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Viewpoint

An overview of time trends in organic contaminant concentrations in marine mammals: Going up or down?

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ABSTRACT

In this article I review recent trends reported in the literature from 2008 to date for organic contaminant concentrations in marine mammal tissues worldwide, in order to get an idea of where we stand currently in relation to the control of hazardous substances. For many contaminants which have been subject to regulation regarding their production and use (e.g. organochlorine pesticides, PBDE and HBCD flame retardants, butyltins) trends are downwards. For perfluorinated compounds, trends are more mixed. For dioxins, furans and dioxin-like CBs, there are no recent data, for either concentrations or trends. For CBs overall, earlier downward trends in concentration in UK harbour porpoises following regulation beginning in the 1980s have stalled, and remain at toxicologically significant levels. This raises concerns for killer whales and bottlenose dolphins who, because of their larger size and greater bioaccumulation potential, have higher levels still, often far above accepted toxicological threshold values.

1. Introduction

In this paper, I summarise what is known about current time trends in the concentrations of organic contaminants in marine mammals. Marine mammals are top predators within the food chains of the marine environment and so represent the major accumulation level for long-lived, lipophilic, contaminants accumulated from lower trophic levels. For some compounds – butyltins and perfluorinated compounds – accumulation is primarily in liver, due to differences in physico-chemical properties. This both makes the concentrations easier to determine, as the concentrations are higher, and also reflects the level of the most serious potential impacts. Impacts have, in fact, been demonstrated, for PCBs at the least (Jepson et al., 2005; Hall et al., 2006), in terms of an increase in infectious disease mortality, probably due to immunosuppression. Also, the ability for concentrations of lipophilic contaminants in blubber to demonstrate clear and statistically robust trend information has been amply demonstrated within the UK Cetacean Strandings Investigation Programme which, for PCBs in harbour porpoises, has probably the world's largest marine mammal dataset (Law et al., 2012a). This overview is based upon articles from the scientific literature published from January 2008 to date, so as to summarise recent trend information from all areas. Study of time trends is important as, for legacy contaminants, it allows the effectiveness of earlier legislation to be assessed and, for novel/emerging contaminants, allows their significance to be assessed and priorities for future study to be defined (see e.g. Covaci et al., 2011; van der Veen and de Boer, 2012).

2. Organochlorine pesticides

No clear trend could be observed in chlordane or DDT (and metabolite) concentrations over the ca. 10 year lifespan of a blue whale taken off California, USA, in 2007, determined from analysis of its waxy earplug (Trumble et al., 2014). Leonel et al. (2010) determined organochlorine pesticide concentrations in blubber of franciscana dolphins from Brazil (1994–2004): concentrations of DDT and its metabolites, HCB, chlordanes and dieldrin showed slight decreases during the study period. Although levels were low, there was no temporal trend in DDT and metabolite concentrations over the period 1991–2005 in blubber of sea lions from the Valdés peninsula in Argentina (Borrell et al., 2010).

Law et al. (2013) determined concentrations of organochlorine pesticides (HCB, HCHs, DDTs and dieldrin) in blubber of bycaught female common dolphins from 1992 to 2006: all trends were downwards but only that for HCHs was statistically significant ($p = < 0.01$).

Decreasing concentrations of *p,p'*-DDE, HCB and HCHs were observed in ringed seals from central west Greenland over the period from the early 1990s to 2010, with the fastest rate of decline for α-HCH (ca. 10% per year) and the slowest for β-HCH (1.9% per year).

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In striped dolphins from Japan, no trend was observed in blubber concentrations of DDTs, chlordanes, HCHs or HCB over the period 1978–2003 (Isobe et al., 2009). In melon-headed whales mass stranded on Japanese coasts in 1982, 2001/2002 and 2006, Kajiwara et al. (2008) reported that levels of DDTs and HCB were lower at the end of the period that at the start. Concentrations of chlordanes showed an increase during the period, and those of HCHs were stable.

3. Brominated flame retardants

HBCD was determined in blubber of juvenile ringed seals from east Greenland (1986–2008) using both fresh and archived samples (Vorkamp et al., 2011). α-HBCD was the only diastereoisomer consistently detected and showed an annual increase over the period of around + 6.1%, although the concentrations were several orders of magnitude lower than those reported from industrialised areas. In the same study, ∑BDE concentrations showed a declining trend from about 2003, in line with the removal of the penta-PBDE product from the market prior to the 2004 ban within the EU. Vorkamp et al. (2012) extended this time series to 2010, during which period HBCD concentrations continued to increase.

