**CONCENTRATIONS OF LEGACY PERSISTENT ORGANIC POLLUTANTS AND NATURALLY
PRODUCED MeO-PBDES IN DUGONGS (*DUGONG DUGON*) FROM MORETON BAY, AUSTRALIA**

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Abstract

Dugongs (*Dugong dugon*) are an iconic and strictly herbivorous species. They inhabit coastal areas, which brings them in contact with urban and agricultural pollutant sources, yet their exposure and susceptibility to environmental pollutants is still largely unknown. The goal of this study was to investigate the presence of several legacy compounds such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and pesticides as well as naturally produced MeO-PBDEs in male and female dugongs from Moreton Bay (n = 24), a semi-enclosed embayment close to Australia's third largest city, Brisbane. Results show that concentrations of all investigated compounds are low in general (< 120 ng/g lipid weight) and below known toxicity thresholds established for marine mammals. However, concentrations found in this study are higher or comparable to concentrations in dugongs outside Moreton Bay or in sirenians worldwide. No temporal trends for POPs from 2001 until 2012 were found for adult animals suggesting that environmental changes are only slowly reflected in dugongs. Finally, pollutant profiles in dugongs are limited to the most persistent PCBs, pesticides and PBDEs that also dominate POP profiles in other marine mammal species in general.

Keywords

POP, PCB, dugongs, MeO-PBDE, herbivorous, Australia

Introduction

Pollution in marine environments is a widespread problem with studies reporting considerable concentrations of environmentally relevant pollutants even in remote areas (Lohmann et al., 2006; Jamieson et al., 2017). Although new and emerging contaminants, such as phthalates, UV filters and personal care products in a range of matrices (e.g. Heudorf et al., 2007; Brausch and Rand, 2011; Tsui et al., 2017) currently attract a lot of attention, studies focusing on legacy POPs (persistent organic pollutants) still abound. Compounds such as PCBs (polychlorinated biphenyls) are probably the best-known POPs because of their known health impacts, worldwide distribution and long use. This pollutant class consists of 209 congeners that were used in varying mixtures throughout the 1950s and 1960s as coolants and dielectric fluids in electronic equipment, and components in glues and paints (ATSDR, 2000). PCBs were banned globally in the 1970s, however, there is evidence for more recent PCB manufacturing sources (Hu and Hornbuckle, 2010; Koh et al., 2015; Herkert et al., 2018) making PCBs both legacy and re-emerging contaminants. PBDEs (polybrominated diphenyl ethers) are a class of flame retardants that also consist of 209 congeners. Voluntary bans of selected PBDEs started in the early 2000s, but it is presumed that a large amount is still stockpiled in PBDE-containing consumer products (Shaw and Kannan, 2009; Wiseman et al., 2011). DDT (dichlorodiphenyltrichloroethane) and its isomers and metabolites (DDE and DDD) are pesticides that were banned in the 1970s, but restricted uses in disease vector control are still currently permitted in areas affected by malaria (Mansouri et al., 2017). Similar to DDT, chlordane and related compounds are insecticides that were banned in the 1980s, while hexachlorobenzene (HCB) is a fungicide that was banned in the 1970s although it may still be produced unintentionally (ATSDR, 1994; Reed et al., 2007). All of these compounds and POP classes are listed under the Stockholm Convention because of their persistence, ability to biomagnify in the food chain, toxic effects and potential for long-range

transport. With the oceans being identified as major sinks for pollutants (Lohmann et al., 2006), these four properties are especially relevant for marine top predators such as several marine mammal species. Consequently, there are numerous studies reporting on the considerable concentrations of pollutants in marine mammals with some studies linking pollution to adverse effects of their reproductive, immune and endocrine systems (e.g. DeLong et al., 1973; Helle et al., 1979; Aguilar and Borrell, 1994; Jepson et al., 2005; Mos et al., 2010).

In toxicology, marine mammals are considered excellent bioindicators for exposure to pollutants in the marine environment (Ross, 2000). Because of their long life spans, large lipid reserves, inability to metabolize several lipophilic POPs, such as PCBs, and top position in the marine food webs, the concentrations found in these animals are usually elevated and of concern. However, not all marine mammals are apex predators.

Dugongs (*Dugong dugon*) are herbivorous and strictly marine animals, a combination that makes them unique among other Sirenia as well as marine mammals in general. As herbivores, they are typically positioned in the lower regions of the trophic food chain, which means that the concentrations of pollutants are expected to be lower than those typically detected in piscivorous (odontocetes) or mammal-eating (killer whales and polar bears) marine mammals. In Australia, studies have indeed reported much lower concentrations of organohalogenes such as PBDEs, PCBs, pesticides and chlordanes in herbivorous dugongs and green turtles compared to piscivorous dolphins and other marine turtle species (Vetter et al., 2001; Gaus et al., 2005; Haynes et al., 2005; Hermanussen et al., 2008; Weijs et al., 2016a; Vijayasarathy et al. 2019). However, these studies either had limited sample sizes and/or a focus on adult females, which are poor representatives of the population due to the offloading of pollutants to their offspring during gestation and lactation (Debieer et al., 2003; Weijs et al., 2010). In addition, dugongs investigated in these six studies were obtained from several

locations along the Queensland coast with only three studies reporting levels of POPs such as PCBs, PBDEs or pesticides in (adult female) dugongs from Moreton Bay in southeast Queensland (Hermanussen et al., 2008; Weijs et al., 2016a; Vijayasathya et al. 2019).

