PCDD/F AND DIOXIN-LIKE PCB EXPOSURE IN A COASTAL COMMUNITY VIA CONSUMPTION OF LOCAL SEAFOOD

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Abstract

A national study highlighted that, similar to other countries, seafood contributes a major proportion to polychlorinated-*p*-dibenzo dioxin (PCDD), dibenzofuran (PCDF) and dioxin-like polychlorinated biphenyl (PCB) exposure of Australians. As typical for national studies, the exposure assessment utilised contaminant concentrations in retail (sea)food. However, more than 80% of the Australian population lives within 50km of the coast, where recreational and/or subsistence fishing of local seafood is prominent. This study assessed TEQ_{DP} exposure for a coastal subpopulation who consume locally caught seafood from an area with low (background) TEQ_{DP} levels in sediments, which is typical for Australian nearshore marine systems. Despite low sediment levels, a previous study showed that TEQ_{DP} concentrations in local seafood were 25 fold higher than retail seafood. This study showed average monthly contaminant intake for the coastal community (ranged between 34 (best case) to 107 (worst case scenario) pg TEQ kg^{-1} bw $month^{-1}$) was an order of magnitude higher than that estimated for the general population and was mainly driven by 2 to 6 fold higher seafood consumption rates. This highlights the need for information on seafood consumption patterns in coastal communities to better assess the contribution of locally sourced seafood to dietary PCDD/F and PCB exposure.

Introduction

In general, dietary intake contributes approximately 90% to the total human background exposure to polychlorinated-*p*-dibenzo dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (PCBs)[1]. The majority of this dietary exposure results from consumption of lipid rich products, including seafood. An Australian national dietary exposure assessment, conducted in 2004, estimated that the monthly dietary intake of PCDD/Fs and PCBs for the population aged over 2 years was 3.7 – 15.6 pg TEQ kg^{-1} bw $month^{-1}$ with seafood consumption contributing the majority (39%)[2]. The exposure assessment was based on a National Nutrition Survey which estimated monthly consumption of between 680 to 870g of seafood for individuals over 19 years of age[3]. Contaminant data was based on the analysis of 19 retail fish samples of unknown origin and 5 samples of canned tuna. A subsequent risk assessment found the mean and 95^{th} percentiles of the Australian population were below the JEFCA tolerable daily intake of 70 pg TEQ kg^{-1} bw $month^{-1}$, indicating the Australian general population has a very low risk from exposure to dioxins through food[2]. As this study was targeted towards the general population, no analysis was undertaken to account for population groups who may consume self-caught and more seafood compared to the national average, such as for example coastal Aboriginal and Torres Strait Islander populations, commercial and/or subsistence fishermen. It was, however, considered unlikely that such population subgroups would be significantly more exposed than the general population[2].

It is well known that sectors of community may be exposed to elevated levels of persistent organic pollutants if they regularly consume food from contaminated areas and/or species accumulating higher contaminant levels. For example, Arctic Indigenous populations have been shown to contain elevated PCB levels in blood serum due to subsistence on traditional foods that include higher trophic sea mammal species such as seal and polar bear[4,5]. Similarly, local commercial and recreational anglers from the PCB contaminated Great Lakes region have been reported to have elevated PCB serum levels compared to a reference group who consume little fish[6].

Despite the relatively low density of typical industrial PCDD/F and PCB point sources around Moreton Bay, Queensland, Australia, and the low TEQ_{DP} concentrations in its sediment, the median TEQ_{DP} level in fish was previously found to be approximately 25 fold higher compared to [7,8]. This indicates that retail fish do not accurately reflect the contaminant levels in seafood originating from Australian nearshore marine waters and are therefore not adequate for evaluation of human exposure via recreational or subsistence fishing. However, in the absence of information on consumption of locally sourced seafood by the Australian population, its contribution to PCDD/Fs and PCBs exposure cannot be estimated adequately. Hence, in a case study approach, a coastal (island) subpopulation in southeast Queensland, Australia was chosen to determine the consumption of locally sourced seafood and to evaluate the associated exposure to PCDD/Fs and PCBs.

Materials and Methods

For this study, two different survey methodologies were employed to estimate typical long-term patterns of seafood consumption by the adult population of North Stradbroke Island in Moreton Bay. A food frequency questionnaire was distributed to residents which presented a pre-determined list of Moreton Bay seafood species for which participants were asked to average consumption over the last year (n=197, response rate 39%). The results were then validated through fortnightly seafood diary records completed by a smaller subset of the community (n=33). For contaminant data, 110 samples from 23 seafood species (including commonly eaten fish parts and traditional seafood; dugong and turtle[9]) were analysed for PCDD/Fs and PCBs. All PCDD/F and PCB analysis was undertaken at Eurofins GfA in Hamburg, Germany as detailed previously[8].

