalves) owing to Hg exposure. Detailed information regarding the calculations and assumptions are provided in the Materials and Methods section.

		No risk	Low risk	Moderate risk	High risk	Severe risk	Reference
Marine mammal	Liver (μg/g)	<16.00	16.0–64.0	64.0–83.0	83.0–123.0	≥123.0	Ronald et al. 1977
Seabird	Egg (μg/g)	<0.11	0.11–0.47	0.47–1.30	1.30–1.70	≥1.70	Ackermann et al. 2016
	Blood equivalent (μg/g)	<0.20	0.20–1.00	1.00–3.00	3.00–4.00	≥4.00	Ackermann et al. 2016
	Body feather (μg/g)	<1.58	0.58–7.92	7.92–23.8	23.8–31.7	≥31.7	Ackermann et al. 2016
Bird of prey	Body feather (μg/g)	<1.58	0.58–7.92	7.92–23.8	23.8–31.7	≥31.7	Ackermann et al. 2016
Fish	Muscle (μg/g)	<0.10	0.10–0.30	0.30–0.50	0.50–2.00	≥2.00	Dillon et al. 2010
Bivalve*	Soft tissue (μg/g)	<0.01	0.01–0.05	0.05–0.15	0.15–0.40	≥0.40	SFT 1997

* Note that the risk categories were estimated for human consumption as no risk data for bivalve exposure exists.

respective analytical methods used for the published data contributing to the present paper. Additional analyses on mercury were performed at the accredited Trace Element Lab of the Aarhus University (Denmark) as well as at the Institute Littoral, Environment and Societies (LIENSs, France). Briefly, total mercury analyses (referred to as Hg throughout this article) were performed on dried tissue using a Direct Mercury Analyser 80 (Milestone, Italy) or an Altec Advanced Mercury Analyser 254 (Altec, Czech Republic) following the USEPA Method 7473 (USEPA, 1998).

The instrumental analytical quality control conducted at the Trace Element Lab of the Department of Bioscience, Aarhus University, Denmark. was verified by analysing procedural blanks, duplicates, aqueous standards (10 ng and 100 ng Hg, prepared from 1000 ± 4 mg L^{-1} stock solution, Sigma-Aldrich, Switzerland), and Certified Reference Material (CRM; DORM-4, National Research Council, Ottawa, Canada). Procedural blanks and CRMs were analysed concurrently every 10 samples. All samples and CRMs were corrected for the average blank amount of Hg (0.07 ± 0.13 ng; n = 131) as well as for the recovery of aqueous standards (108.6 ± 1.3%; n = 21). The measured recovery percentage of the CRMs fell within the acceptable range (105.9 ± 2.3%; n = 52) of the certified value (0.410 ± 0.055 μg g − 1 dry weight). Relative percent difference for duplicate samples ranged from 0.02% to 34.08% (n = 13). As for the analyses conducted at LIENSs, each Hg analysis were repeated two or three times for each sample until the relative standard deviation for the aliquots was < 10%. Samples not meeting this criterion were excluded from the analysis. The mean Hg concentrations for those two measurements were then considered. To ensure the accuracy of measurements, a certified reference material (CRM) was used (Lobster Hepatopancreas Tort-2; NRC, Canada; Hg concentration of 0.27 ± 0.06 mg g^{-1} of dry weight (dw)). The CRM was measured every 10 samples and the average measured value was 0.26 ± 0.01 mg/g dw (n = 113). Additionally, blanks were run at the beginning of each sample set. The detection limit of the method was 0.05 ng of Hg. Further details on the analytical procedure as well as the quality assurance are provided in detail by Sun et al. (2019), Ma et al. (2020), Bustamante et al. (2006) and Fort et al. (2016). The QA/QC for all the employed data of the cited peer-reviewed publications as well as the databases from ICES and the Swedish EPA are provided in the cited articles as well as at the from ICES databases (ICES 2004; ICES Data Centre 2019) and the data from the Swedish EPA are accredited by SWEDAC (Swedish EPA 2020; SWEDAC).

2.3. Risk analysis

We conducted for the first time a risk analysis for potential Hg-associated health effects for marine mammals, seabirds and birds of prey, fish and bivalves in the Baltic, Greater North Sea and North Atlantic. We used five risk thresholds, resulting in five Risk Categories (RCs), i.e. No Risk Category (NRC), Low Risk Category (LRC), Moderate Risk Category (MRC), High Risk Category (HRC), and Severe Risk Category (SRC; Table 1). These categories reflect to which degree measured total Hg concentrations exceed effect threshold concentrations for adverse effects on reproduction, physiology, condition and behaviour. Methylmercury (MeHg) is the most toxic form of Hg. Previous studies demonstrated that Hg in blood, muscle, feather and egg is > 90% MeHg and thus total Hg is considered as a good proxy of MeHg concentrations and toxicity in these tissues (e.g. Dietz et al. 1990; Bond & Diamond 2009; Renedo et al., 2017). Conversely, Hg is mostly in the form of inorganic Hg (iHg) in liver (e.g. Wagemann et al. 1998). It has previously been demonstrated that total Hg concentrations in liver and muscle are significantly correlated, demonstrating that total Hg in liver can be used to assess animal exposure to MeHg and its toxicity. Since total Hg measurements, as apposed for MeHg for which only very few data in tissues is available (Ackerman et al. 2016), have been routinely used to investigate Hg exposure and effects and has led to the established risk thresholds, the current risk assessment focuses on total Hg data only.

The ICES monitoring programme has since 2012 started to monitor MeHg but only on blue mussels. In numbers these analyses so far only represents 7.6% of the total Hg analyses. The corresponding figures from the IVL database is in the same magnitude but these analyses are not accredited. For marine mammals, the hepatic Hg thresholds defined by Ronald et al. (1977) and Dietz et al. (2019) were used, while for seabirds the assessment methodology introduced by Ackerman et al. (2016) was adapted for egg, liver, body feathers, and blood concentrations. For fish, a system comprising five risk categories was established based on expert knowledge (Benjamin Barst Pers. Comm.; Nil Basu Pers. Comm. From Ongoing AMAP Assessment) and two key papers to convert whole body Hg concentrations to muscle Hg concentrations (Dillon et al., 2010; Peterson et al., 2004). With respect to bivalves, an Environmental Quality Standard (EQS) was developed within the EU Common Implementation Strategy for the Water Framework Directive (EU, 2005), and was set at 0.02 μg g^{-1} ww for protection of fish-eating top predators for secondary poisoning of Hg. It is therefore important to note that this threshold does not indicate risk for bivalves but rather to wildlife consuming bivalves, as no risk data for bivalve Hg contamination currently exists. As such, the NRC was based on measured concentrations at < 0.20 μg g^{-1} dw distant from known sources, and it was translated to the EQS by applying a general dry matter percentage of 10%. This EQS was further extrapolated to a system of five categories of increasing severity of risk using the approach developed by Statens Forurensnings Tilsyn (SFT, 1997). The remaining four categories were calculated using a factor of 10 for conversion, resulting in the lower threshold of the SRC to be at 0.40 μg g^{-1} ww, which is close to the EU food safety limit (EU, 2006) of 0.50 μg g^{-1} ww. Altogether, the proposed RCs for bivalves were defined both using expert opinion and empiric monitoring measurements.

