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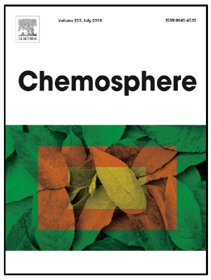

A Retrospective Investigation into the Occurrence and Human Exposure to Polychlorinated Naphthalenes (PCNs), Dibenzo-*p*-dioxins and furans (PCDD/Fs) and PCBs through cod liver products (1972 – 2017)

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A Retrospective Investigation into the Occurrence and Human Exposure to Polychlorinated Naphthalenes (PCNs), Dibenzo-*p*-dioxins and furans (PCDD/Fs) and PCBs through cod liver products (1972 – 2017)

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Abstract

A retrospective analysis of a number of historical medicinal grade cod-liver oil samples produced in Northern Europe revealed relatively high contamination levels of PCNs, PCDD/Fs and PCBs. The total toxic equivalence (TEQ) associated with PCDD/Fs, dl-PCBs and PCNs was in the range 95 to 427 pg g^{-1} for Baltic cod-liver oils and from 70 to 148 pg g^{-1} for oils sourced from the North Atlantic. The corresponding range for canned cod liver products (Baltic Sea) sampled in 2017 ranged from 52 to 104 pg g^{-1} fat (33 to 34 pg g^{-1} ww). The contribution from PCBs to the overall TEQ toxicity was around 3 to 6-fold higher than from PCDD/Fs and ranged from 24 to 318 pg TEQ g^{-1}ww. The estimated summed TEQ intakes of

PCDD/Fs, dl-PCBs and dl-PCNs resulting from the consumption of the daily recommended doses was highest for the Baltic cod-liver oils ranging from 16 to 293 pg kg^{-1} body mass (bm) day^{-1} for an adult, 20 to 183 pg kg^{-1} bm day^{-1} for a teenager and 15 to 131 pg kg^{-1} bm day^{-1} for a child. The contribution to daily adult TEQ intake from PCNs alone, although relatively small is estimated to contribute up to 5-fold above the recent EFSA proposed TWI of 2 pg kg^{-1} bm. The results indicate that although currently produced fish oils may undergo rigorous purification procedures and show low contaminant levels, cod livers sourced from the Baltic and consumed locally, continue to contribute substantially to the dietary intake of these contaminants.

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Highlights

Historical medicinal grade cod-liver oils showed high TEQ levels with PCBs>PCDD/Fs>PCNs

A cod liver oil from the Baltic Sea was most contaminated with a summed TEQ of 427 pg g^{-1}

Maximum estimated intake from recommended oil doses was 293 pg TEQ kg^{-1} bm day^{-1}

1. Introduction

Some polychlorinated dibenzo-*p*-dioxin (PCDD), -furan (PCDF), -biphenyl (PCB) and -naphthalene (PCN) congeners and their brominated- and mixed brominated-/chlorinated analogues are agonists of the aryl hydrocarbon receptor (AhR) in human cells and organs, particularly the liver (Behnisch et al., 2003; Blankenship et al., 2000; Falandysz et al., 2012b, 2014; Safe 1994; van den Berg et al., 2013; Wall et al., 2015; White and Birnbaum, 2015). Commonly referred to as dioxin-like effects mediated by the AhR, these contaminants elicit a range of toxic responses in organisms that are exposed in early-life. A number of responses are observed even at relatively low doses that correspond to concentrations in some items of the diet, although not all effects can be attributed to a dioxin-like pathway (Gregoraszczuk et al., 2016; Hansen, 1999; White and Birnbaum, 2015). The transgenerational toxicity of these contaminants in animals has been little studied so far, as subjects with a long life span including humans can be considered unsuitable for study. This consideration also includes epidemiological results (Baker et al., 2014) with very few accidental exposures of populations such as the Yusho and Yu-Cheng incidents (Li et al., 2013; Tsukimori et al., 2008) and the Seveso accident (EFSA 2018B), that are considered suitable or ethical to investigate.

Food, and particularly sea-food is a major source of human and wild-life exposure to halogenated persistent organic pollutants (POPs) on a global basis. Regional populations of top sea-bird predators have been decimated due to poor breeding success resulting from high body burden of PCBs, DDTs and PCDD/Fs, e.g. white-tailed sea eagle (*Haliaeetus albicilla*) nestlings in the south western region of the Baltic Sea in the 1970-1980s (Falandysz, 1984b and 1986a; Falandysz and Szefer, 1983; Falandysz et al., 1988, 1994a and 1994b; Kannan et al., 2003). Another example is the top sea mammal predator such as the killer whale (*Orcinus orca*) which is under threats of adverse health effects and population collapse from current levels of exposure and high PCB body burdens (Desforges et al., 2018). A general decrease in the trend of some chlorinated POPs including PCDD/Fs and PCBs has been observed in the global environment and foods in the last 2-4 decades as a result of regulation and cleaner waste disposal technologies, although the rate of decline for the regions of the Baltic Sea are rather low (Josefsson et al., 2018; Miller et al., 2014). See also in paragraph 3.4.

In earlier studies, several compounds such as hexachlorobenzene, hexachlorocyclohexanes, chlordanes (CHLs), cyclodiene insecticides (Aldrin, Dieldrin), DDT and metabolites and polychlorinated biphenyls (PCBs) were reported as common contaminants in foods such as canned cod (*Gadus morhua*) livers as well as cod-liver oil that was widely available for medicinal use: orally or topically (e.g. "Maść Tranowa" – oily ointment; 40% cod-liver oil; *Unguentum Olei Jecoris Aselli* FP VI). Among these contaminants, DDT/metabolites and PCBs in particular, were found to dominate the occurrence (Falandysz, 1981; Falandysz and Kannan, 1993 and 1994; Falandysz et al., 1992). Crude (unpurified from halogenated POPs) cod-liver oil that was sold in the past for medicinal use could also be contaminated with other highly toxic compounds such as PCDD/Fs, dl-PCBs or PCNs but there was very little good quality data available in the literature on the occurrence of the latter in cod-liver oil or canned cod liver products. Currently, cod-liver oil and fish oils are advertised widely as nutraceuticals and supplements rich in omega-3 fatty acids. Mothers are advised to feed babies of age > 6 months with cod-liver oil in dose 2.5 mL (2.1 g) daily (Apteka, 2019). It is not known if cod-liver oil was recommended or administered to babies in times before the era of the omega-3 fatty acids supplements.