Crystalball 2000 (Decisioneering Inc.) risk modelling software was used to combine the consumption survey information with contamination results. The software incorporates Monte Carlo probabilistic methods to predict and describe the exposure and estimate the proportion of the population at greater risk from exposure. Most input distributions for the probabilistic analysis were based on empirical data and where sample numbers were limited, distributional recommendations were adopted from similar scenario analyses in the literature. To assess exposure, average monthly dose of contaminants (Cm) (pg TEQ kg^{-1} bw $month^{-1}$) was estimated using the following model:

$$Cm = \frac{\sum_{i=1}^{23} CInt_i}{Bw}$$

where CInt is the total contaminant intake from ingestion of seafood species denoted i=1 to i=23 (TEQ pg $month^{-1}$) and Bw is bodyweight of the consumer adult (kg).

To measure contaminant intake (CInt):

$$CInt_i = Cn_i \bullet Ct_i$$
$$Cn_i = Cf_i \bullet Sw_i \bullet Sn_i$$

where Cn is the consumption amount of individual seafood species (g $month^{-1}$), Ct the toxic equivalent concentration for PCDD/Fs and PCBs (TEQ_{DP})[10] in the respective seafood species (pg g^{-1} fresh weight), Cf the seafood species consumption frequency (times $month^{-1}$), Sw the seafood species portion weight (grams) and Sn the species portion number consumed per meal. The model provides an estimate of monthly contaminant intake for individuals within the population. The resulting contaminant exposure estimate was then compared to the Joint FAO/WHO Expert Committee on Food Additives (JEFCA) guideline of 70 pg TEQ kg^{-1} bw $month^{-1}$ (tolerable monthly intake, TMI) to assess the proportion of the population who are potentially at a greater risk regarding adverse health effects from

PCDD/F and PCB exposure via seafood consumption[11]. The National Health and Medical Research Council of Australia has endorsed the use of the JECFA guideline as a regulatory guideline for risk assessment purposes[12].

Results and Discussion

In Moreton Bay seafood, TEQ_{DP} levels ranged from 8.6 to 190 (median 36; average 47) pg g^{-1} lw in fish, 36 to 39 (median 38) pg g^{-1} lw in squid, 38 to 600 (median 160; average 230) pg g^{-1} lw in crustaceans and 14 to 77 (median 24; average 29) pg g^{-1} lw in shellfish samples (Table 1). Despite the relatively low density of typical industrial PCDD/F and PCB point sources around south-east Queensland, the median TEQ_{DP} level in fish from this study is approximately 25 fold higher compared to recent Australian national data on retail seafood.

Table 1. PCDD/F TEQ and PCB TEQ range (and median) in pg g^{-1} lw (upper bound) and lipid content range (and median) for all seafood species.

	Bonito *Sarda sp*	**Flathead** *Platycephalus fuscus*	**Flounder** *Pseudorhombus jenynsii*	**Garfish** *Hemiramphus robustus*	**Mackerel** *Scomberomorus sp*	**Moses Perch** *Lutjanus russelli*	**Mullet** *Mugil cephalus*	**Snapper** *Pagrus auratus*	**Stingray** *Urolophidae sp*	**Summer Whiting** *Sillago ciliata*	**Tailor** *Pomatomus saltatrix*	**Yellowfin Bream** *Acanthopagrus australis*
	n=1	n=13	n=4	n=3	n=6	n=2	n=8	n=3	n=1	n=4	n=10	n=9
Lipid %	2.39	0.13-0.39 (0.29)	0.13-1.03 (0.7)	0.8-1.2 (0.9)	4.99-11.8 (7.35)	0.96-1.49 (1.23)	0.5-11.6 (2.67)	0.67-1.64 (1.08)	0.68	0.24-0.53 (0.36)	5.1-18.2 (11.2)	0.22-5.21 (1.02)
TEQ PCDD/Fs	18	5.8-53 (18)	17-48 (34)	7.0-9.3 (9.3)	6.3-42 (26)	30-39 (35)	7.3-74 (23)	17-81 (22)	87	4.4-24 (14)	6.5-59 (34)	9.8-80 (38)
TEQ PCBs	5.0	4.9-78 (17)	3.2-14 (7.7)	2.8-4.3 (3.3)	4.5-30 (21)	4.9-5.7 (5.3)	2.6-24 (8.3)	11-13 (11)	62	4.2-8.1 (6.4)	8.0-50 (20)	4.9-110 (14)
Total	**23**	**16-110 (35)**	**21-60 (43)**	**10-14 (12)**	**11-72 (46)**	**36-44 (40)**	**10-98 (34)**	**30-92 (33)**	**150**	**8.6-32 (21)**	**15-81 (54)**	**15-190 (55)**