3. Results and discussion

3.1. Marine mammals

For the Baltic Sea, Greater North Sea and North Atlantic, eight out of 35 (23%) marine mammal groups were within the two highest RCs, i.e. the SRC and the HRC. In an earlier study of Arctic marine mammals it has been shown that 23 out of 69 (33%) of the marine mammal groups were within these two RCs (Dietz et al., 2019). It should, however, be noted that the relative high occurrence of these two RCs for Arctic marine mammal species is linked to apex predator species such as polar bear (*Ursus maritimus*) and killer whale (*Orcinus orca*). Nine out of 34 (26%) of the presented groups of marine mammals must be regarded as quite highly at risk (Fig. 4; SI Table 4, SI Fig. 4).

3.1.1. Grey seal

Among marine mammals, most data were available from the increasing grey seal populations (Hårding and Härkönen, 1999). Hg concentrations in grey seals were generally in the same order of magnitude as found for harbour and ringed seals, though maximum values were lower in the latter two (Fig. 1; SI Fig. 1; SI Table 1). Adult females from the Gulf of Bothnia showed the highest concentrations with 23.5% in the NRC and 29.4% in the SRC. Males and subadults from the same region were lower contaminated, with none above the LRC. Likewise were all yearlings within the NRC. Baltic Proper grey seals showed slightly higher concentrations than those in the Gulf of Bothnia, although a comparison for females was not possible at this point. Nevertheless, RC distribution for subadult and yearling Baltic Proper seals is similar to that of the Gulf of Bothnia. In contrast, adult males from the Baltic Proper occupied all RCs and even up to 11.1% fell within both the HRC and SRC. Finally, adult male and subadult grey seals from the Danish Straits showed similar concentrations as those in the Gulf of Bothnia. No data for yearlings or adult females are available at this point. In this region, most of the adult males and subadults reside in the NRC (50 and 75%, respectively), while some individuals of both groups

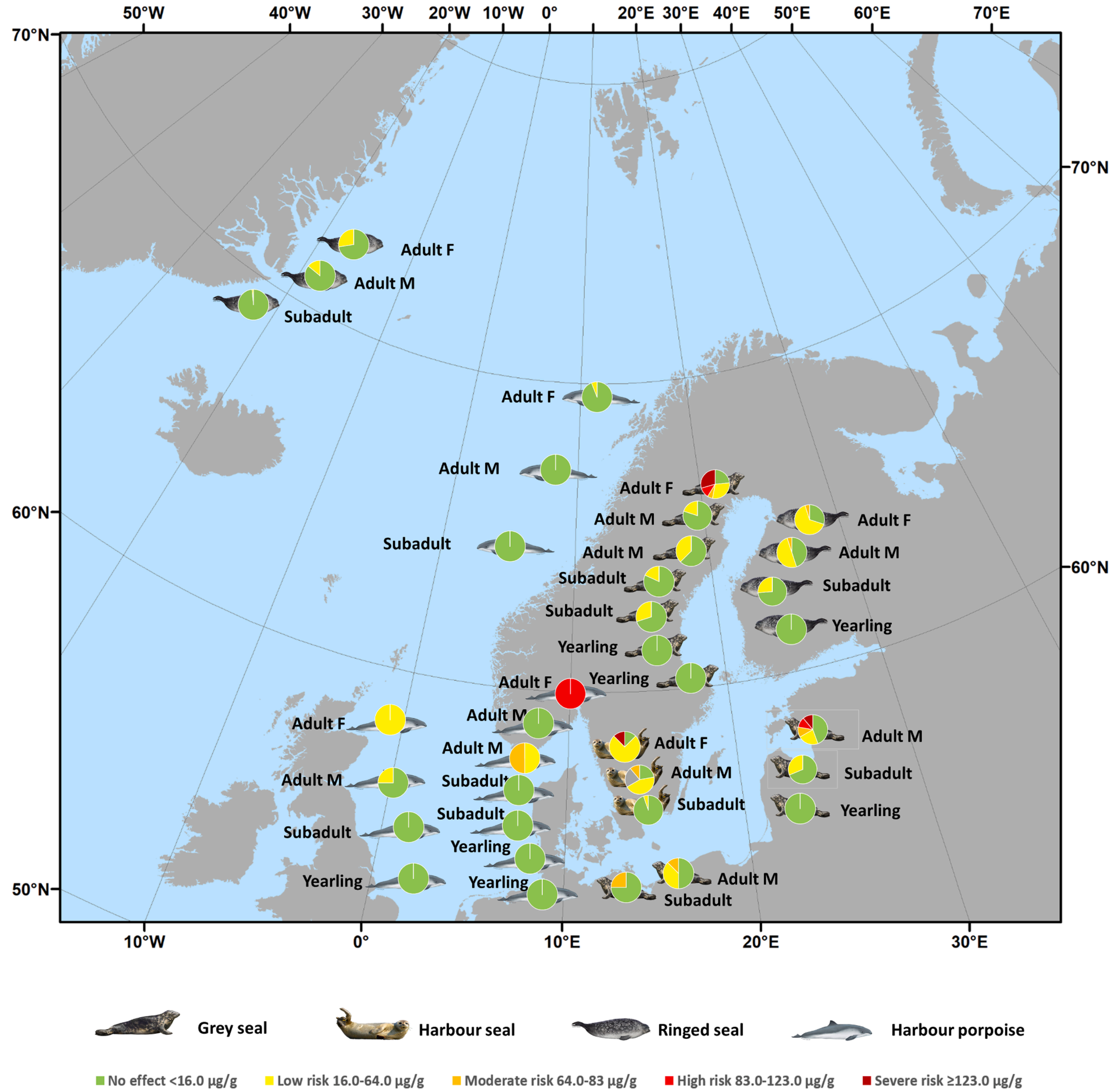


Fig. 1. Geographical overview of the proportion of individuals of a specific marine mammal group, i.e. individuals of a specific maturity in a specific location, at risk of health effects due to contemporary (post-2000) Hg exposure in liver tissue. See SI Table 1 for detailed exposure and Risk Category data, and see SI Fig. 1 for a ranked histogram.

do fall within the HRC (12.5 and 25%, respectively). Unfortunately, no data were available for grey seal Hg concentrations in the Greater North Sea or the North Atlantic, and a comparison in potential health risk was therefore not possible.