In order to obtain an indication of past and recent human exposure from these sources, cod-liver oils produced in Iceland, Norway and Poland and available from 1972-2001 from European markets, and varieties of canned liver products such as cod liver and cod liver/vegetable pate - produced in February 2017 from Baltic Sea cod livers and retailed in

Poland have been examined for the occurrence of PCDD/Fs, dl-PCBs, ndl-PCBs and PCNs using sensitive, reliable and validated methods for food analysis (Fernandes et al., 2004; 2010).

As a historical note it is worth mentioning that although the products investigated here were freely available retail products, there are instances of limited production and distribution, e.g. small-scale production (up to one ton) of cod-liver oil products made in Gdynia, Poland, in 1993 and 2001 was restricted to employees (and probably close relatives etc.). Additionally, although retail cod-liver oil sold in Poland up to the mid 1970's was sourced from the Baltic sea, those produced in the 1950s and 1960s could also include oil from cod livers sourced from the North Sea.

In examining these exposures, it is worth noting that in light of the new epidemiological, experimental and modelling data on the toxicity of dioxins and dl-PCBs, and health concerns arising from the present dietary exposure to low levels of those compounds, the European Food Safety Authority (EFSA) has proposed a new tolerable weekly intake (TWI) of 2 pg TEQ kg^{-1} bodyweight. This proposed revision of the TWI is 7-fold lower than the previous TWI (EFSA 2018). The downward revision of the TWI clearly introduces major challenges - the measurement of these compounds at lower levels, better current insight into many aspects of food and environmental pollution by these contaminants regarding legacy and ongoing emission, and considerations for the prevention/reduction of exposure and the resulting health risk for humans and animals.

2. Materials and methods

2.1. Samples

Cod liver oil (tran) of medical grade sourced from the Baltic Sea or the North Atlantic (all stored all the time in a refrigerator) and canned cod livers (freshly bought food products) made from the Baltic Sea cod were obtained as follows:

a) Tran Leczniczy FP IV; Medicinal cod liver oil purchased in a pharmacy shop in Gdańsk, Poland (1972 – in original brown glass bottle; 100 mL),

b) Cod-liver oil (1993 and 2001) obtained from a processing plant (Zakłady Rybne) in Gdynia, Poland (1993 and 2001 – in brown glass bottles, 500 mL),

c) L-Cod Liver Oil; (Czysty Świeży Tran) oleum morhuae B.P.: (Contents 1 litre - in original can), donated by Red Cross, 1980,

d) M-Tran (Medisin Tran) purchased in a pharmacy shop in Norway (Contents CA 500 mL; original green glass bottle, 1982),

e) Two types of canned cod-liver products: "*Wątróbki rybne w tłuszczu własnym*" (Cod liver in its oil) and "*Pasztet z wątróbek dorszowych*" "" (Pate of cod livers and vegetables) produced in Łeba (Poland) in 2017; contents 150 g (Table 1).

2.2. Analysis

The analysis targeted the seventeen planar PCDD/F congeners, twelve dioxin-like (DL)PCBs (IUPAC Nos #77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 167,189), six non-dioxin-like (NDL) PCBs (IUPAC Nos #28, 52, 101,138, 153 and 180) and nineteen PCNs (PCN-13, 27, 42, 52/60, 53, 63, 65, 66/67, 64/80, 69, 70, 71/72, 73, 74 and 75).

The methods used for the extraction, purification and measurement of these analytes have been documented in full in several previous reports (Fernandes et al., 2004; 2010; 2016). Briefly, samples were internally standardised with ^{13}C-labelled analogues of target compounds (individual PCNs, PCDD/Fs and PCBs purchased as nonane solutions from Cambridge Isotope

Labs, Mass. USA or from Wellington Laboratories Inc. Ontario, Canada) and extracted by cold solvent extraction using a dichloromethane:hexane(40:60) mixture as described in Fernandes et al., 2004. *Ortho*-substituted PCBs were fractionated from non-*ortho* PCBs and PCDD/Fs on an activated carbon column. The two fractions obtained were further purified using activated alumina. For PCN analysis, PCNs were separated from PCBs by a similar fractionation on activated carbon. Congener separation, detection, confirmation and quantification was carried out using high-resolution gas chromatography/high-resolution mass spectrometry (HRGC-HRMS) at a resolution of 10,000 (Waters Autospec Ultima instrument fitted with a Hewlett Packard 6890N gas chromatograph) for the PCNs, PCDD/Fs, and non-*ortho* PCBs, and high-resolution gas chromatography/low resolution mass spectrometry (HRGC-LRMS) for the *ortho*-substituted PCBs (Fernandes et al., 2004). Toxic equivalents (TEQs) were calculated conventionally for PCDD/Fs and PCBs (van den Berg et al., 2013), for PCNs (Fernandes et al., 2010).