	Pacific Oyster *Crassostrea gigas*	**Pearl Oyster** *Pinctada margaritifera*	**Pipi** *Donax deltoides*	**Squid** *Photololigo sp*	**Bay Prawn** *Metapenaeus macleayi*	**Banana Prawn** *Penaeus merguiensis*	**Tiger Prawn** *Penaeus esculentus*	**Mudcrab** *Scylla serrata*	**Sandcrab** *Portunus pelagicus*	**Dugong Blubber**[a] *Dugong dugon*	**Dugong Muscle** *Dugong dugon*
	n=2 pools	n=1 pool	n=4 pools	n=2	n=1 pool	n=1 pool	n=1 pool	n=4	n=2	n=3	n=2
Lipid %	1.7-2.8 (2.3)	0.64	0.81-1.75 (1.6)	1.87-2.0 (1.94)	0.98	0.78	0.31	0.13-0.32 (0.23)	0.23-0.33 (0.28)	37.3-62.1 (53.5)	1.5-10.6 (6.1)
TEQ PCDD/Fs	14-19 (16)	11	23-74 (24)	31-32 (32)	22	32	55	100-540 (190)	320-340 (330)	3.7-8.8 (8.2)	7.1-30 (19)
TEQ PCBs	2.2-2.7 (2.4)	2.8	1.1-3.4 (1.2)	5.3-6.8 (6.0)	16	19	5.0	7.8-60 (27)	32-34 (33)	0.61-1.1 (0.96)	1.7-3.5 (2.6)
Total	**16-21 (19)**	**14**	**24-77 (25)**	**36-39 (38)**	**38**	**51**	**60**	**130-600 (210)**	**350-370 (360)**	**4.3-9.9 (9.1)**	**8.7-34 (21)**

[a]one sample not analysed for mono-ortho PCBs

Of all food frequency questionnaires returned, 95% of respondents consume locally sourced seafood and the majority have lived on the island for 5 years or more (76%). When asked about their current pattern of seafood consumption compared to historical use, 75% consume about the same or more fish, 83% consume about the same or more crustaceans and 69% about the same or more shellfish. This indicates that the majority of consumers on the Island have retained similar consumption patterns over their years of residence.

Based on individual seafood species consumption in the food frequency questionnaire, the monthly average intake of seafood of the island community was 6.4 kg (median 4.5 kg; range 0.30 – 31 kg). This represents more than six fold higher consumption compared to the general Australian population. When compared to other dietary survey techniques, food frequency questionnaires tend to overestimate frequency of consumption due to the reliance on participant recall and their ability to correctly average consumption over a specified time period[13]. Hence, this study investigated seafood consumption using a second survey method (i.e. diary records, which is more time consuming and hence had a much lower participant rate). The diary records indicated that the total average seafood consumption may have been overestimated by up to a factor of three using the food frequency survey methodology. Although the diary records indicate a lower amount of consumption, the average monthly level recorded is still approximately two fold above that of the Australian general population.

Different exposure scenarios were calculated due to this uncertainty in consumption estimates. In a 'worst case' scenario, input distributions for consumption variables were developed directly from individual species estimates within the food frequency questionnaire. The results from the Monte Carlo simulation (10,000 trials) indicated that 44% of the seafood consuming population on the Island is exposed to PCDD/Fs and PCBs above the TMI of 70 pg TEQ kg^{-1} bw $month^{-1}$ (Figure 1). The average intake was 107 pg TEQ kg^{-1} bw $month^{-1}$ (median: 58; 95^{th} percentile: 362). As this scenario does not take into account uncertainty in relation to respondent recall, it is regarded as the 'worst case' scenario.

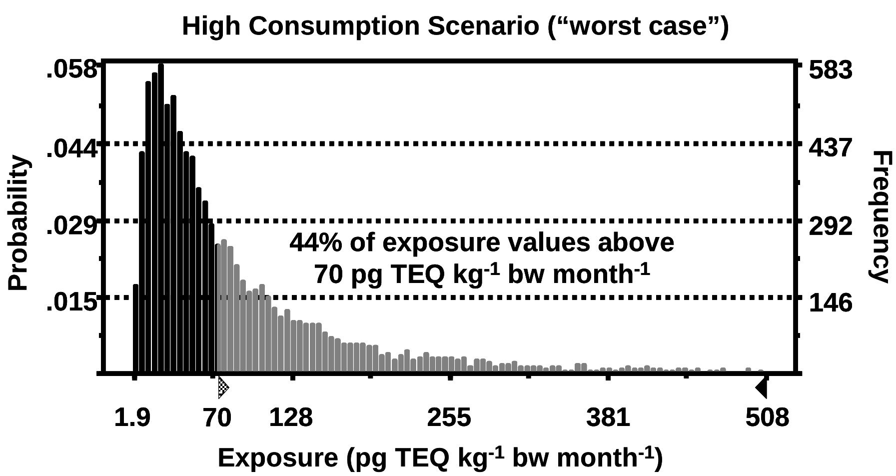

Figure 1. Probability chart showing exposure calculations (10,000 trials) for the coastal island community based on individual species consumption estimates from the food frequency questionnaire (worst case scenario).