3.1.2. Harbour seal

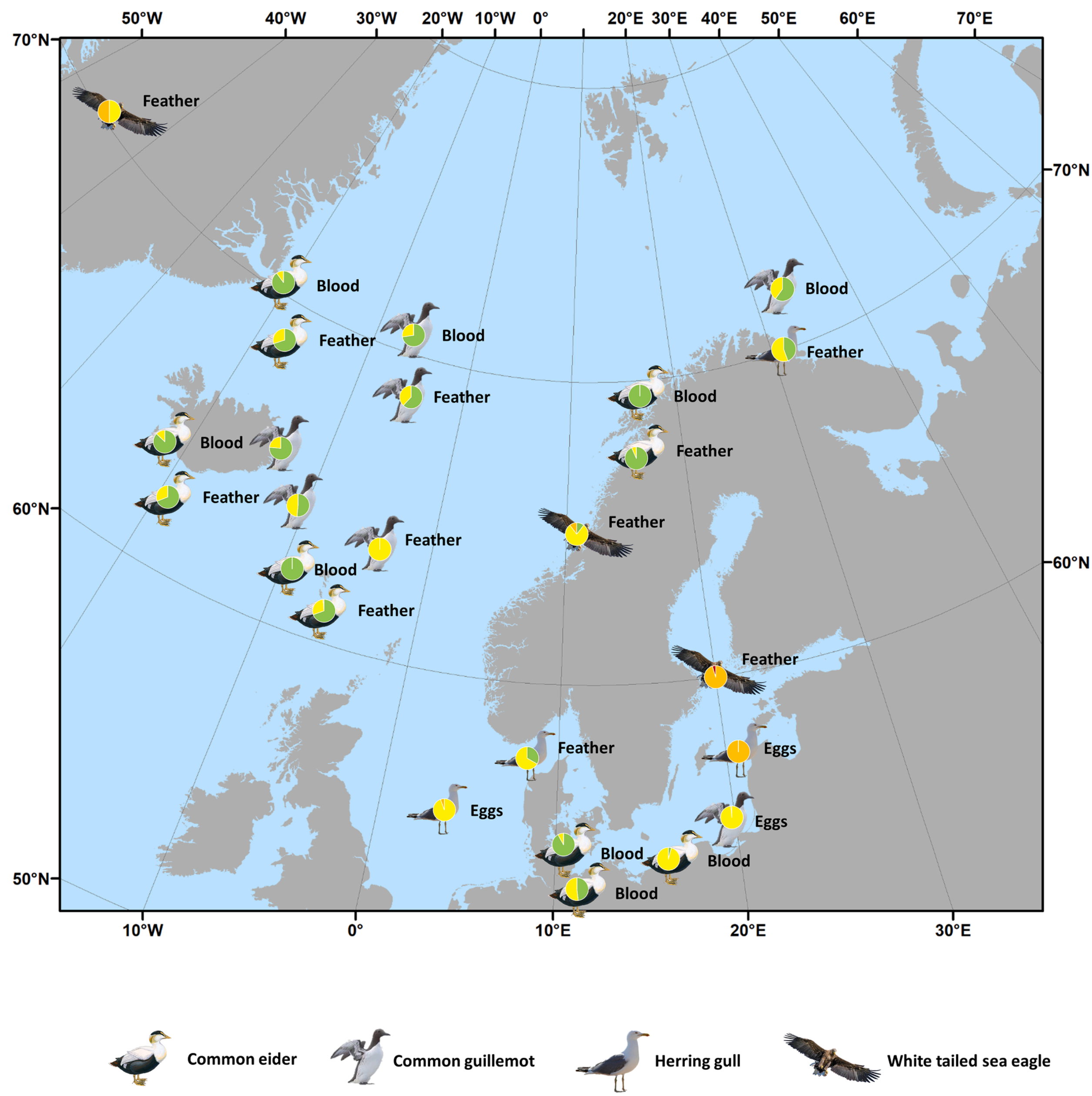
Harbour seal data were only available from the Danish Straits as very few harbour seals (n = 588) inhabit the remaining Baltic Sea regions (Härkönen and Isakson, 2010). Similarly to grey seals, the majority of Hg concentrations in harbour seals were within the NRC and the LRC (Fig. 1; SI Fig. 1; SI Table 1). Up to 75.0% of adult females were within the LRC while only 5.3% of the subadults were. As much as 12.5% of adult females fell within the SRC, however, up to 77.8% of the adult male seals had concentrations associated with a health risk (MRC: 22.2%, HRC: 11.1%). It can be assumed that the individuals in the two highest RCs are most likely old individuals with a substantial lifetime bioaccumulation thus carrying legacy exposure from before the turn of the millennium. Unfortunately, no data were available for harbour seal Hg concentrations in the Greater North Sea or the North Atlantic, and a comparison in potential health risk was therefore not possible.

3.1.3. Ringed seal

Ringed seal Hg concentrations in the Gulf of Bothnia generally fell within the NRC and the LRC (Fig. 1; SI Fig. 1; SI Table 1). All yearlings were likely free of Hg associated health effects while Hg concentrations in subadults were higher and resulted in 26.3% of these individuals to potentially be at low risk. All adults were considerably higher exposed than the subadults and yearlings, and showed 30 and 45%, respectively, for females and males to be at no risk. Females and males showed similar concentrations resulting in 65.0 and 50.0% of the individuals to fall within the LRC, respectively, while each group showed 5% of individuals to be at moderate and high risk. Ringed seals from East Greenland were slightly lower in Hg concentrations than those from the Baltic Sea, resulting in populations being less at risk, with subadult seals having 98.0% of the population at no risk and only 2.0% falling within the LRC. Hg concentrations in the adult East Greenland seals were higher than in the subadult ones, with concentrations being approximately half those found in adult ringed seals in the Gulf of Bothnia. Hence, adult female East Greenland ringed seals was mostly (72.5%) within the NRC while the remaining 27.5% were at low risk. Similarly, the majority of adult Greenland male ringed seals were within the NRC (86.0%) and the remainder within the LRC (14.0%). These proportions were much lower than observed for the Gulf of Bothnia, and, in contrast, moderate or high risk can only be expected in the latter. It should be noted that the spatial differences between East Greenland and Gulf of Bothnia is less pronounced for Hg as compared to other chemical contaminants (e.g. ∑PCBs; Bjurlid et al., 2018; Dietz et al., 2019).

3.1.4. Harbour porpoises

Data on Hg concentrations in harbour porpoises from the Baltic Proper are scarce since this population is very small (n = 500 individuals; SAMBAH, 2016). Thus, in the present study, Hg



	No risk	Low risk	Moderate risk	High risk	Severe risk
Egg (µg/g)	<0.11	0.11–0.47	0.47–1.30	1.30–1.70	≥1.70
Blood (µg/g)	<0.20	0.20–1.00	1.00–3.00	3.00–4.00	≥4.00
Feather (µg/g)	<1.58	0.58-7.92	7.92-23.75	23.79-31.67	≥31.67

Fig. 2. Geographical overview of the proportion of individuals of specific seabirds and birds of prey mammal populations present in the Baltic that are at risk of Hg-mediated health effects extrapolated from blood, feather or egg Hg concentrations; based on post-2000 monitoring data grouped according to sex and maturity where possible. See SI Table 2 for the detailed information upon which this summary graphic is based and a ranked histogram on the same data in SI Fig. 2.

concentrations in harbour porpoises from only the Danish Straits were used to assess the risk of this species to Hg. An adult female harbour porpoise from 1998 had the highest Hg concentration while concentrations in two adult males from the same year were an order of magnitude lower. As expected, Hg concentrations in subadults were lower than those in adults, and concentrations in yearlings were even lower. As for the risk categories, all yearlings, subadults and adult males from 1998 fell within the NRC, while the adult female from 1998 with the highest concentration fell in the HRC. The remaining were categorised with low or moderate risk, while no harbour porpoise was in the severe risk category. Since we were not able to retrieve post-2000 data on Hg-levels from Harbour porpoises from the North Sea, data from 1998 to 1999 were used for comparison. All individuals from the Greater North Sea or Norwegian coast fell within the NRC, with the exception of the adults for which females showed the highest concentrations. Nonetheless, none of these experienced moderate or higher risk of Hg bioaccumulation associated health effects.