The analytical methods have been thoroughly validated and has been extensively used in other studies. Quality control criteria for all analytes was similar to regulated PCDD/Fs and PCBs measurements with the inclusion of a cod liver oil reference material (Fernandes et al., 2018) and procedural blanks which were evaluated prior to quantitation and reporting. The measured concentration of the reference material was within the established limits for all analytes. Typical limits of quantitation (LOQs) which are calculated by incorporating the levels detected in blanks as described in EC guidelines - European commission 2017) ranged from 0.01 to 0.1 pg/g for PCDD/Fs, dl-PCBs and PCNs depending on the congener, and 0.01 ng/g for ndl-PCBs. Analytical recovery typically ranged from 60 - 98% for PCDD/Fs and PCBs and 40 - 80% for PCNs. Further quality assurance measures included the successful participation in international inter-comparison exercises on PCDD/Fs and PCBs (Dioxins, 2018) over the study duration.

Measurement uncertainty (expanded uncertainty with a coverage factor of 2) estimates range from around 20% at ≥ 10x the limit of detection, to around 200% at the limit of detection.

2.3 **Intake estimates**

Practical estimates of daily consumption of cod-liver oil were based on prescribed or recommended doses. Historically, the volume of a daily oral dose for medicinal cod-liver oil prescribed as a source of vitamins (no less than 800 IU and vitamin D: no less than 80 IU in 1 g) in Poland was 5.0 to 15.0 g (FP IV, 1970). Other sources (Igya, 2019) prescribe a dose of one to two tablespoons given 1-2 times a day for adults and one tablespoon 1-2 times a day for children. (However in rare instances intake can be considerably higher as observed during the Tübingen punting race - a traditional boat race of student origin on the Neckar river in Tübingen, Germany. A half-litre of cod-liver oil had to be drunk before the eyes of the spectators, by each of the losing team members as well as those from barges disqualified for violation of the rules) (de.wikipedia, 2019).

It was estimated that the intake was in the range 1 to 4 tablespoons for an adult (body mass 70 kg), 1 to 2 tablespoons for a teenager (age 14, body mass 56 kg), and 1 to 2 teaspoons for a child (age 7, body mass 26 kg). Volumes of 15 mL (equivalent to 12 g) for a tablespoon and 5 mL (equivalent to 4 g) for a teaspoon were assumed. Daily intake was estimated as the product of the contaminant concentration and the quantity of oil or food consumed, divided by the appropriate body mass.

3. Results and discussion

Contaminant concentrations are expressed in conventional units (pg g^{-1} or ng g^{-1}) on a fat weight basis, except for the canned products which are given on a fat and whole weight (ww), (product) basis.

3.1. Absolute concentrations

All cod liver oils and canned livers examined were substantially contaminated with PCNs, PCDD/Fs and PCBs (Table 1). In general, the congener profiles reflecting this contamination were similar for all products and samples. Among the PCDD congeners, the 1,2,3,6,7,8-HxCDD and 2,3,7,8-TCDD dominated in the Baltic Sea cod-liver oils produced between 1993 and 2001 and in canned liver products, with 2,3,7,8-TCDD occurring at approximately half the concentration of 1,2,3,6,7,8-HxCDD. The profile of PCDDs in the Baltic Sea cod-liver oil produced in 1972 contrasted with the oil for later years but was comparable to that for the Atlantic (Iceland and Norwegian Sea) cod-liver oils for which both 2,3,7,8-TCDD and 1,2,3,6,7,8-HxCDD but also OCDD were the dominant congeners. 1,2,3,4,7,8-HxCDD was only detectable in the liver and vegetable pate sample which could suggest a non-cod-liver origin (Table 1). 2,3,7,8-TCDF, 1,2,3,7,8-PeCDF and 2,3,4,7,8-PeCDF were the dominant PCDF congeners in all cod livers products.

The congener profile for the NDL-PCBs seen in Figure 1 shows that PCBs #101, 138 and 153 were the main contributors to the NDL-PCB sum which ranged from 680 to 2393 ng g^{-1} fat for the Baltic cod-liver oils and from 494 to 714 ng g^{-1} for the North Atlantic oils. The NDL-PCB sum for the canned products were 277 to 619 ng g^{-1}. The data is not directly comparable to earlier reported Baltic Sea contamination levels (which were higher than the present study) as the historical PCB concentrations for cod-liver oils sourced from both the North Atlantic and the Baltic Sea were reported as the sum of all detected congeners, but PCBs-101, 138 and 153 were the major contributors in most cases (Falandysz et al., 1994b; 1994d). PCBs still remain the major chlorinated and most studied contaminant in the biota and sediments of the Baltic Sea. The legacy deposits are considered to be overlaid by more recent marine sedimentation but these concealed sources remain a potentially major extinction threat to top marine food-chain wildlife such as the killer whale but also humans (EFSA, 2018).

Concentrations for canned cod liver products in the present study are only around 3-fold lower in comparison to canned livers produced 27 years earlier in 1990 (Falandysz et al., 1992). Products from 1990 were also contaminated with DDT and its metabolites, DDTs (1,000 ± 140 ng g^{-1} ww) with lower levels of other organochlorine pesticides (Falandysz et al., 1993).

