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Legacy and emerging organic pollutants in liver and plasma of long-finned pilot whales (*Globicephala melas*) from waters surrounding the Faroe Islands

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HIGHLIGHTS

- POPs and metabolites were analysed in liver and plasma of pilot whales
- Lipid normalized POP concentrations were mostly similar in liver and plasma
- POP concentrations in adult females were lower than in juveniles
- Relative concentrations of POPs differed between age groups
- Concentrations of metabolites were low in both adult females and juveniles

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ABSTRACT

Concentrations of PCBs, organochlorine pesticides (OCPs), brominated flame retardants and a suite of relevant metabolites of these POPs, in all 175 different compounds, were determined in liver and plasma of traditionally hunted pilot whales ($n = 14$ males and $n = 13$ females of different age groups) from the Faroe Islands.
The main objectives of this study were to determine differences in the presence and concentrations of the compounds in the liver and plasma, how they depend on developmental stage (calves, sub adults, and adult females), and to assess maternal transfer of the compounds to suckling calves.
Generally, the lipid weight (lw) concentrations of quantified POPs in the liver and plasma of pilot whales were positively correlated, and lw concentrations of most POPs did not differ between these matrices. However, concentrations of some individual POPs differed significantly ($p < 0.05$) between plasma and liver; CB-153 ($p = 0.044$), CB-174 ($p = 0.027$) and BDE-47 ($p = 0.017$) were higher in plasma than in liver, whereas p,p′-DDE ($p = 0.004$) and HCB ($p < 0.001$) were higher in liver than in plasma.
POP concentrations differed between age/gender groups with lower levels in adult females than in juveniles. The relative distribution of compounds also differed between the age groups, due to the influence of the maternal transfer of the compounds. The results indicated that larger, more hydrophobic POPs were transferred to the offspring less efficiently than smaller or less lipid soluble compounds.
Very low levels of both OH- and/or $MeSO_2$-PCB and PBDE metabolites were found in all age groups, with no significant ($p > 0.05$) differences between the groups, strongly suggesting a very low metabolic capacity for their formation in pilot whales. The lack of difference in the metabolite concentrations between the age groups also indicates less maternal transfer of these contaminant groups compared to the precursor compounds.

1. Introduction

Persistent organic pollutants (POPs), and in particular those that are halogenated, are widely distributed in the environment and bioaccumulate in marine food webs. Marine mammals occupy high trophic positions in their respective marine food webs, which along with

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physiological and life trait characteristics such as long life span, late reproduction, and a thick subcutaneous lipid layer (blubber), generally lead to accumulation of high concentrations of lipophilic POPs in marine mammals (Muir et al., 1988; Borrell, 1993; Troisi et al., 1998; Tilbury et al., 1999; Weisbrod et al., 2000, 2001; Tornero et al., 2005, 2006; Kajiwara et al., 2006; Fair et al., 2010; Letcher et al., 2010; Villanger et al., 2011).

Long-finned pilot whales (*Globicephala melas*) that inhabit the North-East Atlantic Ocean waters have been hunted as part of the Faroese traditional diet for centuries. In hunted pilot whales from the Faroe Islands, high concentrations of POPs like polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), dichlorodiphenyltrichloroethanes (DDTs), toxaphenes and other organochlorine pesticides (OCPs) have been documented (Borrell, 1993; Borrell and Aguilar, 1993; Lindstrom et al., 1999; Dam and Bloch, 2000; Dam, 2001; Hoydal and Dam, 2003, 2005, 2009; Nielsen et al., 2014). Although the concentrations of PCBs and DDTs in Faroese pilot whales have been reported as intermediate compared to pilot whales from other parts of the North Atlantic (Borrell and Aguilar, 1993), the concentrations in the 1990s were among the highest reported in mammals from the Arctic (AMAP, 2004). The highest reported concentrations were similar to or exceeded those expected to be linked to reproductive failure in other cetaceans (Reddy et al., 2001). The first reports of PBDEs in pilot whale blubber from the Faroe Islands were among the highest concentrations of PBDEs measured in biological samples (Lindstrom et al., 1999) at that time. PBDE determinations in blubber samples collected since then have not shown equally high concentrations to be common (van Bavel et al., 2001; Rotander et al., 2012a). Thus, concentrations of PBDEs in pilot whale blubber from the Faroe Islands appear to show a temporal decreasing trend (Rotander et al., 2012a; Nielsen et al., 2014).

POPs and their metabolites are known to interfere with several biological systems that can affect the health and fitness of exposed individual wildlife (Letcher et al., 2010). High levels of POPs have been associated with effects on reproduction, the immune system, and growth and development in marine mammals (Helle, 1976; Van Loveren et al., 2000; Reddy et al., 2001; Jepson et al., 2005). Thus, the high levels of contaminants in the tissues of pilot whales raise concerns for their health. It is also possible that continuous exposure to high POP loads in their tissues can cause subsequent effects at the population level.