3.2. Seabirds and birds of prey

In order to perform a risk assessment for seabirds and birds of prey we focussed on four species, i.e. common eider (*Somateria mollissima*), common guillemot (*Uria aalge*), European herring gull (*Larus argentatus*), and white-tailed eagle (*Haliaeetus albicilla*; Fig. 2; SI Table 2; SI Fig. 2). Only one out of 21 groups (4.8%), i.e. body feathers from adult white-tailed eagles from the Baltic Proper) showed individuals with concentrations within the SRC, while the majority of the remaining bird groups fell within the MRC (Fig. 2, SI Table 2; SI Fig. 2). Similar risk grouping has been demonstrated for North American and Arctic birds (Ackerman et al., 2016; Dietz et al., 2019). In the extensive work by Ackerman et al. (2016), 30 out of 69 groups (43.5%) contained individuals within the HRC and SRC. Despite being more remote from anthropogenic Hg sources, seven out of 53 Arctic species groups (13.2%) showed to contain individuals within these two RCs, which is in high contrast to the present study (Dietz et al., 2019). It is unlikely that these regional variations are explained by differences in the trophic level of study birds only. Indeed, the present study focused on low, intermediate and high trophic level species feeding on bivalves up to predatory fish, and we did not observe any major variations in the risk categories between species. These results thus suggest lower Hg contaminations and associated risks for seabirds in the Baltic Sea, the Greater North Sea and the northeast Atlantic when compared to Arctic and North American regions. At smaller spatial scale, we also found regional differences within the regions investigated in this study. Indeed, seabirds from the Baltic Sea were found in higher proportion in the LRC and MRC (mean: 75.1%) than those from the Greater North Sea and northeast Atlantic (mean: 46.3%) suggesting higher Hg contamination and associated health risk in the Baltic Sea (see SI Table S2).

3.2.1. Common eider

An assessment using blood equivalent risk thresholds showed a clear West-East gradient among Baltic Sea colonies with increasing concentrations from the two Danish Straits colonies at Hov Røn and Agersø to the Baltic Proper colony at Christiansø (Fig. 2; SI Table 2; SI Fig. 2). This concentration gradient results in individuals at Hov Røn to be 92.0% within the NRC (only 8.0% within the LRC), those at Agersø to be 48.3% within the NRC (up to 51.7% within the LRC), while at Christiansø only 4.3% of the individuals fall within the NRC and as much as 95.7% fall within the LRC. Seabird contamination in the Baltic Sea generally contrasts with those found in North Atlantic colonies, such as those at Tromsø, Faroe Islands and Ittoqqortoormiit in East Greenland, where the majority (>90.0%) of the individuals falls within the NRC, if not completely (at Tromsø and Faroe Islands) (Fig. 2; SI Table 2; SI Fig. 2). Moreover, body feather-based RCs for the same North Atlantic populations show similar conclusions, with the exception that 1.4% of Faroese eiders may also be at moderate risk and that overall a lower proportion (69.4–93.3%) of eiders is at no suspected risk. Body feather Hg data for the Baltic Sea colonies was not available at this point.

3.2.2. Common guillemot

While no blood equivalent RCs can be constructed for Baltic Sea common guillemots, post-2000 egg data ($n = 160$) was available for the Baltic Proper (Fig. 2; SI Table 2; SI Fig. 2). Egg Hg concentrations reflect the females' short-term dietary exposure prior to egg laying in this income breeder species, and may be regarded as a health risk assessment for foetal exposure (Ackerman et al. 2016). Most eggs concentrations fell within the LRC (98.1%), and the remaining 1.9% belonged to the MRC. While none of the measured eggs signified no risk for foetus, there was also a general lack of individuals in the higher RCs, likely due to the overall decrease in Hg contamination observed in common guillemot within the Baltic Proper (Bignert and Helander, 2015). These declines corresponded well with the declines in Hg in body feathers of white-tailed eagles (Sun et al., 2019) and are in agreement with observations made by Rigét et al. (2011) for decreasing Hg time trends towards the Scandinavian regions. Here, we were able to make a geographic comparison with some North Atlantic colonies, such as those at Hornøya and Jan Mayen in Norway and the one at the Faroe Islands (Fig. 2; SI Table 2; SI Fig. 2). In sharp contrast to the Baltic Sea egg RC assessment, the RC for the North Atlantic colonies were based on blood equivalents or body feathers and did not show any presence above the LRC, but rather the majority of adults to reside within the NRC (60.0–72.5%). For these North Atlantic colonies, no egg concentrations was available, and hence a comparison on foetal risk was not possible. Nevertheless, the high percentage of common guillemots from the Baltic Sea in the LRC was similar to Hg concentrations and associated risk for adult birds from the Faroe Islands (SI Table 2), whereas the remaining populations showed variable percentages within the No risk and the Low risk categories (Fig. 2; SI Table 2, SI Fig. 2).

3.2.3. European herring gull

Like for the common guillemot, Hg data for the Baltic Sea were collected at the Baltic Proper only and were restricted to egg concentrations. Based on these, it was clear that the foetal risk fell completely within the LRC, similarly as in the Greater North Sea (LRC: 94.4%). In the latter, also all individuals were above the threshold at which risks are expected and, moreover, a small proportion of the population was at moderate risk (5.6%). Within that same Greater North Sea region, body feather-based RCs for a colony at Vest-Agder showed 66.6% fall within the LRC and the remaining 33.3% to be at no risk, indicating that there potentially was large spatial variability among herring gull colonies. Finally, body feather concentrations from a herring gull colony close to the above-mentioned common guillemot colony at Hornøya seemed to confirm a similar RC profile for the same location (NRC: 44.4% and LRC: 55.6%). This profile also again confirmed that inter-colony differences may correspond to contrasting on abiotic and biotic pathways, rather than to a simple gradient of decreasing Hg concentrations northwards, as was seemingly indicated by the common eider spatial variations.

3.2.4. White-tailed eagle

A clear geographical difference can be observed between body feather Hg concentrations in adults from the Baltic Proper compared to those from the Norwegian coast. In fact, within the Baltic Proper no individuals were in the NRC while 95.2% fell within the MRC, and 4.8% still within the SRC despite declining time trends (Sun et al., 2019). The key legacy source of Hg in Sweden came from chlor-alkali plants and from metal production during the 1950 s and 1960 s (Lindqvist et al., 1991). In contrast, along the Norwegian coast 10.5% of the individuals fell within the NRC and none were above moderate risk, with 79.0 and 10.5% at low and moderate risk, respectively. AS for the more remote and supposed pristine adult eagles from W Greenland, had half-half inthe LRC and MRC, but none in the NRC.