3.2. TEQs

In relation to the regulated limit (European Commission, 2011) for PCDD/F and DL-PCB TEQ in fish oils sold in the EU and even in relation to historical levels over the last 15-20 years (Fernandes et al., 2006), higher concentration were recorded in this study. PCDD/F TEQ in the Baltic Sea products ranged from 14.4 to 98 pg g^{-1} for cod-liver oils and from 12.3 to 15.0 pg g^{-1} in the canned liver products (4.8 to 7.7 pg g^{-1} ww). For the North Sea sourced oils, PCDD/F TEQ was 11.3 pg g^{-1} (Iceland) and 21 pg g^{-1} (Norway). The corresponding range for PCB TEQ was 79 to 318 pg TEQg^{-1} for Baltic cod-liver oils and from 38.5 to 87 pg TEQ g^{-1}fat for the canned products (24 to 28 pg g^{-1} ww). The PCB TEQ in the North Atlantic sourced oils ranged from 58 pg TEQ g^{-1} (Iceland) to 124 pg TEQ g^{-1} (Norway) (Table 1). In keeping with the observed TEQ profiles for fish and fish products (Fernandes et al., 2009) dl-PCB TEQ was considerably higher than PCDD/F TEQ. As observed elsewhere (Falandysz, 2003; Fernandes et al., 2017; 2018), the TEQ contribution from PCNs is generally lower than PCDD/F and PCB TEQ in marine products. In the current study the TEQ range arising from the most potent AhR active PCN congeners (Falandysz et al., 2019a), was 1.8 to 15.9 pg g^{-1} for the Baltic cod-liver oils; 1.3 to 2.3 pg g^{-1} in the canned liver products (0.8 pg g^{-1} ww), and from 1.2 pg g^{-1} (Iceland) to 2.6 pg g^{-1} (Norway) in the North Atlantic oils (Table 1).

TEQ arising from dioxin-like contaminants is considered to elicit a cumulative response (Fernandes et al., 2014) and although the PCN contribution to the summed TEQ is relatively small, it is significant, particularly when compared to the regulated limit for PCDD/F TEQ at 1.75 pg g^{-1}. The total TEQ of PCDD/Fs, dl-PCBs and PCNs was in the range 95 to 427 pg g^{-1} for Baltic cod-liver oils and from 52 to 104 pg g^{-1} in the canned liver products (33 to 34 pg g^{-1} ww). The values for the North Atlantic sourced oils were 70 pg g^{-1} (Iceland) and 148 pg g^{-1} (Norway).

3.3. Regulated Maximum levels (MLs)

The occurrence of PCDD/Fs and PCBs in cod liver oil and cod liver is regulated through the specification of MLs within the EU (EC, 2011). For fish liver, the limit for PCDD/F plus PCB TEQ stands at 20.0 pg g^{-1} ww, and at 200 ng g^{-1} ww for ndl-PCBs (EC, 2011). The combined PCDD/F and PCB TEQ values in processed cod livers in this study exceeded the ML by just over 1.5 fold (Table 1), while the concentration of ndl-PCBs was at, or below the ML (Table 2). However the MLs for fish liver oil are lower for PCDD/F-TEQ at 1.75 pg g^{-1} fat, rising to 6.0 pg g^{-1} when PCB-TEQ is included and 200 ng g^{-1} fat for ndl-PCBs. The PCDD/F TEQ values in the Baltic Sea cod-liver oils were eight to 56-fold higher than the ML, and six to twelve fold higher in the North Atlantic cod-liver oils. When combined with dl-PCBs, the TEQs for the Baltic Sea oils were 15 to 69-fold higher than the ML (12 to 24-fold higher for the North Atlantic cod-liver oils). In relation to the regulated combined (PCDD/Fs and PCBs) limit these values are very high at almost an order to an order and a half of magnitude, greater. PCN TEQ alone in the Baltic Sea oils ranged from just above, to up to 9-fold greater than, the ML for PCDD/Fs-TEQ (Table 1). Given the latest conclusion of the EFSA Panel that the existing TWI for PCDD/Fs and dl-PCBs should be revised downward by a factor of seven these findings indicate potential health concerns for those who consumed these products in the past.

The ndl-PCB concentrations of the cod-liver oils sourced from the Baltic Sea, North Atlantic (Iceland) and North Atlantic (Norway) exceeded the ML by 3.4 to 12-fold, 2.5-fold and 3.6-fold respectively.

3.4. Historical data on PCDD/Fs, PCBs, DDT and HCB

Although there is very little data on PCN concentrations in cod liver and cod liver oil sourced from North Atlantic regions, including the Baltic Sea. However, some historic data on other contaminants, such as PCDD/Fs, PCBs, DDT and HCB has been reported, and these have been summarised in Table 3 along with the data from the current study. The period covered by this compilation is contemporaneous with all but the most recent samples in this study, and literature references are provided in the right hand column. Much of the historic data up to the late 1980s covers PCBs, HCB and DDT, with PCDD/Fs being investigated in the more recent reports, but the differences in reported parameters underline the earlier comment about the difficulties in making comparisons between data sets. While there may be some indication of a declining trend in dl-PCB concentrations in the cod liver oil - 933 ng TEQ kg^{-1} in 1971 to 310 ng TEQ kg^{-1} in 2001, this is not reflected in either the PCDD/F TEQ (14 ng TEQ kg^{-1} in 1972 to 98 ng TEQ kg^{-1} in 2001) or the ndl-PCBs (3000 µg kg^{-1} in 1971 to 2400 µg kg^{-1} in 2001). In general, it would appear that the highest concentrations for PCDD/Fs and PCBs in cod liver oil from the Baltic Sea were observed during the 1980s unlike DDT and HCB where peak concentrations were seen in the 1970s. It is difficult to make a similar comparison for the cod liver as most of the reported data for PCDD/Fs and PCBs date to 2006 (Table 3).

3.5. Estimated intake

Based on these parameters, the daily dioxin-like TEQ intake arising from the consumption by adults, teenagers and children was calculated individually for PCDD/F TEQ, PCDD/Fs + PCB TEQ, PCN TEQ, PCDD/Fs + PCB + PCN TEQ.

The estimated daily intakes of PCDD/Fs + dl-PCBs + dl-PCNs-TEQ from the Baltic cod-liver oils were in the range 16 to 293 pg kg^{-1} body mass (bm) day^{-1} for an adult, 20 to 183 pg kg^{-1} bm day^{-1} for a teenager and 15 to 131 pg kg^{-1} bm day^{-1} for a child (Table 4). As observed earlier, PCNs make a smaller contribution, but the daily adult intake from these minor contaminants alone could contribute a maximum of up to 5-fold above the recent EFSA proposed (EFSA 2018) tolerable weekly intake of 2 pg TEQ kg^{-1} bm kg^{-1} week^{-1}.