While dugongs are distributed across the coast of southeast Queensland, previous research has shown that the animals from Moreton Bay are distinguished genetically from other populations outside the Moreton Bay area thereby implying a high site fidelity for dugongs in this region (Seddon et al., 2014). Moreton Bay is a semi-enclosed and shallow embayment that receives inputs from several rivers and waterways, such as the Brisbane river, which flows through a highly urbanized city with a population over 2.4 million people (Brisbane, Queensland). In a previous study, concentrations of PCBs and DDT in blubber of some humpback dolphins (*Sousa sahulensis*) from Moreton Bay were found to be comparable to concentrations found in highly contaminated transient killer whales and to exceed several toxicity thresholds associated with immunotoxicity and reproductive effects (Weijs et al., 2016b). These results show that legacy POPs are present in the Moreton Bay ecosystem at considerable concentrations, which raises the question of how these chemicals bioaccumulate in this dugong population at large rather than only in adult females.

The present study aims to fill the knowledge gaps about the toxicological status of dugongs in Moreton Bay as a contaminated and urbanized area in southeast Queensland, Australia. More specifically, this study aims to 1) investigate the influence of age, year of death and gender on the bioaccumulation of legacy POPs in dugongs, 2) compare the results to concentrations for dugongs and marine mammals in the literature, and 3) investigate, for the first time, the presence of some naturally produced methoxylated PBDEs (MeO-PBDEs) in dugongs in Moreton Bay. Despite their natural origin, these compounds can be transformed into hydroxylated PBDEs (HO-PBDEs), which often have a greater toxicity than PBDEs (Wiseman et al., 2011).

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Materials & Methods

Samples and target analytes. Blubber (n = 22) and liver (n = 11) samples from 24 dugongs (*Dugong dugon*) were included in this study. All samples were obtained from dead animals (e.g. stranded, victims of fisheries by-catch, indigenous hunting) from 1998 until 2012. All animals were from the Moreton Bay region (Queensland, Australia) except for one animal that originated from northern Queensland (Torres Strait), which was included for comparison reasons. For all individuals, detailed information is available in Tables S1 (females and animals with unknown gender) and S2 (males) in the Supporting Information. In all samples, 23 polychlorinated biphenyl congeners (PCBs; IUPAC numbers 28, 52, 74, 99, 101, 105, 118, 128, 138, 146, 153, 156, 170, 171, 177, 180, 183, 187, 194, 196/203, 199, 206, 209), 5 chlordanes (CHLs; oxychlordanes-OxC, *trans*-nonachlor-TN, *cis*-nonachlor-CN, *trans*-chlordanes-TC, *cis*-chlordanes-CC), 3 hexachlorocyclohexanes (HCHs; α , β , γ), hexachlorobenzene (HCB), 2 pesticides (DDXs; *p,p'*-DDE, *p,p'*-DDT), 7 polybrominated diphenyl ethers (PBDEs; IUPAC numbers 28, 47, 99, 100, 153, 154, 183) and 2 naturally occurring methoxylated PBDEs (MeO-PBDEs; 6-MeO-BDE47 and 2'-MeO-BDE68) were targeted.

Sample preparation. Approximately 0.150 g of blubber or 1.50 g of liver was homogenized with anhydrous Na₂SO₄ and spiked with internal standards which included PCB 143, ϵ -HCH and PBDE 77. Homogenized samples were added to a glass column, covered with a mixture of acetone/n-hexane (1/3, v/v) and extracted 3 times: homogenized samples were left covered by the solvent mixture for a minimum of 2 h (twice) and overnight (once). The extracts were combined (per sample) and evaporated to approximately 10 mL. About 1/8th of each of the resulting extracts was used for gravimetric lipid determination after evaporating the aliquot on aluminium dishes in the oven (100°C, 1 h). The remaining 7/8th of each extract was cleaned up on approximately 8 g of acid silica (H₂SO₄, 44%) with 20 mL n-hexane and 15 mL

dichloromethane (DCM). Extracts were evaporated to near dryness, re-dissolved in 100 μ L recovery standard (PCB 207 in iso-octane) and kept at -20°C until GC-MS analysis.

Instrumental analysis. All analytes (PCBs, DDTs, HCB, chlordanes, HCHs, PBDEs and MeO-PBDEs) were quantified with an Agilent 6890 gas chromatograph coupled with a 5973 mass spectrometer system (GC-MS) (Weijjs et al., 2009). The GC was equipped with a 30 m $\times$ 0.25 mm $\times$ 0.25 μ m DB-5 capillary column. The MS operated in electron capture negative ionisation (ECNI) mode and was used in the selected ion-monitoring (SIM) mode with 2 ions monitored for each analyte of homologue group.

Quality Assurance/Quality Control (QA/QC). Mean recoveries $\pm$ SD (both expressed in %) for internal standards were 100 $\pm$ 6, 68 $\pm$ 4 and 78 $\pm$ 5 for PCB 143, ϵ -HCH and PBDE 77, respectively. For each analyte, the mean procedural blank value was used for subtraction. After blank subtraction, the limit of quantification (LOQ) was set at 3 times the standard deviation of the procedural blank, which ensures > 99 % certainty that the reported value is originating from the sample. For analytes that were not detected in procedural blanks, LOQs were calculated for signal to noise ratio equal to 10. LOQs depended on the sample intake and on the analyte and ranged between 2 and 20 pg/mL extract. QC was performed by regular analyses of procedural blanks, by random injection of standards and solvent blanks.