3.3. Fish

In order to perform a risk assessment for fish we focussed on 11 species, i.e. the Atlantic cod (*Gadus morhua*), Atlantic herring (*Clupea harengus*), common bream (*Abramis brama*), common dab (*Limanda limanda*), common roach (*Rutilus rutilus*), common whitefish (*Coregonus lavaretus*), European flounder (*Platichthys flesus*), European perch (*Perca fluviatilis*), northern pike (*Esox lucius*), round goby (*Neogobius melanostomus*), and viviparous eelpout (*Zoarces viviparus*; Fig. 3; SI Table 3; SI Fig. 3). For most of these fish species, we were able to compare different Baltic Sea study regions, while there was still a grave lack of data to provide a consistent comparison among study regions for all species. For five of them, i.e. the Atlantic cod, Atlantic herring, common dab, European flounder, and viviparous eelpout, we were even able to provide a wider geographical comparison between the Baltic Sea and the Greater North Sea, and even with some North Atlantic stocks (in the case of Atlantic cod and common dab). None of the Baltic stocks showed individuals that fell under the two highest RCs, i.e. the HRC and SRC, with the exception of European perch. In general though, also including the neighbouring waters, no observations fell within the SRC, while 40.0% of the fish stocks in the Greater North Sea and North Atlantic seemed to have concentrations within the HRC.

Atlantic cod from the Baltic Proper and Danish Straits predominantly occured within the NRC (95.2–99.6%) with only a low proportion of the individuals occurring in the LRC (0.5–4.8%). The RC profiles in the Faroese and Icelandic Waters as well as in the Barents Sea were similar while Greater North Sea and Norwegian Sea had higher proportions of their stocks in the LRC and some even in the MRC (7.9 and 3.9%, respectively) and HRC (2.1 and 1.6%, respectively).

In all Baltic Sea study regions the stocks of Atlantic herring fell largely within the NRC (94.3–100.0%), while only the Gulf of Bothnia and the Baltic Proper also presented the MRC, as it seemed to be almost twice that for the Danish Straits and Gulf of Finland stocks. The Danish Straits presented Hg concentrations at which no risk for health effects

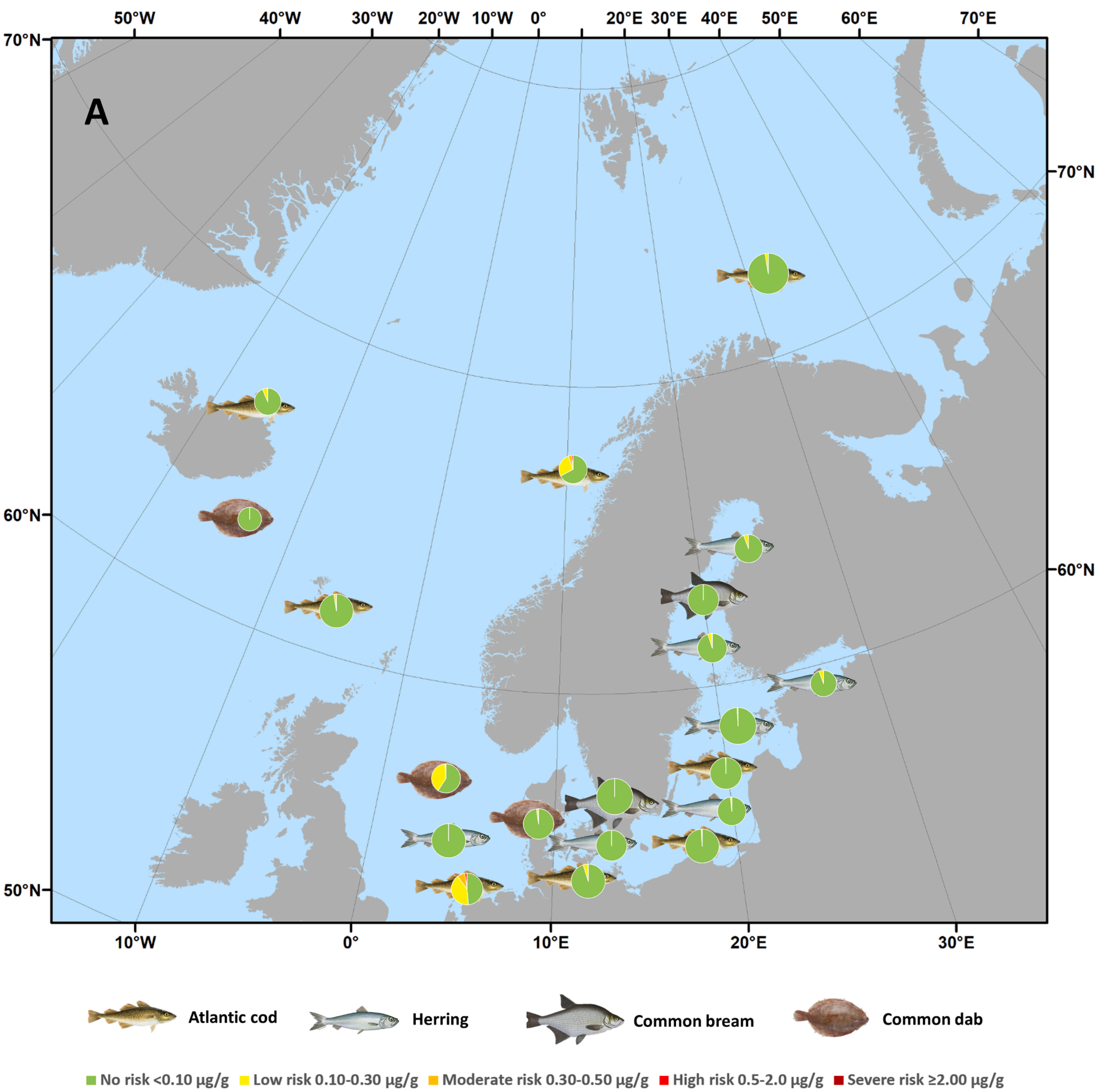


Fig. 3. A and B Geographical overview of the proportion of individuals of specific marine fish populations present in the Baltic that are at risk of Hg-mediated health effects extrapolated from mucle Hg contrations; based on post-2000 monitoring data grouped according to sex and maturity where possible. See SI Table 3 for the detailed information upon which this summary graphic is based and a ranked histogram on the same data in SI Fig. 3.