TEQ intakes for PCDD/Fs + dl-PCBs + dl-PCNs resulting from a daily dose of cod-liver oils varied depending on the age, but nonetheless exceeded this TWI from 7.5 to 146-fold. For the oils sourced from the North Atlantic, intakes of PCDD/Fs + dl-PCBs + dl-PCNs-TEQ resulting from daily doses of the Norwegian oil would range from 25 to 101 pg kg^{-1} bm day^{-1}. In common with the intakes estimated for the Baltic Sea oils, the intakes of summed TEQ arising from consumption of these older cod liver oils sourced from North Atlantic, are high, either approaching or exceeding the recently proposed EFSA TWI.

Concentrations of the measured contaminants in the more recent (2017) canned cod liver products unsurprisingly show similar profiles to the oils as seen in Fig. 1 and Table 1. These products are currently still available in retail outlets in Poland and the profile of PCB occurrence reflects previous data reported 27 years ago (Falandysz et al., 1992 and 1993) highlighting the relatively high content of PCBs but also PCDD/Fs and PCNs in this foodstuff (Table 1). The estimated dietary intakes of all the measured contaminants arising from the consumption of a typical portions of these foods is shown in Table 4, but must be considered in combination with intake from the rest of the diet. Current data for Poland is not available but an indication of this contribution may be obtained from other European countries, e.g. adult consumption from the average UK diet was in the range 0.5–0.6 pg TEQ kg^{-1} bm day^{-1} (Mortimer et al., 2013).

Currently produced cod-liver oils undergo rigorous purification procedures which exclude most of these types of contaminants, and TEQ concentrations for PCDD/F + PCB TEQ in these oils is typically lower, e.g. of the order of 0.2 pg g^{-1} (PCN TEQ < 0.01 pg g^{-1}) (Fernandes, 2017). Clearly the intake of contaminants resulting from consumption of these oils would be expected to be much lower.

Work on this study will be ongoing with further dissemination on the profiles, the half-life of PCNs in Baltic cod and the contribution to dioxin-like TEQ from other similar contaminants (Falandysz et al., 2019a; 2019b), but many questions may remain unanswered. It is likely that in common with a number of European countries, cod-liver oil may have been administered to children, either medically prescribed, or provided as a general dietary supplement during the sampling period covered by the study. Any resulting long-term (18 to 47 years have elapsed since the sample collection dates) health effects that are attributed to dioxin-like toxicity (White and Birnbaum, 2009) may in many cases still have to manifest themselves.

4. Conclusions

Cod liver oils and cod liver products produced in Northern Europe over the the last 40-50 years covered by this study were found to show relatively high levels of PCDD/F, PCB and PCN contamination. Cod liver oils from the Baltic Sea were found to show higher levels of contamination than the North Atlantic sourced samples. The number of samples investigated during this study does not support the elucidation of trends or direct comparison between locations, but the similarity in profile and the continuing presence of raised contaminant levels in recent liver products from the Batic Sea indicate that although currently produced fish oils may undergo rigourous purification procedures to remove contamination, food produced from fish livers continues to contribute substantially to human exposure to these contaminants.

Disclaimer

The authors assert no conflict of interest.

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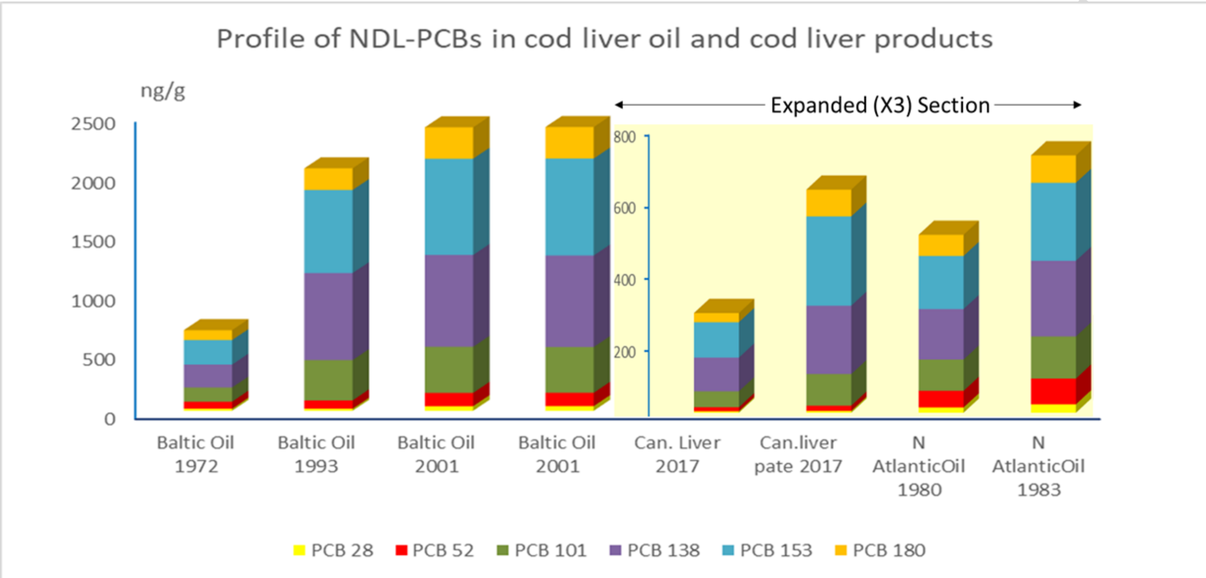

Figure 1. Profile of NDL-PCBs in cod-liver oil and cod liver products (in colors available on-line only).