In addition, 4 dugongs that were analysed for PCDD/Fs and PCBs in Vijayasathay et al (2019) were re-analysed in order to get data on PBDEs, DDXs, HCB, HCHs, CHLs, and MeO-PBDEs. This provides the opportunity to compare the PCB results as sample preparation and analysis were done differently and in different laboratories (Table S3). Finally, blubber of a humpback dolphin (sample ID BIO-328) was included as a laboratory reference sample. This sample was analysed in three different institutions with different procedures for sample preparation and analysis thereby giving the opportunity to assess the

performance of the current method (Table S4). The results of this QA/QC framework can be found in the Supporting Information (Table S3 for dugongs, Table S4 for dolphin BIO-328).

Statistics. The impacts of gender and year of death (independent variables) were tested on the concentrations of pollutants (sums of POP classes with PCBs divided into groups according to the number of chlorine atoms) in blubber while accounting for the body size (body length as a proxy for age; covariate) through two-way ANCOVA statistical tests. Due to the limited number of liver samples, the impact of the year of death could not be included. Therefore, for liver, the impact of gender (independent variable) was tested on the pollutant concentrations while accounting for the body size (covariate) through one-way ANCOVA statistical tests. Tests were performed using SPSS (IBM SPSS Statistics version 25) and the level of statistical significance was set at $\alpha = 0.05$. For compounds detected in more than 50% of all samples, concentrations below LOQ were replaced by a value of f (frequency of detection) * LOQ (James et al., 2002). All compound levels were log-transformed to achieve normality (with $\log(x+1)$ to avoid negative values). Animals for which information about gender and body size was not available (BIO-69 and BIO-329) were not included in the statistical analysis. The one animal from the Torres Strait (BIO-600) was also excluded from any statistical test. Year of death for the Moreton Bay animals varied from 2001 until 2012 and this variable was divided into 3 groups of 4 years (YI = 2001-2004; YII = 2005-2008; YIII = 2009-2012) for statistical purposes only as there were several years with limited sample sizes ($n = 1$ or 2). All graphs were made with GraphPadPrism (version 8).

Results

Lipid percentages and non-detects

The average lipid percentage in the blubber of females was $67\% \pm 15\%$ (min 43%, max 80%; Table S1). The average lipid percentage in the blubber of males was $63\% \pm 25\%$ (min 3%, max 84%; Table S2). Lipid percentages in blubber of the two animals from unknown gender and age were 62 and 67% (Table S1). Lipid percentages in the liver were lower compared to blubber with percentages ranging from 3.7 to 5.1% for the females, 2.8 to 5.2 % for the males and 2.8% for one animal with unknown gender (Tables S1 and S2).

PCBs 28, 52, 74, 101, 99, 206, 209, α -HCH, β -HCH, γ -HCH, and PBDEs 28, 153, 183, 100 were not detected in any sample (blubber or liver). PCBs 171, 156, 199, 194, OxC, *p*'*p*-DDT, PBDE 99 and 6-MeO-BDE47 were found in less than 50% of all samples with OxC, *p*'*p*-DDT, PBDE 99 and 6-MeO-BDE47 not detected in any liver sample. All these compounds were excluded from any calculations and statistical analysis.

Influence of gender, year of death and body size on the bioaccumulation of POPs

All results are provided in Table 1 and S5. Partly because of the relatively low sample size, there were only few statistically significant differences in pollutant levels in the present study. Results showed that the concentrations of all pollutant groups in blubber of females were comparable from 2001-2012 (YI, YII and YIII) and, when compared to males from 2001-2012, the only statistically significant differences were found with the concentrations in blubber of males in 2009-2012 (YIII) for hexa-, hepta-, and octa-PCBs ($p = 0.002$, 0.002 and 0.003 for hexa, hepta and octa, respectively; Fig 1 and Fig S2). For the males, only levels of hexa-, hepta-, and octa-PCBs were higher in the YIII group ($p = 0.008$, 0.003 , and 0.002 for hexa, hepta and octa, respectively) while there were no differences in concentrations between the YI and YII groups (Fig 1 and Fig S2).

For the liver, no statistically significant differences were found for any pollutant group between males and females while taking into account the body sizes (one-way ANCOVA; p -values ranged from 0.515 to 0.898).

Profiles

All compounds. Proportions of p,p' -DDE were highest in the majority of all samples (21 out of 27 samples) ranging between 15 and 60% of the total sum of all pollutants (Fig 2). DDE was followed by the sum of PCBs (mean $\pm$ SD for all blubber samples = $31 \pm 9\%$; for all liver samples = $30 \pm 16\%$). For 6 out of 27 samples, proportions of the sum of PCBs were higher than p,p' -DDE. For blubber, 2'-MeO-BDE 68 had the highest proportions after the sum of PCBs ($12 \pm 7\%$) (Fig 2A). However, for liver, 2'-MeO-BDE 68 only came after HCB ($9 \pm 7\%$) and chlordanes ($7 \pm 4\%$) with a mean $\pm$ SD of $6 \pm 4\%$ (Fig 2B). Between the adult males, adult females and juveniles, the biggest differences could be found in the proportions of 2'-MeO-BDE 68 and p,p' -DDE to the total sum of all organohalogens. The overall pattern of the calf resembled that of the adult females in the blubber, but not in the liver. Blubber from an adult male from the Torres Strait was analysed for comparison (Fig 2A). This animal had a distinctly different overall profile compared to the Moreton Bay animals in this study with HCB and sum PCBs (both 24%) as most dominant compound (groups) followed by 2'-MeO-BDE 68 (18%), sum of PBDEs (16%) and p,p' -DDE and sum of chlordanes (both 9%) (Fig 2A). The profiles in the blubber from the two animals with unknown gender and age (or body size) were similar to the profiles of the juveniles (Fig 2A).

