EXPOSURE AND POTENTIAL RISKS OF DIOXINS TO THE MARINE MAMMAL DUGONG

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Introduction

Atmospheric transport and subsequent deposition is considered a key pathway of PCDD/F input into marine systems. However, their strong affinity for particulates can result in significant sediment/soil associated PCDD/F transport from local emissions, in particular within riverine systems. In the subtropical/tropical coastal zone of Queensland, Australia, numerous large-scale tributaries feed into shallow embayments of the near shore marine environment. This environment sustains diverse and unique ecosystems associated with the world heritage Great Barrier Reef Marine Park. These include extensive seagrass habitats providing the food source for the only strictly herbivorous marine mammal dugong (*Dugong dugon*). Dugongs represent the last extant species in the family Dugongidae of the order Sirenia (the latter also including three species of manatee) and are listed as vulnerable to extinction by the IUCN Red List of Threatened Species. While Australia supports the most significant portion of the worlds remaining dugong populations, surveys have indicted a strong decline to only 3% of the population estimated in the 1960s[1]. Dugongs frequent coastal waters with habitats predominantly in wide shallow protected bays with a relatively small home range, varying from 5 to 64 km^2 [2]. Dugongs have a long life-span (~70 years), low reproductive rate and relatively long gestation and lactation periods. They are hindgut fermenters with highly specialized dietary requirements, uprooting whole seagrass plants and often selectively foraging on a few pioneer seagrass species[3]. Low metabolic rates, high seagrass consumption rates and inordinately long passage times of digesta through the gut (~6-7 days)[4], compensate for the low nutrient containing seagrass diet. Long life span, high fat repositories, narrow home ranges and highly specialized food requirements render the dugong potentially vulnerable to the regional PCDD/F contamination observed in Queensland. The present study aimed to establish baseline information for PCDD/F concentrations in dugongs from Queensland, to determine the exposure pathways involved and evaluate potential associated risks.

Materials and Methods

Blubber tissue from dugongs was obtained from animals found stranded within six regions along the coastline of Queensland and one region in the Northern Territory (Darwin). Throughout this paper, the regions in Queensland (Far North, Cairns, Hinchinbrook, Whitsundays, Hervey Bay and Moreton Bay), are referred to as Region 1 to 6 (respectively). Blubber samples were taken from the outer layer of blubber and together with previously reported data from dugong tissue[5, 6] a total of 7 adult males and 15 adult females were included in results and interpretation for this paper. The tissue was digested using 4 molar HCl at 60 °C and lipid content was determined gravimetrically. All samples were analysed for full PCDD/F profiles at ERGO Forschungsgesellschaft mbH in Germany using standardized techniques[5, 6].

For calculation of average daily intake (ADI), the following parameters have been used: (1) Dugong body weight: average 370 kg (320-470 kg) – based on records obtained for the animals of this study. (2) Seagrass consumption: average 35 kg wet weight (4 kg dry weight) per day – this is within the range of estimated consumption rates[2, 4]. (3) Sediment consumption: 2 kg dry weight per day. No information on sediment consumption rate exists, and the estimate was based on seagrass:sediment weight ratios determined from samples rinsed thoroughly in seawater[7]. (4) Total body lipid: average 35% (30-40%) of body weight – no records could be obtained for dugongs and the estimate was based on reports from other marine mammals. (5) Steady state conditions were assumed for estimation of half-life (T_2) using the equation $T_2=(\ln(2)*B_{Dug})/(f*I_{Dug})$[8], where B_{Dug} is the body burden of dugongs (ng kg^{-1}), f is the fraction absorbed (%) and I_{Dug} is the intake (ng kg^{-1} day^{-1}).

Results and Discussion

PCDD/Fs were detected in all dugong samples analysed (range ∑PCDD/Fs 225-2000 and 80-1200 pg g^{-1} lipid adult males and females, respectively). While considerable differences were present in dugong ∑PCDD/F concentrations from all regions (Coefficient of Variation (CV) = 104 and 79% males and females, respectively), variability was relatively low in animals from the same region, in particular among adult males (range CV = 2.2-25% and 27-50% males and females, respectively). This suggests that the PCDD/F exposure of dugongs from Queensland is governed by localized pathways. Differences in dugong PCDD/F concentrations between Queensland habitat regions were previously observed from initial data and were suggested to be correlated to the sediment OCDD contamination observed in seagrass habitats (OCDD sediment screening was previously established as a reliable proxy for ∑PCDD/F contamination in Queensland)[5]. The additional animals analysed for this study confirm a significant ($p<0.0005$) linear correlation between the ∑PCDD/F concentrations in dugongs and average OCDD concentrations in sediments from their habitats (Figure 1). Hence, the relatively simple OCDD screening method may provide a cost-effective initial strategy for dugong PCDD/F exposure assessment in Queensland. In general, PCDD/F body burdens of most, higher trophic, marine mammals are influenced by various uptake and transfer processes, including complex interactions within the food web, which often also encompass a wide spatial extent within the marine system. In the relatively regionalized, herbivorous dugong, most food web processes and large-scale spatial influences are removed. Hence, the PCDD/F concentrations in dugongs reflect the regional PCDD/F contamination of near shore marine sediments in Queensland. Preliminary results from other studies in Queensland suggest that the key input pathway for this contamination, in turn, is associated with sediment/soil transport via riverine runoff[9].