Table 1. PCDD/Fs, dl-PCBs – non-*ortho* (pg g^{-1} fat), mono-*ortho* (ng g^{-1} fat) and TEQs of PCDD/Fs and dl-PCBs (pg g^{-1} fat) in cod-liver oil and canned liver products (ng g^{-1} fat // ng g^{-1} ww) made from livers of cod caught in the regions of the North Atlantic in 1972 – 2017

Compound	Region of the North Atlantic							
	Baltic Sea						North Atlantic - Iceland	North Atlantic - Norway
	Medicinal Tran (cod-liver oil)	Cod-liver oil (tran)	Cod-liver oil (tran)	Cod-liver oil (tran)	Canned liver products		Lysi Tran (cod-liver oil)	Möllers Tran (cod-liver oil)
	1972	1993	2001	2001	2017[A]	2017[B]	1980	1982
pg g^{-1}								
PCDD/Fs								
2,3,7,8-TCDD	3.68	11.04	20.29	21.42	2.5 // 2.2	3.51 // 1.13	2.64	6.39
1,2,3,7,8-PeCDD	1.21	9.68	13	12.2	1.7 // 1.5	1.22 // 0.39	1.03	1.7
1,2,3,4,7,8-HxCDD	< 0.05	< 0.05	< 0.11	< 0.08	< 0.08 // < 0.07	0.12 // 0.04	< 0.05	< 0.04
1,2,3,6,7,8-HxCDD	4.47	21.15	43.87	45.95	3.89 // 3.43	6.67 // 2.16	3.19	5.85
1,2,3,7,8,9-HxCDD	1.09	2.63	6.76	6.99	0.72 // 0.63	1.16 // 0.38	0.83	1.46
1,2,3,4,6,7,8-HpCDD	2.17	3.82	7.4	6.77	1.17 // 1.03	1.58 // 0.51	1.4	3.54
OCDD	4.52	3.39	2.44	2.59	0.46 // 0.4	0.94 // 0.3	3.94	31.4
2,3,7,8-TCDF	51.3	104.3	170.5	170.0	30.4 // 26.8	38.8 // 12.5	43.1	68.4
1,2,3,7,8-PeCDF	13.33	36.05	74.01	77.64	7.26 // 6.29	13.24 // 4.28	10.11	21.42
2,3,4,7,8-PeCDF	7.56	90.81	99.82	95.64	11.37 // 10.01	11.15 // 3.61	5.99	9.78
1,2,3,4,7,8-HxCDF	2.83	11.27	28.57	28.58	2.33 // 2.05	3.78 // 1.22	2.09	3.68
1,2,3,6,7,8-HxCDF	3.76	17.58	38.61	38.06	3.54 // 3.12	7.33 // 2.37	2.96	6.07
1,2,3,7,8,9-HxCDF	< 0.12	< 0.42	1.33	1.27	< 0.11 // < 0.1	0.3 // 0.1	< 0.07	< 0.25
2,3,4,6,7,8-HxCDF	4.24	12.16	30.32	29.99	3.22 // 2.84	6.68 // 2.16	2.94	6.25
1,2,3,4,6,7,8-HpCDF	1.56	2.41	8.75	9.2	0.68 // 0.6	1.75 // 0.57	1.19	3.32
1,2,3,4,7,8,9-HpCDF	0.25	0.37	1.51	1.38	< 0.13 // < 0.12	0.26 // 0.08	0.17	0.57
OCDF	0.42	< 0.23	0.72	0.89	< 0.12 // < 0.1	0.31 // 0.1	0.62	7.24
Non-*ortho* PCBs								
PCB 77	1001	2374	3673	3069	568 // 500	705 // 228	802	1311

PCB 81	50	63	173	182	16 // 14	22 // 7.0	40	72
PCB 126	691	2033	2747	2826	339 // 298	747 // 242	509	1118
PCB 169	133	501	555	540	86 // 76	261 // 85	98	176
Mono-*ortho* PCBs								
ng g^{-1}								
PCB 105	35.74	124.88	127.6	124.95	13.57 // 11.96	25.7 // 8.31	26.84	45.98
PCB 114	2.76	7.37	8.6	8.42	0.82 // 0.72	1.58 // 0.51	2.03	3.33
PCB 118	113	341	377	375	40 // 35	84 // 27	84	145
PCB 123	1.45	5.09	6.57	6.41	0.69 // 0.6	0.69 // 0.22	0.98	1.56
PCB 156	13.51	49.6	54.97	57.65	6.15 // 5.42	14.64 // 4.74	8.84	15.92
PCB 157	3.88	12.95	14.29	14.54	1.5 // 1.32	3.61 // 1.17	2.81	4.41
PCB 167	6.73	19.26	26.12	24.36	2.84 // 2.5	7.82 // 2.53	4.26	7.11
PCB 189	1.34	1.93	5.44	5.35	0.41 // 0.36	1.2 // 0.39	0.91	1.47
TEQ pg g^{-1}								
PCDD/Fs	14.4	66	98	97	12.3 // 10.8	15.0 // 4.8	11.3	21.0
PCBs	79	235	310	318	38.5 // 34	87 // 28	58	124
PCNs#	1.8	7.5	15.9	12.9	1.3 // 0.8	2.3 // 0.8	1.2	2.6
Total	**94.7**	**309.0**	**423.9**	**427.4**	**52.1 // 46**	**104.1 // 34**	**70.4**	**147.5**

Notes: A and B = two types of canned cod-liver products: "cod livers in own juice"[A] and "pate, cod liver & vegetables"[B] produced in the town of Łeba (Poland) in February 2017; #Data from Falandysz et al., (2019a)

Table 2. Indicative (ICES-6) PCBs in cod-liver oil (ng g^{-1} fat) and canned liver products (ng g^{-1} fat // ng g^{-1} ww) made from livers of cod caught in the regions of the North Atlantic in 1972 – 2017