Compound groups. The PCB profiles were dominated by PCB 153 in all animals regardless of tissue type, age, body size or gender. PCB 153 was followed by PCB 138 in all samples, except for the blubber and liver of the male calf (BIO-59) where PCB 118 was present at

higher concentration than PCB 138. The order was hexa-PCBs > hepta-PCBs > penta-PCBs > octa-PCBs in all animals except for the blubber in some males (2 adult males, 1 juvenile male and 1 calf from Moreton Bay, 1 adult male from Torres Strait) and in 1 of the 2 animals with unknown gender or age where penta-PCBs were more prevalent than hepta-PCBs.

Among the chlordanes, there were no clear trends in the blubber while CC had the highest levels in 9 out of 11 liver samples. For the PBDEs, PBDE 47 was predominant in 25 out of 31 samples while PBDE 154 was predominant in the remaining 6 out of 31 samples. 2'-MeO-BDE 68 was detected in all samples except for the blubber and liver of one adult male (BIO-61) and 6-MeO-BDE 47 was only found in the blubber of one adult female.

Tissue distribution of POPs

Paired blubber and liver samples of 3 adult females, 4 adult males, 1 male calf and 1 animal with unknown gender/age were available. For these animals, liver/blubber (L/B) ratios were calculated based on the concentrations in lw (lipid weight) with L/B ratios < 1 being indicative for a preferential accumulation in the blubber, L/B ratios > 1 indicating a preferential accumulation in the liver and L/B around 1 indicating no preferential accumulation in both tissue types. L/B ratios for all animals are given in Fig 3 and Fig S3. The high L/B ratios calculated for BIO-61 are a result of the low concentrations detected in the blubber of this animal where most compounds were only detected at a level close to the LOQ. Overall, most penta- and hexa-PCBs, nonachlors (TN and CN), PBDEs, *p,p'*-DDE and 2'-MeO-BDE 68 were found to accumulate preferentially in the blubber, while the chlordanes (TC and CC), HCB, hepta- and octa-PCBs accumulated preferentially in the liver.

Discussion

Legacy compounds in sediments and seagrass in Moreton Bay

Moreton Bay is one of the largest estuarine bays in Australia and lies between the mouth of the Brisbane River and large barrier islands, i.e. Moreton Island and Stradbroke Island (Fig S1) (Coates-Marnane et al., 2018). With the Brisbane River running through Brisbane, which is Australia's third largest city with 2.4 million inhabitants in 2018, as well as other waterways ending in Moreton Bay, pollutants from urban and agricultural sources are continuously flushed into the bay (Toms et al., 2006; Mueller et al., 2011; Duodu et al., 2017; Coates-Marnane et al., 2018). From a toxicological point of view, Sirenia are one of the least investigated groups among marine mammals (O'Shea et al., 2018). Like other marine mammal species, dugongs accumulate compounds primarily through their diet. Unlike other marine mammal species, dugongs are strictly marine and herbivorous, with a diet consisting mainly of seagrass and associated sediment (Tol et al., 2016; O'Shea et al., 2018). On one hand, these food types are low in the food web and are unlikely to accumulate lipophilic compounds to the same extent as prey species at higher trophic levels. On the other hand, they are only found in relatively shallow coastal and estuarine waters, bringing dugongs in close proximity to urban and agricultural pollutant sources (Haynes et al., 2000; McLachlan et al., 2001). Although both sediments and seagrass can contain pollutants depending on the physicochemical properties of the chemicals, only limited information is available for Moreton Bay. Haynes et al. (2000) could not detect PCBs and a range of organochlorine pesticides in seagrass and sediments in Moreton Bay. Other studies focused more on sediments only and showed that the concentrations of selected legacy pollutants (PBDEs, DDT) from the present study are low in sediments of Moreton Bay compared to sediments in the Brisbane River as well as sediments in similar catchments worldwide (Toms et al., 2006; Mueller et al., 2011). Declines in organochlorine pesticide concentrations in the Brisbane

River and Moreton Bay between 1998 and 2008 have been reported as well as almost 25 times higher levels of DDT in the mouth of the Brisbane River compared to more upstream locations (Mueller et al., 2011). However, in a more recent study, increases in sediment concentrations of several organochlorine pesticides were found after the floods of 2011 and 2013 (Duodu et al., 2017). These extreme weather events have a direct impact on dugong populations because of seagrass die-offs but potential indirect implications on the toxicological status of dugongs have not been assessed yet. To our knowledge, the present study is the first to analyse legacy compounds, such as PCBs and organochlorine pesticides in male dugongs from Moreton Bay in southeast Australia. The present study is also the first to analyse dugongs from after the floods (5 animals from 2011 and 2012) and to report on the presence of naturally produced MeO-PBDEs in Moreton Bay. While the sample size in the present study was relatively high compared to most other studies investigating Sirenia, the low numbers of animals in some groups have an influence on the robustness and reliability of the results and associated interpretations. Consequently, findings are presented as potential explanations that require further research with a larger sample size rather than straightforward conclusions.