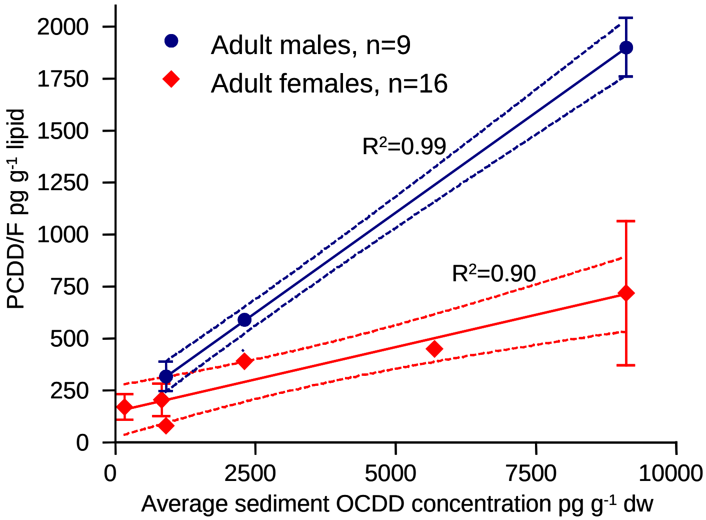

Figure 1. PCDD/F concentrations in adult male and adult female dugongs compared to the average sediment OCDD concentrations from different regions along the Queensland coastline. Error bars represent standard deviations and 95 percentiles are given above and below the calculated mean.

Highest PCDD/F concentrations were observed in adult male dugongs, and within a given region, concentrations were on average 3.4-fold lower in adult females (Figure 1). Lower concentrations in adult females, in combination with the higher variability described above in adult female compared to adult male dugongs within a region, is most likely due to the females' potential to eliminate PCDD/Fs via gestation and lactation. Therefore, while the PCDD/F concentrations of dugongs are a function of habitat contamination, the variation within a region, and importantly the slope of the function itself, is a reflection of dugong specific biological and toxicokinetic processes, such as PCDD/F absorption, metabolism and reproductive status.

Compared to other mammals, the PCDD/F concentrations in dugongs from most regions in Queensland exceed those reported in dugongs from Thailand and higher trophic marine mammals from locations remote to or with relatively low industrial activities and population densities such as the Arctic[10], New Zealand[11] and South Australia[12]. PCDD/F concentrations in dugongs from the highest contaminated Region 5, (average ∑PCDD/Fs 1900 and 718 pg g^{-1} lipid in adult male and females, respectively) exceed those reported in a range of higher trophic adult marine mammals near relatively polluted areas such as the Mediterranean, Baltic and Japan, and similar concentrations were reported for adult male killer whales off the British Columbian coast[13] (1250-2400 pg g^{-1} lipid). While the dugong is mostly removed from potential biomagnifications of lower chlorinated, more toxic PCDD/F congeners through the food chain (due to its herbivorous nature), and sediment TEQ levels are low (average 1.6, range 0.072-7.0 pg g^{-1} dw), PCDD/F concentrations expressed as TEQ in dugong blubber were surprisingly high (range 5-140 and 0.92-55 pg g^{-1} lipid in adult males and females, respectively) compared to those reported for higher trophic marine mammals elsewhere. Similar to ∑PCDD/F concentrations, TEQ levels in dugongs were highly dependant on habitat regions. However, it should be highlighted that, in contrast to many other marine mammals, PCB concentrations analysed from some dugongs in Queensland indicate that their contribution to the TEQ are relatively low[14].