	Region of the North Atlantic							
	Baltic Sea						Lysi Tran (cod-liver oil)	Möllers Tran (cod-liver oil)
	Medicinal Tran (cod-liver oil)	Cod-liver oil (tran)	Cod-liver oil (tran)	Cod-liver oil (tran)	Canned liver products		Lysi Tran (cod-liver oil)	Möllers Tran
Compound	1972	1993	2001	2001	2017[A]	2017[B]	1980	1982
PCB 28	17.9	17.9	39	39	4.2 // 3.7	4.9 // 1.6	14.6	23
PCB 52	59	70	112	112	11 // 9.7	14.7 // 4.76	46	72
PCB 101	120	340	389	386	44 // 39	88 // 28	87	117
PCB 138	193	734	774	772	94 // 82.5	190 // 61	140	210
PCB 153	206	700	811	817	98 // 86.5	248 // 80	147	216
PCB 180	84	182	265	266	25 // 22.4	74 // 24	59	76
Sum	680	2044	2390	2393	277 // 244	619 // 200	494	714

Notes: A and B = two types of canned cod-liver products: “cod livers in own juice”[A] and “pate, cod liver & vegetables”B produced in the town of Łeba (Poland) in 2017.

Table 3. Literature data on contamination with PCDD/Fs, PCBs and some pesticides of cod livers and cod liver products (data adapted, respectively, all values rounded)

Product and place of origin	Year(s) and	PCDD/Fs TEQ ng kg^{-1} ww	dl-PCBs TEQ ng kg^{-1} ww	Σ ndl-PCBs µg kg^{-1} ww	Total PCBs µg kg^{-1} ww	DDTs µg kg^{-1} ww	HCB µg kg^{-1} ww	Ref.
Cod livers (fresh)								
Baltic Sea, western areas	1973; n = 32				11000 (2000 – 53000)	5300 (930 – 53000)	220 (10 – 1600)	No 76
Baltic Proper	1974; n = 2x0.5 kg				13000	7500		ICES 77
Baltic Proper	1976; n = 10				13000 (9200 – 21000)	10000 (5900 – 17000)		Hu 77
Baltic Proper	1976-1977; n = 100				10000 (9200 – 11000)	19000 (10000 – 27000)		Lu 78
Baltic Sea, Gulf of Gdańsk	1981; n = 158				4000 ± 600 – 51000 ± 21000		30 ± 5 – 120 ± 40	Fa 83
Baltic Sea, southern part	1981; n = 471				4600 ± 1100 – 31000 ± 13000	1200 ± 200 – 18000 ± 6000	190 ± 5 –130 ± 20	Fa 84a
Baltic Sea, southern part	1983; n = 210[a]				7200 (3600 ± 400 - 9800 ± 1400)	2600 (1500 ± 100 –3400 ± 300)	96 (28 ± 15 – 170 ± 20)	Fa 86
Baltic Sea, southern part	2006; n = 3(19)	9.2 (7.3 – 10.	80 (70 – 83)					Karl 09
Baltic Sea, western part	2006; n = 5(33)	8.7 (6.5 - 15)	69 (56 - 96)	670 (480 - 1050)				Karl 16
Baltic Sea, Gulf of Gdańsk	2006; n = 1(4)	8.2	60					Karl 09
Baltic Sea, Lithuania	2006; n = 1(8)	14	71					Karl 09
Baltic Sea, central region	2006; n = 3(46)	15 (14 – 18)	77 (61 – 115)					Karl 09
Kattegat	2007; n = 2(24)	9.3 – 16	64 – 96					Karl 09
Bay of Kiel	2007; n = 1(14)	12	112					Karl 09
North Sea	2007; n = 3(15)	14 (10 - 18)	68 (34 – 100)					Karl 09
North Sea	2007-09; n =5(20)	9.5 (2.0 – 17)	29 (7.5 - 93)	300 (53 – 760)				Karl 16
Southern Norway	2009-10; n = 5(22)	6.5 (5.8 – 7.7)	29 (11 - 38)	280 (95 – 830)				Karl 16
Northern Norway	2010-12; n = 7(56)	2.7 (1.3 – 4.1)	11 (5.7 – 16)	230 (11 – 32)				Karl 16
Barents Sea	2010-12; n = 9(82)	1.1 (0.90 – 2.1)	5.1 (4.0 – 8.9)	47 (36 – 73)				Karl 16
Greenland	2006; n = 3(15)	0.5 (0.4 – 0.7)	2.9 (2.0 – 3.3)					Karl 09
Greenland	2006-10; n = 7(35)	0.90 (0.37 – 1.4)	3.0 (1.4 – 4.1)	37 (26 - 41)				Karl 16
Canned cod liver products								
Baltic Sea, Poland	1993; n = 3		370 - 460	740 (500 – 900)	2000 ± 400 (1200 – 2600)	1000 ± 100	50 ± 6	Fa 93, Fa 92
Baltic Sea, Poland	2017; n = 2	4.8 - 11	28 - 34	200 - 240				C.Study
Cod liver oil (medicinal)								
Baltic Sea	1971; n = 1		933	3000	8000			Fa 94, 94c
Baltic Sea	1972; n = 1	14	79	680				C.Study
Baltic Sea	1975; n = 1		880	2450	6600			Fa 94, 94c
Baltic Sea	1980; n = 1		2300	6700	17000			Fa 94, 94c
Baltic Sea	1985; n = 1		1200	2900	8000			Fa 94b,c
Baltic Sea	1986; n = 1				10000			Fa 94
Baltic Sea	1971-80; n = 10				14000 (9100 – 18000)	16000 (9400 – 25000)	370 (290 – 460)	Fa 94d
Baltic Sea	1981-89; n = 10				10000 (7000 – 14000)	6400 (3100 - 9000)	260 (170 – 340)	Fa 94d
Baltic Sea	1989; n = 1		1200	3100	9500			Fa 94, 94c
Baltic Sea	1993; n = 1	66	235	2000				C.Study
Baltic Sea	2001; n = 2	97 - 98	310 - 318	2400 - 2400				C.Study
North Sea	1982; n = 1				4600	1700	130	Fa 94, 94d
Norwegian Sea	1980; n = 1	21	120	710				C.Study
Norwegian Sea	1982; n = 1				5000	1400	110	Fa 94d
North Atlantic, Iceland	1980; n = 1	11	58	490				C.Study
North Atlantic, Iceland	1984; n = 1				2000			Fa 94
North Atlantic, Iceland	1984-1987; n = 8				1900[b]	860 (650 – 950)	87 (73 – 100)	Fa 94d