Bioaccumulation of POPs in dugongs

Bioaccumulation patterns of POPs in marine mammals are often characterized by peak concentrations in juveniles followed by an overall decrease in concentrations in all animals due to a diet switch and a slower growth (Weijjs et al., 2010). Usually, this overall decline turns into a further stepwise decrease for females as they go through successive pregnancies in contrast to a gradual increase in concentrations for adult males (Weijjs et al., 2010). Some of these trends were also seen in the present study: the highest concentrations of POPs were detected in the juveniles and adult females exhibited decreasing POP concentrations with

body size (Fig 1 and S2). The more stable concentrations for all POP classes with body size in adult males, however, deviate from the expected patterns. There are several possible explanations for this finding, ranging from age-dependent modifications in the dugong's physiology to changes in environmental exposure concentrations. Changes in absorption or assimilation efficiencies and capacities for metabolic biotransformation of pollutants between young animals and adults have been reported for cows, humans and marine mammals (Ginsberg et al., 2004; Kierkegaard et al., 2009; Boon et al., 1987; McKinney et al., 2006; Routti et al., 2008). Both processes have also been suggested for dugongs, but only for PCDD/Fs (Haynes et al., 1999; McLachlan et al., 2001). Declining environmental concentrations in Moreton Bay could also be a potential explanation for the more stable concentrations found in the adult males. Mueller et al. (2011) reported decreasing pesticide (DDT, DDE, DDD) concentrations in sediments from Brisbane River from 1998-2008. However, increases in POP concentrations have also been reported after the floods in 2011 and 2013 (Duodu et al., 2017). An increase in PCB levels was also found in juveniles ($n = 2$) and an adult male ($n=1$) from 2011 and 2012 compared to dugongs from years before the floods (Fig 1). For the juveniles, the effect of the floods may contribute to the peak levels of PCBs to a larger extent than the typical bioaccumulation process in which the juveniles can have peak levels of selected POPs as a result of a prolonged lipid-rich milk diet. For the adult male, the effect of the floods may seem apparent as this animal had higher PCB levels compared to all other adult animals (Fig 1). However, only one adult male and two juveniles from the period after the 2011 floods was available for analysis thereby making it difficult to draw definitive conclusions. Furthermore, no statistically significant differences were found between adult male or female dugongs over a period of more than 10 years, including the male from after the floods. This suggests that the concentrations in dugongs change at a lower rate than environmental concentrations.

Profiles

The lack of information about the pollutant profiles in sediments and seagrass and about the dugong's potential capacity for metabolic biotransformation or breakdown makes it hard to interpret the profiles in these animals. In the majority of the samples, contributions of *p,p'*-DDE to the total sum were higher than contributions of PCBs. This is unusual compared to humpback dolphins (Weijs et al., 2016b) and green turtles (Weltmeyer et al., personal communication) from Moreton Bay where proportions of PCBs were consistently higher than *p,p'*-DDE. Green turtles are omnivorous and inhabit open waters in the first years of their lives after which they adopt a coastal lifestyle and switch to an herbivorous diet for the rest of their lives. Despite having a similar herbivorous lifestyle in Moreton Bay, dugongs seem to accumulate higher *p,p'*-DDE and lower PCB concentrations than green turtles. Potential explanations could be that dugongs are able to metabolise PCBs faster than green turtles or species-specific differences in absorption and excretion of PCBs and/or *p,p'*-DDE. However, there is currently not enough evidence for either suggestion.

Within each class, profiles resembled the patterns commonly found in most marine mammal species worldwide and more specifically in the humpback dolphins from Moreton Bay (Weijs et al., 2016b). *p,p'*-DDE was the only DDT-related compound that was found in the dugongs suggesting that there has not been any recent input of DDXs in Moreton Bay, confirming the findings of Mueller et al. (2011) in sediments and recent results in green turtles from Moreton Bay (Weltmeyer et al., personal communication). Although detected at lower concentrations than *p,p'*-DDE, both *p,p'*-DDD and *p,p'*-DDT were found in sediments and humpback dolphins from Moreton Bay indicating that these pesticides are present in the Moreton Bay ecosystem and available for accumulation by marine wildlife. Despite this, *p,p'*-DDD could

not be found in any dugong sample while *p,p'*-DDT was detected only in 4 blubber samples. This may mean that these compounds do not bioaccumulate readily as they were not detected in green turtles from the same region either (Weltmeyer et al., personal communication) but a capacity for metabolic biotransformation of some POPs in dugongs cannot be ruled out. Among PCBs, PCB 153 was the predominant PCB congener in all tissues followed by either PCB 138 or PCB 118, a common trend for marine mammals in general (e.g. Boon et al., 1997; Weijs et al., 2009; Storelli et al., 2012). With only 2 congeners detected of the lower chlorinated PCBs, dugongs seem to preferentially accumulate higher chlorinated PCBs (hexa to octa) (Fig 2), which likely reflects the persistence of the hexa- and hepta PCBs in the environment. Other explanations could be an ability to metabolically biotransform lower chlorinated PCBs by dugongs or a lack of lower chlorinated PCBs associated with seagrass. No specific pattern could be distinguished for the CHLs in the blubber samples, while *cis*-chlordane had the highest concentration in 9 out of 11 liver samples. This corresponds partly with the findings from the humpback dolphin study where *cis*-chlordane also had the highest concentration in liver, but the lowest concentration in blubber (Weijs et al., 2016b). PBDEs 47 and 154 were the only two PBDE congeners detected in the dugongs, in contrast with the humpback dolphins where 11 PBDEs were detected (Weijs et al., 2016b) as well as with a green turtle where only PBDEs 99 and 100 were found (Hermanussen et al., 2008). The results of the humpback dolphins (Weijs et al., 2016b), green turtle (Hermanussen et al., 2008) and sediments (Toms et al., 2006) show that PBDEs are bioavailable in Moreton Bay. Debromination (i.e. formation of lower brominated PBDEs from higher brominated PBDEs) and metabolic biotransformation (formation of hydroxylated PBDEs) have been suggested for some marine mammal species (McKinney et al., 2006) previously. However, differences in absorption between the species may also be an explanation for the different PBDE profiles found in tissues of the three species.