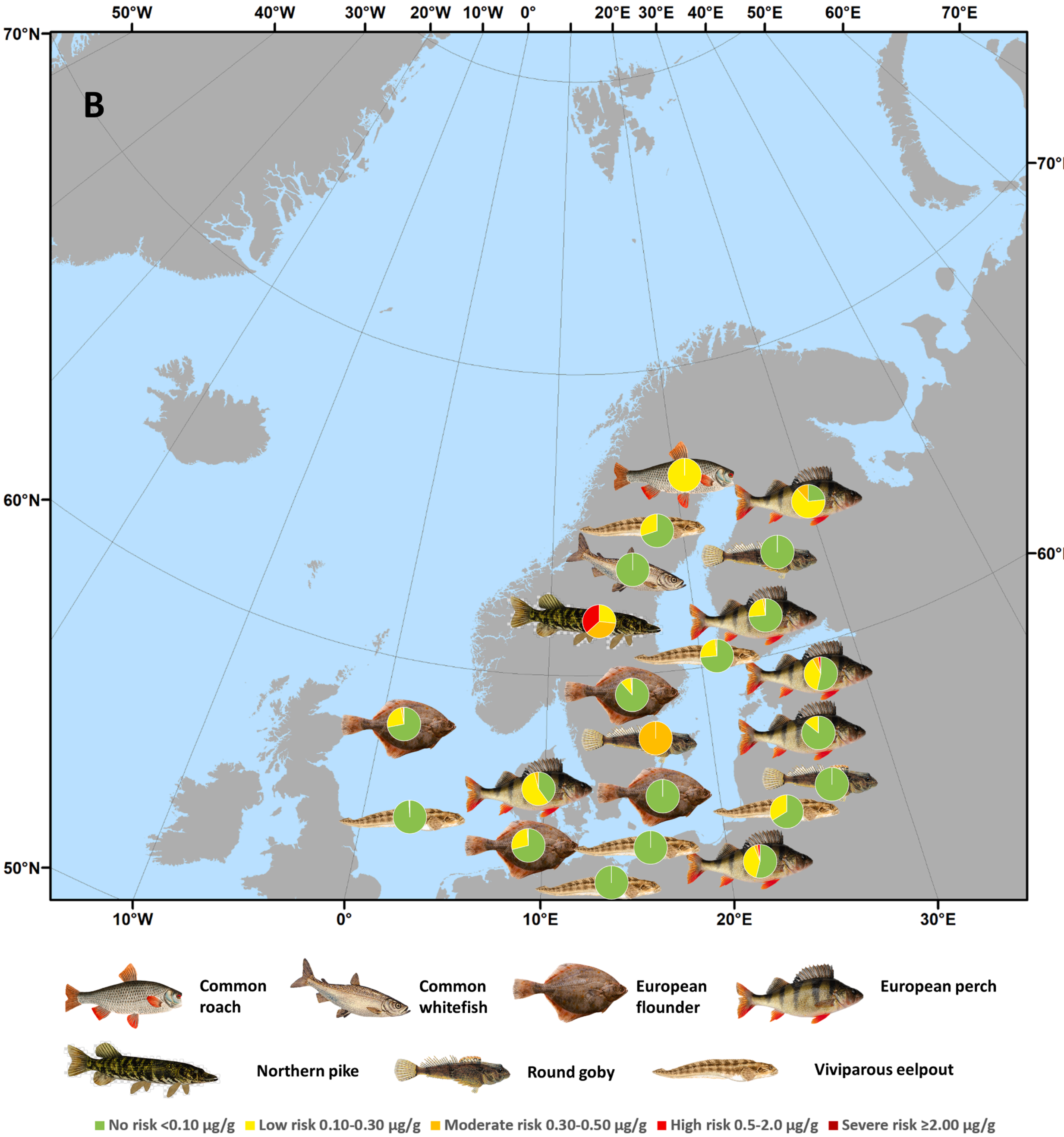


Fig. 3. (*continued*).

can be identified, similarly as the observations made for the Greater North Sea.

For common bream, common whitefish, round goby and common roach all individuals showed a high consistency in RC, though this may be because sample sizes were very small. All of the former bream fell within the NRC, with the exception of common roach that was entirely categorised under the LRC. Northern pike, then again, showed the opposite, where all individuals, all from the Gulf of Bothnia, were all at risk, while all were almost equally spread out over the LRC, MRC and HRC.

Data for the common dab were only available for the Danish Straits, showing that 97.8% is in the NRC, similar as to the Icelandic Waters stock, while the greater North Sea seemed to present high Hg concentrations resulting in only 59.1% being in the NRC and the remaining individuals to the LRC (39.7%), MRC (0.9%) or HRC (0.3%).

European flounder from the Baltic Proper had lower Hg concentrations than those in the Danish Straits, which seemed to be more similar in concentrations and RC profile than the Greater North Sea stocks. The latter had lower incidence (71.1–71.9%) in the NRC than for the Baltic Proper (88.4–100.0%).

European perch showed the highest proportion of its individuals within the NRC in the Baltic Proper and Gulf of Bothnia (23.4–85.6%), while the Gulf of Finland and Danish Straits had a smaller proportion in the NRC (40.0–53.5%) and likewise a higher in the LRC (38.4–56.0%). Viviparous eelpout almost showed the opposite, having all individuals free of risk in the Danish Straits, similar to the stocks in the Greater North Sea (NRC: 99.2 and MRC: 0.8%). This species still showed most of its individuals to be in the NRC (66.3–73.7%) though a part fell within the LRC (24.6–33.8%) and even within the MRC in the Baltic Proper (1.8%).

Finally, of the Viviparous eelpout from the Gulf of Bothnia and the Baltic Proper, 66.3–70.0% was in the NRC and the remaining

30.0–33.8% in the LRC (Fig. 3; SI Table 3; SI Fig. 3).

3.4. Bivalves

In order to perform a risk assessment for bivalves we focussed on four species, i.e. the Baltic macoma (*Macoma baltica*), blue mussel (*Mytilus edulis*), softshell clam (*Mya arenaria*), and zebra mussel (*Dreissena polymorpha*; Fig. 4; SI Table 4; SI Fig. 4). The dataset was extensive for blue mussel (n = 6,188) while data for the other three species remained spurious, i.e. <50 observations each. Thus, only preliminary conclusions can be drawn for the species with low sample sizes. Moreover, for these latter three species, we were not able to provide a comparison with the neighbouring waters, while for the blue mussel we can provide an extensive geographic comparison. The majority of the bivalves fell within the NRC or LRC, with only the Baltic macoma and blue mussel being represented by individuals at higher risk, within the MRC, and even within the SRC. No previous studies have ever presented a similar risk assessment procedure or such a large-scale evaluation. Finally, we would like to point out that the here-presented risk assessment would not be possible when only using MeHg data, which may have provided a more causal link though is very data-poor. Therefore we recommend the here obtained results and discussion to be taken as rough indications, as a result of large fluctuations in the proportions of MeHg of the total Hg content.