Notes: [a]Length class 40 – 50 cm; [b]n = 1; Σ ndl-PCBs – Sum of PCBs# 28, 52, 101, 138, 153 &180; Ref., respectively: Norén and Rosén, 1976; ICES Coop Res. Rep., 1977; Huschenbeth, 1977; Luckas et al., 1978; Falandysz, 1983,1984a, 1986; Karl and Lahrssen-Wiederholt, 2009; Karl et al., 2016; Falandysz et al., 1993; C.Study (Current study); Falandysz et al., 1994b, 1994c, 1994d.

Table 4. The estimated adult and children oral intakes of TCDD TEQs of PCDD/Fs, dl-PCBs, dl-PCNs and iPCBs arising from the consumption of a typical portion of examined cod-liver oils daily or intakes arising from the consumption of canned livers products

Parameters	Product intake (g)	PCDD/Fs		PCDD/Fs + dl-PCBs		dl-PCNs		PCDD/Fs + dl-PCBs + dl-PCNs	
		TEQ pg	TEQ pg kg^{-1} bm	TEQ pg	TEQ pg kg^{-1} bm	TEQ pg	TEQ pg kg^{-1} bm	TEQ pg	TEQ pg kg^{-1} bm
Cod-liver oil									
Baltic Sea - Poland									
Adult (70 kg bm)	12	173 - 1176	2.5 - 17	1121 - 4980	16 - 71	21.6 - 191	0.31 - 2.7	1136 - 5129	16 - 73
- " -	24	346 - 2352	4.9 - 34	2242 - 9960	32 - 142	43.2 - 382	0.62 - 5.4	2273 - 10258	32 - 146
- " -	48	691 - 4707	9.9 - 67	4483 - 19920	64 - 285	86.4 - 763	1.2 - 11	4546 - 29615	65 - 293
Teen age 14 (56 kg bm)	12	173 - 1176	3.1 - 21	1121 - 4980	20 - 89	21.6 - 191	0.39 - 3.4	1136 - 5129	20 - 92
- " -	24	346 - 2352	6.2 - 42	2242 - 9960	40 - 178	43.2 - 382	0.77 - 6.8	2273 - 10258	41 - 183
Child age 7 (26 kg bm)	4	57.6 - 392	2.2 - 15	374 - 1660	14.4 - 63.8	7.2 - 63.6	0.28 - 2.5	379 - 1710	15 - 66
- " -	8	115.2 - 784	4.4 - 30	747 - 3320	13.3 - 128	14.4 - 127	0.54 - 4.9	758 - 3420	30 - 131
Atlantic - Norway									
Adult (70 kg bm)	12	252	3.6	1740	25	31.2	0.45	1770	25
- " -	24	504	7.2	3480	50	62.4	0.89	3540	51
- " -	48	1008	14.4	6960	99	125	1.8	7080	101
Teen age 14 (56 kg bm)	12	252	4.5	1740	31	31.2	0.56	1770	32
- " -	24	504	9.0	3480	62	62.4	1.1	3540	63
Child age 7 (26 kg bm)	4	84	3.2	580	22	10.4	0.40	590	23
- " -	8	168	6.5	1160	45	20.8	0.80	1180	45
Atlantic - Iceland									
Adult (70 kg bm)	12	136	1.9	832	12	14.4	0.21	845	12
- " -	24	271	3.9	1663	24	28.8	0.41	1690	24
- " -	48	542	7.8	3326	47	57.6	0.82	3379	48
Teen age 14 (56 kg bm)	12	136	2.4	832	15	14.4	0.26	845	15
- " -	24	271	4.8	1663	30	28.8	0.51	1690	30
Child age 7 (26 kg bm)	4	45.2	1.7	277	11	4.8	0.18	282	11
- " -	8	90.4	3.5	554	21	9.6	0.37	563	22
Canned cod livers									
Adult (70 kg bm)	105 g	504 - 1134	7.2 - 16.2	3444 - 4704	49 - 67	84 - 84	1.2	3570 - 4830	51 - 69
- " -	150 g	720 - 1620	10.3 - 23.1	4920 - 6720	70 - 96	120 - 120	1.7	5100 - 6900	73 - 99
Teen age 14 (56 kg bm)	52 g	250 - 562	4.5 - 10.0	1706 - 2330	30.5 - 41.6	41.6 - 41.6	0.74	1768 - 2392	32 - 43
- " -	75 g	360 - 810	6.4 - 14.5	2460 - 3360	43.9 - 60.0	60 - 60	1.0	2550 - 3450	45 - 62
Child age 7 (26 kg bm)	26 g	125 - 281	4.8 - 10.8	853 - 1165	32.8 - 44.8	20.8 - 20.8	0.8	884 - 1196	34 - 46

- " -	37 g	130 - 400	5.0 - 15.4	1214 - 1658	46.7 - 63.8	29.6 - 29.6	1.1	1258 - 1702	48 - 65