Although very persistent, POPs can be eliminated by metabolism. In mammals the cytochrome P450 monoxygenases are the major enzyme class that catalyses the metabolism of xenobiotic, lipid soluble anthropogenic compounds, such as POPs. Although the presence and activities of several CYPs have been identified in different odontocete species (White et al., 1994; McKinney et al., 2004; Garrick et al., 2006; Fossi et al., 2008; Montie et al., 2008; Wilson et al., 2010; Godard-Codding et al., 2011), including pilot whales (Celander et al., 2000; White et al., 2000; Dam et al., 2010), the capacity of toothed whales to metabolize POPs is low compared to other mammalian species (Tanabe et al., 1988; Boon et al., 1992). As a consequence, relatively low levels of hydroxylated (OH) and methylsulfonyl ($MeSO_2$) metabolites of PCB and PBDE congeners (such as OH-PCBs and -PBDEs, and $MeSO_2$-PCBs and -DDEs) have been reported in odontocetes and the ratios between OH-PCB and PCB concentration are therefore also very low (McKinney et al., 2006; Houde et al., 2009; Montie et al., 2009; Weijs et al., 2009a; Nomiyama et al., 2010; Ochiai et al., 2013).

POPs are transferred from mother to offspring during gestation and lactation and a decrease in POP levels with age is often found in marine mammal females (Borrell et al., 1995; Sørmo et al., 2003; Vanden Berghe et al., 2010, 2012). In Faroe Island pilot whales, the transfer of organochlorines (OCs) to the offspring has been estimated to represent 60–100% of the mother's body load during lactation and only 4–10% during gestation (Borrell et al., 1995). Thus, suckling is the most important exposure route for OCs in young whales. Maternal transfer rates differ between different POP groups and specific compounds, where the difference has been linked to the lipophilicity, molecular size and chlorination of the compounds (Sørmo et al., 2003; Yordy, 2009; Bytingsvik et al., 2012). The more lipophilic chemicals are less transferable from the pregnant female to the foetus than less lipophilic chemicals (Borrell et al., 1995). Lower relative proportions of the higher chlorinated PCBs in young pilot whales compared to adults have been found in Faroese pilot whales, confirming a lower tendency of the most lipophilic compounds to be transferred to the offspring (Dam and Bloch, 2000).

It has also been reported that POP metabolites, such as OH-PCBs and OH-PBDEs, are transferred from the mother to the offspring in marine mammals, such as polar bears (*Ursus maritimus*) and seals, albeit to a much lower extent than for PCBs due to the more hydrophilic properties of these metabolites due to the hydroxyl group (Bytingsvik et al., 2012; Vanden Berghe et al., 2012). Since these metabolites have endocrine disrupting properties, there is particular concern about their harmful effects in young developing mammals, particularly with respect to thyroid disrupting effects (Bytingsvik et al., 2012, 2013; Simon et al., 2013). To our knowledge, there are no reports on the maternal transfer of these compounds in toothed whales. Information about levels in lactating calves of such whales would assist in risk assessment of POPs in these species.

The production and usage of PCBs and OCPs have been regulated globally since the 1970s, and the levels have been reported to have decreased in marine top predators that are known to have high capacities for metabolising POPs, such as polar bears (McKinney et al., 2010; Bytingsvik et al., 2012; Dietz et al., 2013). However, there is limited information on current levels and thus trends in marine mammals that have poor abilities to metabolize POPs, such as toothed whales. Thus, information on more recent levels of POPs in Faroe Island pilot whales will provide knowledge on temporal trends of POPs in a group of animals that is characterized by very low capacities to metabolize and excrete POPs.

Knowledge of the partitioning of contaminants between tissues is needed when assessing levels and effects of contaminants in animals. Blubber is the primary site of POP accumulation and in toothed whales more than 90% of the total body load of contaminants is found in the blubber (Yordy et al., 2010). Blood may however be a more accurate indicator of the bioavailable contaminant concentrations (Yordy et al., 2010). In other cetaceans it has been shown that the levels of PCBs are highly correlated in blubber and blood and that concentrations in blubber can be used to estimate the levels in blood and vice versa (Yordy et al., 2010). Levels of PCBs and PBDEs have also been found to be highly correlated between liver and blubber and the partitioning between the tissues is governed by the lipid content (Raach et al., 2011), although liver may be more representative of recent contamination of lipophilic compounds due to a slower partitioning of contaminants into blubber (Raach et al., 2011).

In the present study we determined the liver and plasma concentrations of PCBs (74 congeners), OCPs (20 compounds), brominated flame retardants (17 congeners/compounds) and relevant metabolites of some of these POPs: 33 OH-PCBs, 14 OH-PBDEs, 16 $MeSO_2$-PCBs, pentachlorophenol (PCP), 4-OH-heptachlorostyrene, and 3-$MeSO_2$-*p,p'*-DDE, in different age groups of pilot whales (0–2 year old calves, subadults, and adult females) from the Faroe Islands sampled in 2010 and 2011. The main objectives were to 1) determine differences in the presence and concentrations of the compounds in the liver and plasma; 2) determine how the presence and concentrations of POPs depend on developmental stage (0–2 year calves, subadults, and adult females); 3) assess maternal transfer of the compounds to lactating calves based on the differences in concentrations between adult females and calves. Based on the previously reported low concentrations of PCB and PBDE metabolites in toothed whales, we hypothesised the concentrations of these contaminant groups to be low in the adult females, even lower in calves and intermediate in the sub-adult pilot whales.