Tissue distribution

The liver and blubber are different in terms of composition as well as function, which translates into different affinities for chemicals. Understanding the fate of chemicals in an animal ideally requires a whole-body assessment. This can be done by developing toxicokinetic models, however, some information can also be gained from looking at partitioning processes. Sorted from low (HCB, 5.73; Hansch et al., 1995) to high (PBDE 154, 7.82; Braekevelt et al. 2003) log K_{ow} values as a proxy for a chemical's attraction to either a lipophilic (octanol) or hydrophilic (water) phase, there appears to be a shift with compounds accumulating preferentially in the liver when log K_{ow} values < 6 and in the blubber when log $K_{ow} > 6$ (Fig 3 and Fig S3). This shift is incomplete and clear relationships between L/B ratios and log K_{ow} values cannot be established as preferential accumulation in the liver of some animals (BIO-64, 59 and 329) was still reported for selected compounds with log $K_{ow} > 6$ (Fig 3 and Fig S3).

The results imply that there are no distinct differences between males and females in terms of where compounds are stored, which is confirmed by the similar profiles for males and females in blubber or liver (Fig 2). Nevertheless, concentrations in liver and blubber seem to be at equilibrium ($L/B = 1$) in males more than in females (Fig 3). This finding can be interpreted from a reproduction perspective as chemicals are likely more in a state of flux in adult females through regular offloading of compounds to offspring by gestation and lactation compared to males.

2'-MeO-BDE 68

Methoxylated PBDEs are structural analogues of PBDEs that are naturally occurring (Wiseman et al., 2011). In the present study, 6-MeO-BDE 47 and 2'-MeO-BDE 68 were

targeted with 2'-MeO-BDE 68 detected in 30 out of 33 samples and 6-MeO-BDE 47 only found in blubber in less than 50% of all samples. Both congeners have previously been analysed in marine mammals and are associated with specific sources according to the hypothesis raised by Vetter (2006), i.e. (red) algae for 6-MeO-BDE 47 and sponges for 2'-MeO-BDE 68. Results indicate that 2'-MeO-BDE 68 is preferentially stored in the blubber, which is in accordance with its high log K_{ow} value (Fig 3). There were no statistically significant differences in concentrations of 2'-MeO-BDE 68 between males and females (Fig S2), which suggests that 2'-MeO-BDE 68 is not readily passed on by the adult females to their offspring. In addition, the levels of 2'-MeO-BDE 68 seem to increase with body size in both genders showing that there is bioaccumulation with body size (proxy for age) rather than a stepwise decrease in concentrations in adult females due to offloading during and right after pregnancy (Fig S2).

Maternal offloading

Comparing the ratios between the blubber of adult females and the calf (C/F ratios) and the two juveniles (J/F ratios), shows that C/F ratios for 2'-MeO-BDE 68, hepta-PCBs and octa-PCBs are similar (0.58, 0.58 and 0.56, respectively) but much smaller than the C/F ratios for all other compound classes (ranging from 0.86 for *p,p'*-DDE to 1.66 for HCB). Likewise, J/F ratios for 2'-MeO-BDE 68 and octa-PCBs (2.86 and 1.04, respectively) are much smaller than those for all other POP classes (ranging from 9.25 for hepta-PCBs to 15.29 for PBDEs). As the calf and two juveniles in the present study are not associated with any of the adult females, these C/F and J/F ratios do not represent true matched mother/offspring pairs and should be interpreted with caution. Nevertheless, it has been shown previously that offloading of POPs from mothers to offspring is a selective process that favours lower chlorinated compounds (Debier et al., 2003). The results from this study confirm this principle and

suggest that 2'-MeO-BDE 68 is only transferred from mothers to offspring to the same limited degree as higher chlorinated PCBs.

Comparisons with marine wildlife in- and outside Australia

The contaminant body burdens in the dugongs were compared to other marine mammals (Table 2). The POP concentrations found in the dugongs in this study were lower than those in humpback dolphins from Moreton Bay. The largest difference was for PCBs, with the highest PCB concentration found in blubber of the dugongs in this study being almost 25 times lower than the lowest PCB concentration reported for the dolphins (Weijs et al., 2016b). Compared to other dugongs, PCB concentrations in dugongs from Moreton Bay were at the lower end of the range reported in dugongs along the Queensland coastline (Vetter et al., 2001). However, they were two orders of magnitude higher than those of a dugong from the Northern Territory (Gaus et al., 2005), up to 40 times higher than a dugong from the Torres Strait and more than 10 times higher than PCB concentrations reported in any Sirenia species worldwide.

Similarly, the concentrations of DDXs in the dugongs from the present study were lower than those in humpback dolphins from Moreton Bay (Weijs et al., 2016b) and dugongs from several locations across the Queensland coast (Vetter et al., 2001), but higher compared to DDX levels in Sirenia elsewhere. Concentrations of CHLs and HCB were also highest in the humpback dolphins, followed by the dugongs from Moreton Bay. Both compound groups could not be quantified in dugongs from Queensland (Vetter et al., 2001) or were not targeted in any other study (Table 2).