For the low consumption scenario, consumption correction factors were applied by comparing average monthly consumption levels of seafood types between the diary records and the summed individual species estimates from the food frequency questionnaire. Application of the correction factors into the Monte Carlo simulation subsequently reduced the predicted proportion of the population above the TMI to 11%. The average intake was 34 pg TEQ kg^{-1} bw $month^{-1}$ (median: 20; 95^{th} percentile: 114).

The impact of each input variable on resultant contaminant exposure was quantified through a sensitivity analysis. Each variable was tested independently (over the range of its input distribution values) by holding all other variables static at their median value. Resultant exposure levels were plotted at nominated percentiles of the tested input distribution. The input variables which contribute most significantly to variance in the exposure forecast are related to consumption of seafood, in particular the fattier fish species tailor, mackerel and mullet (Figure 2). It is the upper tails of input distributions that have the greater impact with intake levels increasing from approximately the 70^{th} percentile. This indicates that higher consumption of these lipid rich fish considerably increases exposure levels.

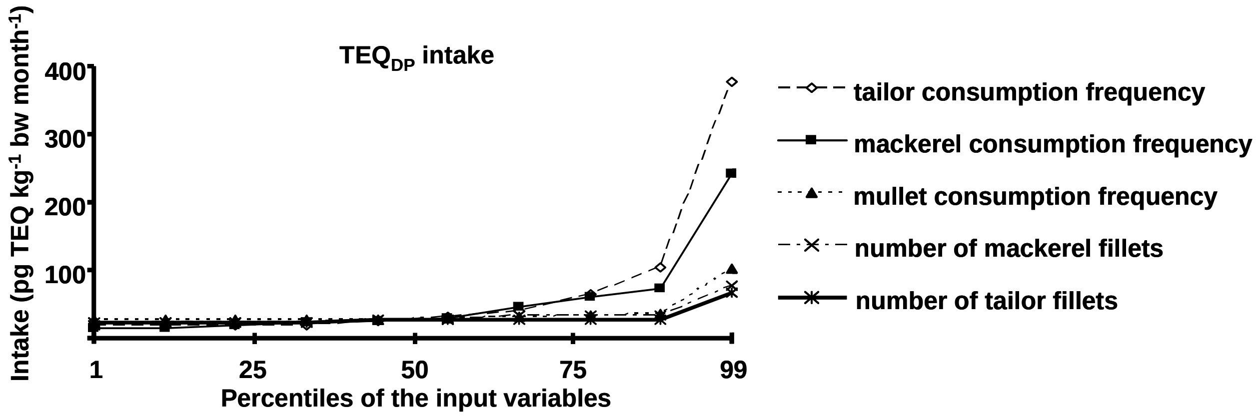

Figure 2. Sensitivity analysis showing top five input variables with largest effect on contaminant exposure.

This case study indicates that coastal subpopulations in Australia can be exposed to considerably higher levels of PCDD/F and PCB concentrations compared to the general population through local seafood sources, even in areas of relatively low TEQ_{DP} concentrations in sediments. This, combined with increased consumption of seafood by local communities, can result in exposure above the TMI in a relatively high proportion of such communities. The results from this study also highlight the inadequacy of utilising general consumption estimates and retail seafood contamination data for risk assessments considering subpopulations. Given that the majority of study participants have lived on the island for over 5 years and their pattern of seafood consumption has remained similar over this time, it is likely that the measured intake levels have been sustained over considerable periods and are relevant with respect to the chronic exposure safe intake guideline of 70 pg TEQ kg^{-1} bw $month^{-1}$. The study suggests that between 11% (low consumption scenario) and 44% (high consumption scenario) of community are above this guideline and risk management strategies which can effectively reduce contaminant intake while at the same time have low impact on the important and beneficial aspects of seafood consumption, would be appropriate for consideration in high level seafood consumers. The finding that consumption is driving elevated exposures demonstrates the need for better information on local seafood consumption patterns to better assess risk for coastal subpopulations.

Acknowledgements

The authors thank the Quandamooka community and John Page for provision of seafood samples, Jan Alderhoven and Peter Spinner for assistance with survey collection and Siobhan Hermanussen, Eva Holt and Sharon Grant for survey and laboratory assistance. Continuous support and professional advice from staff at Eurofins Gfa is gratefully acknowledged. This study is funded by the National Health and Medical Research Council (grant no: 351572). The National Research Centre for Environmental Toxicology is co-funded by Queensland Health Scientific Services (QHSS).

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