The concentrations in the Baltic macoma was similar to earlier observations made for the Baltic Sea along the Polish coast (Polak-Juszczak, 2012; Falandysz, 1994), and the concentration in blue mussels were within the typical observed range (Larsen et al., 2011; Briant et al, 2017). In all study regions both Baltic macomas and blue mussels fell within the NRC, LRC or MRC, with individuals in the Gulf of Finland and Danish Straits being those of lowest risk (NRC: 72.5–100.0%). In the Baltic Proper, macoma and blue mussels fell for a minor proportion within the MRC (6.5% and 1.5%, respectively), and in the Danish Straits a minor proportion (0.7%) of blue mussels was at severe risk. It is likely

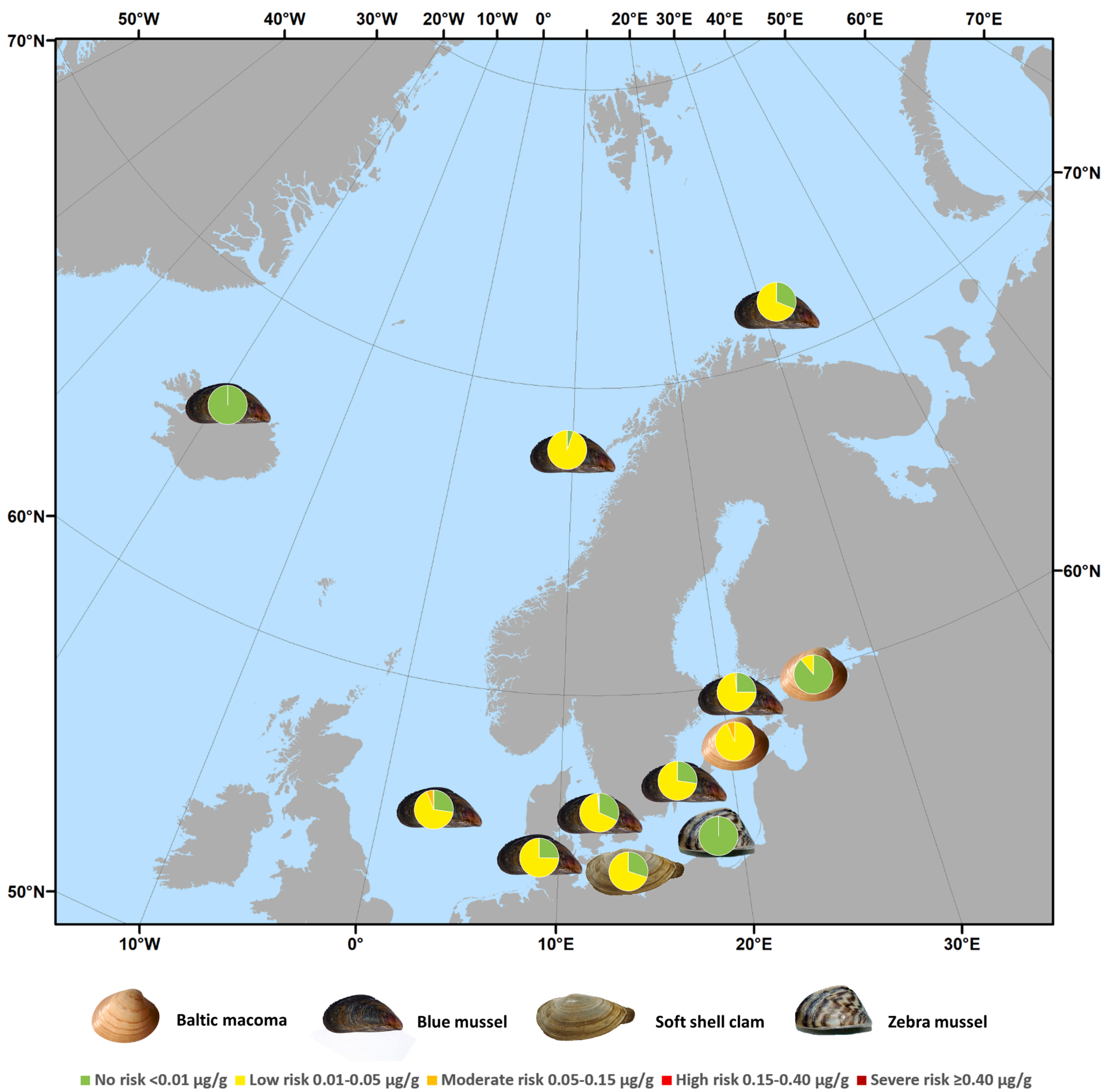


Fig. 4. Geographical overview of the proportion of individuals of specific bivalve populations present in the Baltic that are at risk of Hg-mediated health effects extrapolated from soft tissue concentrations; based on post-2000 monitoring data grouped according to sex and maturity where possible. See SI Table 4 for the detailed information upon which this summary graphic is based and a ranked histogram on the same data in SI Fig. 4.

that these few individuals were representing local sources not found in a larger region, or that these individuals carried a legacy of high body residues due to Hg bioaccumulation prior to the year 2000. For blue mussels, we can present a large-scale geographic comparison, showing that only two out of the eight groups (25.0%) contained individuals within the HRC and SRC, those having originated from the Greater North Sea and the Danish Straits. In the Greater North Sea, the majority of individuals (56.0%) still fell within the NRC and 37.9% were at low risk. Similar proportions were observed for the Norwegian Sea, while in the Barents Sea and Icelandic waters the majority (>96.5%) fell within the NRC, and only a small proportion (<3.5%) of the population was at low risk.

Concentrations in both the zebra mussel and softshell clam species fell within the low risk threshold. Moreover, zebra mussels from the Baltic Proper were believed to be at no risk, and most softshell clams, originating from the Danish Straits, fell predominantly within the NRC (80.0%) (Lepom et al., 2012). Unfortunately, no data were available for Hg concentrations in these two species from the Greater North Sea or the North Atlantic, and a comparison in potential health risk was therefore not possible. Finally, we would like to point out again that there RCs constructed for bivalves are pertinent to human consumption of these species, rather than indicative of health effects on the bivalves themselves, as to the present day no clear threshold levels can be discerned.

3.5. Limitations of the current risk assessment, monitoring programmes and recommendations

Traditional environmental risks assessment frameworks in for example the EU water policies are based on exposure assessments from intake of Hg as outlined in the Technical Guidance Document for deriving Environmental Quality Standards (EC, 2018). However, despite the existence of considerable weight-of-evidence regarding the biological effects of Hg exposure in Arctic and temperate species, major knowledge gaps remain when assessing concentration levels accumulated in organisms (e.g. AMAP 2011, 2018; Dietz et al. 2013, 2019). Recent risk assessments draw attention to the urgent need to establish better threshold concentrations for biologically relevant health effects for all monitored species including top predatory marine mammals (AMAP 2018; Dietz et al. 2019). For each studied species, it should ideally include the influence of variable maturity, sex or study region, in order to further develop reliable risk evaluations.