PBDE levels detected in the present study were more than 7 times higher than those previously reported in an adult female dugong from Moreton Bay (Hermanussen et al., 2008)

and an adult male dugong from Torres Strait. The latter animal also had lower concentrations of 2'-MeO-BDE 68 than the dugongs from Moreton Bay.

Generally, there is a lack of information about the concentrations of organohalogens in Sirenia worldwide and studies that are available usually have a much lower sample sizes compared to the present study. Nevertheless, the comparisons suggest that dugongs in Queensland, Australia, are exposed to higher concentrations of several types of legacy contaminants compared to dugongs from north Australia (Torres Strait, Northern Territory) or Sirenia anywhere else. Within Moreton Bay, the concentrations of any compound class investigated in the present study were higher than those reported previously for dugongs but much lower than those reported for humpback dolphins. The first finding illustrates that adult females are poor representatives of the population, while the second highlights the difference in bioaccumulation between piscivorous and herbivorous species.

Toxic effects?

The concentrations in dugongs reported here are several orders of magnitude lower than previously published PCB or DDT thresholds for reproductive effects and disease in various marine mammal species (Helle et al., 1976; DeLong et al., 1973; Aguilar and Borrell, 1994; Jepson et al., 2005; Mos et al., 2010). Although low concentrations may still be toxic to a sensitive species or may contribute to mixture toxicity (Altenburger et al., 2015), the concentrations of the compounds analysed here are unlikely causing adverse effects individually.

Conclusions

In the present study, POPs in dugongs were found at higher concentrations than those previously reported by studies that only investigated adult females and were comparable to or higher than previous results for Sirenia (dugongs or manatees) worldwide. In contrast, the results were several orders of magnitude lower than POP concentrations in humpback dolphins from Moreton Bay and toxicity thresholds established for several marine mammal species worldwide. Although the relatively low sample size ($n = 24$) warrants further research, our results illustrate that several principles for the bioaccumulation of POPs in piscivorous marine mammals also apply to dugongs. For example, adult females can selectively transfer a portion of their body burden to calves and juveniles through gestation and lactation. However, there are also important differences that should be investigated further. An example of this would be the potential ability of dugongs to metabolically biotransform some POPs. While a large part of the low POP concentrations can be explained by the dugongs' low trophic web position and the low concentrations of several lipophilic compounds in seagrass and sediments, there are some findings that may point towards metabolic biotransformation capacities in dugongs. For this reason, future recommendations are to analyse POP metabolites in tissues of dugongs, to investigate species with a similar diet (green turtles) or physiology (cetaceans) living in the same area, and to conduct toxicity tests as some metabolites are known to be more toxic than the parent compounds. Finally, fluctuations in sediment concentrations do not necessarily result in changes in the tissue concentration in dugongs, which appear to have stabilized in adult animals. This suggests that monitoring the environment may not give an accurate impression of tissue concentrations and temporal trends in wildlife and that changes in the environment need more than a decade to become visible in wildlife.

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Tables and Figures

Table 1. Summary of all contaminant concentrations found in dugongs from Moreton Bay and Torres Strait. Results are mean $\pm$ SD (median) and are expressed in ng/g lw. Detailed info and data for all individual dugongs can be found in Tables S1, S2 and S5 in Supporting Information. AF = adult females, AM = adult males, JM = juvenile males, C = calf.

	Blubber						Liver			
	AF	AM	JM	C	Unknown	Torres Strait	AF	AM	C	Unknown
n	8	7	2	1	2	1	4	4	1	1
Penta-PCBs	0.78 $\pm$ 0.58 (0.72)	1.25 $\pm$ 0.75 (1.33)	10.1 $\pm$ 1.15 (10.1)	1.01	2.53 $\pm$ 1.93 (2.53)	0.23	0.74 $\pm$ 0.34 (0.58)	1.62 $\pm$ 1.91 (0.83)	0.73	0.86
Hexa-PCBs	3.28 $\pm$ 1.94 (3.11)	4.88 $\pm$ 3.06 (3.80)	36.4 $\pm$ 3.13 (36.4)	2.61	9.60 $\pm$ 1.84 (9.60)	0.87	3.83 $\pm$ 2.48 (2.87)	8.11 $\pm$ 10.7 (3.40)	1.62	5.23
Hepta-PCBs	1.20 $\pm$ 0.93 (0.95)	1.68 $\pm$ 1.35 (1.01)	11.1 $\pm$ 5.02 (11.1)	0.69	2.60 $\pm$ 0.03 (2.60)	0.38	1.92 $\pm$ 1.65 (1.45)	3.23 $\pm$ 4.81 (1.06)	0.95	2.97
Octa-PCBs	0.09 $\pm$ 0.05 (0.08)	0.12 $\pm$ 0.09 (0.10)	0.94 $\pm$ 0.88 (0.94)	<0.10	0.09 $\pm$ 0.06 (0.09)	<0.10	0.17 $\pm$ 0.15 (0.14)	0.26 $\pm$ 0.36 (0.09)	<0.10	0.32
CHLs	0.92 $\pm$ 0.64 (0.80)	1.15 $\pm$ 0.60 (0.85)	13.4 $\pm$ 0.01 (13.4)	0.89	2.21 $\pm$ 1.64 (2.21)	0.58	1.05 $\pm$ 0.30 (0.95)	1.56 $\pm$ 1.52 (0.92)	1.30	1.08
HCB	0.61 $\pm$ 0.53 (0.51)	0.53 $\pm$ 0.25 (0.56)	5.69 $\pm$ 0.98 (5.69)	1.01	0.53 $\pm$ 0.42 (0.53)	1.50	1.29 $\pm$ 0.42 (1.32)	1.07 $\pm$ 0.69 (0.87)	2.29	1.10
<i>p,p'</i> -DDE	6.74 $\pm$ 4.79 (6.28)	13.0 $\pm$ 9.55 (13.3)	100 $\pm$ 22.6 (100)	5.78	24.5 $\pm$ 16.3 (24.5)	0.60	5.94 $\pm$ 3.49 (4.56)	15.8 $\pm$ 19.2 (8.22)	1.38	5.86
PBDEs	0.41 $\pm$ 0.23 (0.32)	0.48 $\pm$ 0.21 (0.50)	6.27 $\pm$ 4.47 (6.27)	0.66	0.50 $\pm$ 0.10 (0.50)	1.00	0.34 $\pm$ 0.14 (0.38)	0.60 $\pm$ 0.69 (0.32)	0.61	0.63
2'-MeO-BDE 68	2.49 $\pm$ 1.58 (1.93)	2.64 $\pm$ 2.79 (1.82)	7.11 $\pm$ 4.71 (7.11)	1.44	1.77 $\pm$ 0.78 (1.77)	1.18	1.06 $\pm$ 0.36 (0.99)	0.89 $\pm$ 1.31 (0.31)	0.32	0.47