The PCDD/F congener profiles in all adult animals were dominated by OCDD (average 50% of ∑PCDD/Fs), followed by 1,2,3,4,6,7,9-HpCDD (average 14%), whereas PCDFs only contributed on average 11% to the total ∑PCDD/F concentrations, and most PCDF congeners were present at levels below the limit of detection. In addition to the laterally chlorinated PCDD/Fs, non-2,3,7,8-substituted PCDD/Fs were present in dugong blubber, with remarkably high contributions within each homologue group, in particular the 1,4,6,9-substitued PCDD isomers (e.g. 33% for 1,2,4,7/1,2,4,8/1,3,7,8/1,4,6,9-TCDD, 8% for 1,2,4,6,9-PnCDD, 2.2% for 1,2,4,6,7,9/1,2,4,6,8,9-HxCDD) (Figure 2). Comparing the PCDD/F isomer distributions found in dugongs with those found in sediments and seagrass previously reported from near shore environments in Queensland, it was apparent that the patterns of non-2,3,7,8-substituted congeners in the animals follow closely those observed in sediments (Figure 2) and their food source, seagrass. While this contrasts the almost exclusive accumulation of laterally substituted congeners generally observed in mammals, the presence of non-2,3,7,8-substituted PCDD/Fs in dugongs likely represents the overriding contribution of these isomers in the food source, in combination with the dugongs low trophic position and high seagrass/sediment consumption rates, which may exceed its elimination capacities for non-lateral chlorinated PCDD/Fs. In fact, bioaccumulation factors (dugong to sediment plus seagrass concentration ratios) for all non-lateral substituted PCDD isomers were considerably lower (<1) compared to the 2,3,7,8-PCDDs (>30 for TCDD and PnCDD, 6-24 for HxCDDs, and 3 for HpCDD; (0.2 for OCDD)).

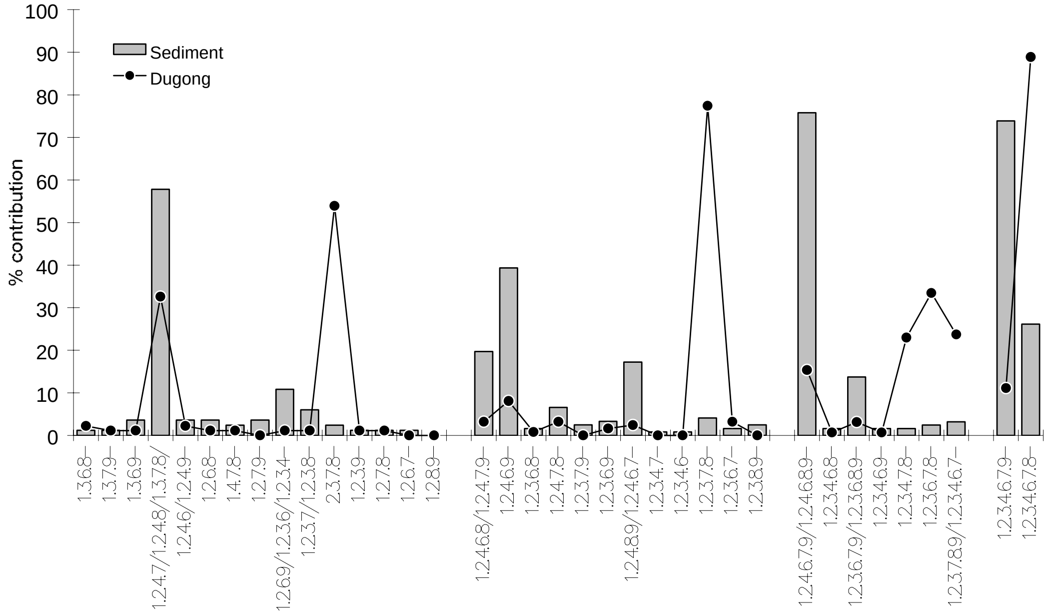

Figure 2. PCDD isomer distributions (% concentration of ∑homologue concentration) in a representative sediment and dugong blubber sample from Queensland.

The results of the present study suggest that exposure to PCDD/Fs by dugongs is related to the local contaminant concentrations in their inshore habitats, which in turn may be governed by terrestrial runoff and sediment transport within river plumes in Queensland[9]. Accumulation of sediment-bound PCDD/Fs in the near-shore coastal zone leads to elevated concentrations in the sediment-seagrass system. From sediments and seagrass, PCDD/Fs are redistributed to dugongs via preferred bioaccumulation (and selective transfer to offspring, unpublished data) of toxicological relevant PCDD/Fs. These results, together with laboratory and field evidence implicating toxic effects at relatively low levels in mammals, may represent risks for existing and future dugong populations. The results from the present study have been used to semi-quantify such risks. To accurately quantify risks, however, the sensitivity in dugongs would have to be determined. As these data are not available to date, it is highlighted that the following presents an estimate of potential risks in comparison to sensitive biological endpoints observed in other mammals.