No clear trend could be observed in BDE47 concentrations over the ca. 10 year lifespan of a blue whale taken off California, USA, in 2007, determined from analysis of its waxy earplug (Trumble et al., 2014). Earlier studies in sea lions had also shown no significant time trends for BDEs or HBCD over the period 1993–2003 (Stapleton et al., 2006). Ross et al. (2013) demonstrated an exponential increase in ∑BDE concentrations in blubber of harbour seals from the Salish Sea, North America, during 1984–2003, but concentrations then stabilised from around 2005 and 2009. In beluga whales from the St Lawrence estuary (Canada), the concentrations of neither BDE congeners nor homologue groups showed any trend in concentration during 1993–2007 (Raach et al., 2011). In harbour seals from the northwest Atlantic during 1991–2005, Shaw et al. (2008) observed no significant time trend for $\sum BDE_{25}$ concentrations. Congener profiles changed over time, but exposure was mainly to the penta-mix PBDE formulation.

In blubber of harbour porpoises stranded on coasts of the eastern North Sea $\sum BDE_7$ concentrations were 132–4770 μg kg^{-1} lipid weight in 1990–1998 and 82–1830 μg kg^{-1} lipid weight in 2000–2008, an apparent decrease (Weijs et al., 2010). In UK harbour porpoises, concentrations of both BDEs and HBCD in blubber were seen to be declining following earlier increases, during the periods 1992–2008 and 1994–2003, respectively (Law et al., 2012a).

In the Arctic, BFR concentrations were reviewed in 2010 (de Wit et al., 2010). Temporal trends of tetra- to hepta-BDEs and HBCD showed increasing or stabilising concentrations, with no clear picture emerging. A range of novel/emerging flame retardant compounds were detected, indicating that these compounds are subject to long-range atmospheric transport and that concentrations will likely increase in the future.

In striped dolphins from Japan, increasing trends were observed in blubber concentrations of PBDEs and HBCD over the period 1978–2003 (Isobe et al., 2009). In melon-headed whales mass stranded on Japanese coasts in 1982, 2001/2002 and 2006, Kajiwara et al. (2008) reported that levels of PBDEs increased during the period of the study.

4. Dioxins, furans and dioxin-like PCBs

No recent temporal trend data are available and this represents a knowledge gap, particularly as high concentrations are still found in fish from some areas, such as the Baltic Sea, in regard of which concerns have already been raised relating to human consumption of fish from these locations (European Food Safety Authority, 2005). Further study of these compounds in marine mammals is indicated.

5. Polychlorinated naphthalenes

Few studies have addressed polychlorinated naphthalenes (PCNs) and, in the UK at least, there is considerable reluctance within government to do so as they are regarded as purely a legacy problem and so of no current interest. Therefore, no current data are available. Rotander et al. (2012) studied PCNs in marine mammals from the Arctic and sub-Arctic from 1986 to 2009. Species studied were long-finned pilot whales, ringed seal, minke whale, fin whale, harbour porpoise, hooded seal and Atlantic white-sided dolphins. No statistically significant trends could be observed, although, in minke whales off Norway, the lowest concentrations were found in samples from the latest sampling period. Ross et al. (2013) found a rapidly declining trend in PCN concentrations in harbour seals from the Salish Sea (NW North America) during 1984–2009.

6. Perfluorinated compounds

Kratzer et al. (2011) determined concentrations of polyfluoroalkyl compounds (PFCs) in grey seal livers from the Baltic Sea over the period 1974–2008. Seventeen of forty-three compounds were detected. Of the compounds detected, most showed peak concentrations in either 1997 or 1998, followed by declines subsequently. In the Baltic and North Seas, Huber et al. (2012) reported that, over the period 1991–2008, concentrations of perfluoralkyl sulphonates and perfluorooctane sulphonamide in harbour porpoise livers decreased over time while the perfluoroalkyl carboxylate concentrations increased. The highest concentrations were seen in the Baltic Sea and the lowest around Iceland.

Rigét et al. (2013a,b) assessed the concentrations of perfluorinated alkylated substances in ringed seals from Greenland following a major reduction in production in the early 2000s. Concentrations of many of these compounds have begun to decline, from around 2006. Others have not, although a time-lag of up to 10 years due to atmospheric transport processes may partly be the reason.

Galatius et al. (2011) studied perfluorochemicals in harbour porpoises from the Danish North Sea during the period 1980–2005. The highest concentrations were found in neonates, suckling juveniles and lactating females. Despite efforts to reduce emissions, no decreasing temporal trends were observed for any compounds. Concentrations of perfluorinated carboxylic acids were found to be increasing. In the German Bight, Ahrens et al. (2009) studied temporal trends of polyfluoroalkyl compounds in livers of harbour seals over the period 1999–2008. Decreasing trends were observed in some classes of these compounds, but this may be result from substitution with shorter-chain analogues, which may be less persistent, rather than total cessation of use. Overall, trends were mixed.

In livers of melon headed whales stranded along the Japanese coast in 1982, 2001/2002 and 2006, Hart et al. (2008) determined concentrations of perfluorinated compounds. The authors noted that concentrations of the original major compound in use, PFOS (perfluorooctane sulphonate) had been declining in European and North American coastal waters since the early 2000s, but that no data were available for Asian waters. The predominant compounds detected were PFOS and PFOSA (perfluoroctanesulphonamide). Concentrations of these compounds rose by approximately 10× between 1982 and 2001/2002, and PFOSA concentrations fell by