Table 2. Comparisons between the concentrations reported here and results from the literature in Moreton Bay (all marine mammal species) and outside of Moreton Bay (all Sirenia). All results are in blubber unless specified otherwise and all concentrations are expressed in ng/g lw. Note that the number and types of PCB congeners can differ between studies. F = all females, NQ = not quantified due to low level or interference, ND = not detected.

Species	Location	Pollutant class	Min-Max	Reference
Moreton Bay	Dugong	PCBs	1.81 – 64.14	Present study
		CHLs	0.33 – 13.40	
		HCb	<0.10 – 6.39	
		<i>p,p'</i> -DDE	1.41 – 116.34	
		PBDEs	0.19 – 9.43	
		2'-MeO-BDE 68	NQ – 10.43	
	Humpback dolphin	PCBs	1,590 – 370,000	Weijs et al. (2016)
		<i>p,p'</i> -DDE	1,700 – 7,200	
		CHLs	174 - 386	
		HCb	9 - 17	
	Dugong (F)	PCBs	1.07 – 12.92	Vijayasathya et al. (In press)
	Dugong (F)	PBDEs	1.40	Hermanussen et al. (2008)
Australia	Dugong	PCBs	1.53	Present study
		CHLs	0.58	
		HCb	1.50	
		<i>p,p'</i> -DDE	0.60	
		PBDEs	1.00	
		2'-MeO-BDE 68	1.18	
	Dugong	PCBs	45.3 - 362	Vetter et al. (2001)
		DDXs	NQ - 173	
		HCb	NQ	
		CHLs	NQ	
	Dugong	DDE	0.3 – 15.0	Haynes et al. (2005)
		HCb	ND	
	Dugong	PCBs	0.49	Gaus et al. (2005)

Worldwide	Dugong*	Thailand	PCBs	3.60 – 4.04	Kumar et al. (2003)
	Manatee	Florida, USA	PCBs	ND	Ames and Van Vleet (1996)
	Dugong*	Sulawesi Island, Indonesia	PCBs	ND	Miyazaki et al. (1979)
			DDT	ND	
	Manatee	Florida, USA	PCBs	0.67 – 6.13**	O'Shea et al. (1984)
			DDXs	0.19 – 0.37**	

* Muscle tissue instead of blubber tissue

** Values recalculated from ppm ww to ng/g lw with a lipid percentage of 75%

Fig 1. Concentrations (log transformed) of hexa-, hepta-, and octa-PCBs with body size in blubber of female and male dugongs from Moreton Bay, Australia. The two animals with unknown gender, age (or body size) were not included in any statistical analysis and are therefore not plotted in these graphs. Similar graphs for other compound groups are in SI.

Fig 2. Proportion of each pollutant group to the total of all lipophilic organohalogen measured in the (A) blubber and (B) liver of females (all adults, first column) and males (adults in second column, juveniles and calves in third column). 'Unknown' category represents the animals of which age and gender are not known. 'TS Male' is an adult male from the Torres Strait ($n = 1$; northern part of Queensland). Error bars represent standard deviation (SD).

Fig 3. Heatmap showing the preferential accumulation of the individual compounds in the liver or blubber of the dugongs of which paired liver and blubber samples were available. Compounds are sorted by increasing log K_{ow} values. Ratios were calculated as concentration in liver divided by the concentration in blubber, both on a lipid weight basis (L/B) (no unit). There is no preferential accumulation in liver or blubber when $L/B = 1$ (black colour in heatmap). There is preferential accumulation in liver when $L/B > 1$ (red colours in heatmap) and in the blubber when $L/B < 1$ (blue colours in heatmap).

Highlights

- Dugongs from Moreton Bay (Queensland, Australia) were investigated
- Levels of all detected POPs were low (< 120 ng/g lw)
- Concentrations of POPs were below known toxicity thresholds for marine mammals
- Concentrations of POPs were higher or comparable to levels in sirenians worldwide