Although the toxic effects of PCDD/Fs act over a common mediator (binding to the Ah-receptor), there are considerable species-specific differences in the responses elicited by these compounds[15]. In light of the unknown susceptibility of dugongs to PCDD/Fs, however, a conservative approach would be to expect that the endpoints for dugongs are at least comparable to those determined for experimental animals with respect to adverse health effects. Lowest dose (LOAELs), at which statistical significant effects on most sensitive endpoints were observed in mammals, have resulted in TEQ body burdens of 28 to 73 ng kg^{-1}. These effects include neurotoxic, reproductive, endocrine and immunotoxic responses[8]. Biochemical endpoints, observed at body burdens as low as 3 ng kg^{-1}, were not included in this assessment.

Average daily TEQ intake (ADI) of adult dugongs (calculated using the equation presented in materials and methods) was estimated at 3.1, 104, 42, 17, 170 and 15 pg kg-1 in the region 1 to six, respectively. These exposure levels resulted in average body burdens of 11, 3.1 and 47 ng kg-1 in adult males from region 3, 4 and 5, and 1,3, 4,0 and 1.5 ng kg-1 in adult females from region 1,2 and 6, respectively. Using the above LOAELs determined for other mammals (28-72 ng kg-1), and average daily intake and body burdens determined for the present study, the PCDD/F intake rate, at which adverse effects may occur in dugongs are in the range of 104 to 270 pg kg-1 day-1. Since the LOAELs are based on experimental animals, an uncertainty factor of 10 is commonly used to account for species differences in toxicokinetics. By applying the same uncertainty for dugongs, a TDI of 10 to 27 pg kg-1 day-1 was calculated. Dugongs from regions 2,3,and 5 exceed this TDI by 3.9-10, 1.6-4.2 and 6.3-17 fold, respectively.

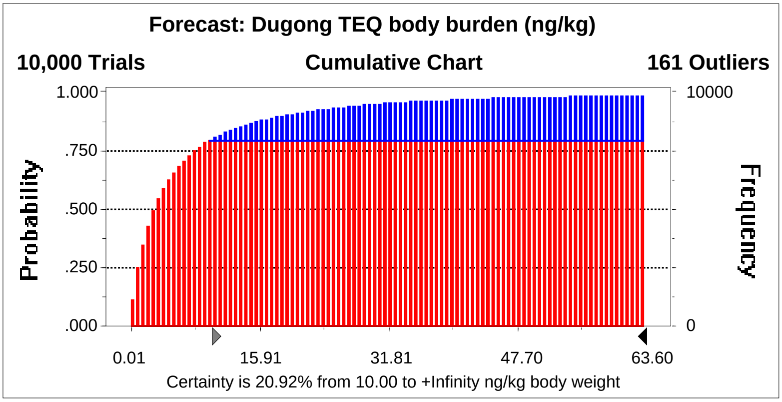

Figure 3. Probabilistic distribution of body burdens in the dugong population of Queensland. The blue area indicates the fraction of the population at or above the LOAEL of 10 pg/kg (immunotoxicity) observed in other mammals.

In order to provide an initial quantitative estimate of the risks to the dugong population of Queensland from exposure to PCDD/Fs, the probabilistic distribution of TEQ body burdens in adult dugongs (including males and females) was calculated. The calculation was based on the lognormal distributions of lipid TEQ concentrations and body weight observed for this study as well as an assumed lognormal distribution of body lipid content. The output distribution of TEQ body burden in dugongs is presented in Figure 3. Assuming that the data obtained for this study is representative for the dugong population of Queensland, and compared to the LOAELs above, up to 1.6-21% of the population are at or above the range of levels (LOAEL) where physiological effects have been observed in other mammals. It has to be noted, however, that this assessment is compromised by the lack of information on dugong specific sensitivities to dioxin-like compounds and actual risks may be lower (or higher). Nevertheless, the relatively high PCDD/F contamination of the dugongs' food source, its high seagrass consumption rates, preferred retention of toxicologically more relevant congeners, low elimination/high bioavailability and selective transfer to offspring, highlight the need for a detailed investigation of the sensitivity of dugongs to PCDD/Fs. This is of particular interest, since dugongs, due to their near shore habitats, are exposed to numerous other anthropogenic pressures, and an excess mortality of only 5% is considered unsustainable for these populations[16].

Acknowledgements
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