Furthermore, while the risk assessment frameworks employed in the present study has proven to allow for large-scale assessment, it does not take into account the potential tissue and species variation in organic Hg content (mainly the toxic MeHg). For most higher trophic levels as fish, birds and mammals, the MeHg is > 70% of total Hg (e.g. Dietz et al. 1990; Bloom 1992; Bond & Diamond 2009; Renedo et al. 2017). For lower trophic levels, and bivalves in particular, large variation and a range of 10–40% of MeHg to Hg ratios has been observed in Danish waters (Strand et al. 2010). In another study, Dietz et al. (1990) documented that almost all Hg was organic (both MeHg/CH_3Hg and CH_5Hg) in muscle tissue of seabirds and marine mammals. In mammals organic Hg in liver never exceeded concentrations of 2 μg g^{-1} even if the total Hg reached concentrations of up to 100 μg g^{-1}. Furthermore, the inorganic form of Hg is toxic to liver and kidney tissues due to its co-enzyme inhibition via high affinity to various microsome and mitochondria sulfhydryl-group enzymes affecting adenosine triphosphate-synthesis and its induction of oxidative stress (Branco et al. Goyer and Clarkson 2001). In addition to this, the detoxifying potential of selenium (Se), the effects of the essential Se being bound to Hg as well as information on inorganic Hg has not been included, as such data are very scarce and would not allow unravelling comprehensive interspecies or

spatial trends. MeHg is of specific concern due to its developmental and neurotoxicity, as well as high potential for bioaccumulation (Dietz et al. 2013). This dilemma has previously been brought forward by Ackerman et al (2016), who conducted a similar risk assessment for North American birds. In their dataset, <1% of the observations were on MeHg concentrations. In fact, under the Baltic Monitoring programmes, Hg exposure is not commonly determined as tissue MeHg concentrations. Ackerman et al. (2016) likewise stated that all Hg in feathers, eggs, whole blood and muscle is in the methylated form (Ackerman et al. 2013; Rimmer et al. 2005; Scheuhammer et al. 1998; Thompson and Furness 1989a). Therefore, total Hg in these are equivalent with the toxic methylated species. Opposite of this, a significant proportion of Hg in liver and kidney is in the inorganic form often bound to Se constituting the inert complex tiemanite (Thompson and Furness 1989b; Scheuhammer et al. 1998; Dietz et al. 2000; Raymond and Ralston 2004; Eagles-Smith et al. 2009). The toxicity of organic and inorganic Hg speciation differs as organic Hg entering the circulatory system reaches and passes the blood–brain-barrier (BBB) thereby resulting in high toxicity (Aschner and Aschner 1990). The target tissues upregulate subcellular synthesis of methallothionein and selenide complex binding detoxify Hg as it becomes inert (Dietz et al. 2013, 2019; Raymond and Ralston 2004). Looking at the molar ratio of Hg:Se is therefore important as it gives the information if Se is in surplus or deficit and thereby capable of detoxifying Hg by forming tiemanite complexes (Dietz et al. 2013, 2019). In the marine ecosystem, Se is in surplus while it is not always the case in freshwater systems. Therefore, Hg exposure will pose a greater threat to terrestrial species with risk of oxidative stress and neuro-endocrine disruption.

The present work faces the exact same challenge with respect to a lack of knowledge of interactive effect thresholds for multiple contaminant exposure as well as other environmental stressors including food and energy deprivation, parasite loads or climate change. Moreover, different tissues may have contrasting integration times and may thus reflect Hg contamination of individuals over different periods during which these may partly occupy different areas. However, combining Hg concentrations with temporal and spatial movements is challenging and have been scarcely investigated (Fort et al. 2016). Furthermore, we are mindful that spatial, species and maturity differences in Hg concentrations may be attributed to individual and species plasticity in dietary habits. We did, however, not endeavour the inclusion of biogeochemical proxies as earlier observed variation in their values is well-known to be also attributed to spatial and metabolic differences due to environmental conditions. The present risk assessment is hence the best possible assessment trying to evaluate the consequences of Hg exposure on health effects and should be taken with precaution, while the relative difference between study regions, species, maturity and sex remain reliable. Ideally, this information should be evaluated at population level effects as done for PCB in killer whales by Desforges et al. (2018a,b,c). Assessing the effect of Hg exposure and accumulation at the population-level is challenging for any species requiring long-term population monitoring to determine the link between observed tissue Hg levels and relevant long-term fitness metrics such as adult survival, reproductive success and recruitment, and ultimately population growth rates (Dietz et al. in review). Future research and monitoring programmes should also aim to concurrently analyses for MeHg, inorganic Hg and Se in order to underpin risk assessment better with physiologically-informed assessment of pathways.

3.6. Conclusions and considerations

A wide range of functional groups from the Baltic Sea and adjacent waters, i.e. the Greater North Sea and the North Atlantic, were investigated, delivering a risk assessment of post-2000 Hg exposure-associated health effects in a wide range of marine mammals, seabirds, birds of prey, fish and bivalves. We found that RC profiles were highly species-specific and that caution is warranted when attempting to discern general latitudinal or longitudinal trends, within the Baltic Sea region but also when comparing to the neighbouring waters. Generally, though, adults seemed to be more prone to carry a legacy of lifetime bioaccumulation whereas juveniles and yearlings were at much lower, or even no risk. Overall, over the last five decades, the situation for Baltic Sea inhabiting species has improved considerably with respect to Hg exposure, though it indeed still carries a legacy of elevated contaminant levels resulting from high industrial and agricultural activity and the Baltic Sea's central position among highly populated countries and slow water turnover. Generally, the associated health risk associated to the here presented exposure in a plethora of Baltic Sea species is not considerably higher compared to the same species in the Greater North Sea or the North Atlantic.

It is critical to pay more effort to better measure individual effects and upscale these to population-level effects using various modelling approaches, taking fertility, energy allocation, immune and endocrine functioning into account (Svensson et al., 2011). This requires a combination of *in vitro* dose–response studies as well as *in vivo* studies of key species in the Baltic (Desforges et al. 2016, 2017, 2018a,b,c). As for the ongoing monitoring programmes and available data in the literature, the majority of the information on Hg exposure is on total Hg with very limited information available on specific Hg species or Selenium, and therefore comes with certain degree of patchiness and unbalance of the dataset that unavoidably presents a uncertainty that is at the present day however acceptable, especially as no other methods are available to allow for this spectrum of interspecies and spatial comparisons. However, future strides to improve risk assessment approaches should be made utilising information on the toxic Hg species, i.e. MeHg, spanning food webs and large spatial management areas. This should be more and more realistic since the analytical methods for MeHg analysis have become more reliable and cost-effective in recent years (Azemard and Vassileva, 2015).

Finally, a thorough assessment of conventional risk thresholds with these recently or newly established ones remains to be endeavoured. For example, the risk evaluation for white-tailed eagle Hg exposure conducted earlier by Sun et al. (2019) does not seem to directly align with the one presented in the present study. The former did indeed use more conservative threshold values, where Hg exposure associated health effect are believed to occur only at body feather concentrations above 40.0 μg g^{-1} dw, a concentration well within the here proposed SRC. Nonetheless, the conservative threshold of natural biogeochemical Hg concentrations (5.00 μg g^{-1} dw) agrees with the lower RC at which low risk is expected. The former thresholds were based on a series of observations in white-tailed eagle and bald eagle to be exposed at this magnitude of exposure but without indicating compromised health. This discrepancy in risk evaluation is not necessarily an issue for the present study aiming to provide a large-scale geographical identification of hotspots for several species. It does, however, indicate that the here discussed health risks should be considered indicative, rather than definitive, as a multitude of local biogeochemical, trophic ecological and physiological factors are likely demanding risk thresholds that are sex, age, species and perhaps even location-specific.

Conflict of interest statements

We report that there are no conflicts of interests, and that the submitted manuscript has been reviewed and approved by all co-authors, and is not under consideration for publication elsewhere.

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

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2020.106178.

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