4.5 Estimation of human exposure to halogenated contaminants from wild and farmed fish, shellfish and shrimp

Abstract
Fish and fish products contribute substantially to the human exposure to organohalogenated contaminants. In recent years, several organohalogen contaminant surveys on wild and farmed fish consumed in The Netherlands were carried out. This resulted in a substantial amount of data on the concentrations of polychlorinated dibenzo-p-dioxins and -furans (PCDD/Fs), polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB), p,p’-DDT, -DDD and -DDE, polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD) and perfluorooctane sulfate (PFOS). These datasets are integrated and combined contaminant exposures are estimated. The objective of this study was to specify (i) the main contributing fish species to the human exposure in The Netherlands; (ii) the main contributing contaminants, and (iii) to determine the contribution of recently introduced farmed species like pangasius and tilapia. The exposure is dominated by PCBs (sum of seven indicator congeners) and PFOS. PFOS shows a distinct exposure pattern as compared to the other contaminants (e.g. because there is no contribution from salmon). From a species point of view, herring and farmed salmon are the main contributors to the contaminant exposure from fish, followed by cod, plaice and mussels. The contribution of farmed tilapia, pangasius and shrimp was very low (<1% for all species).

Introduction
The human exposure to organohalogenated contaminants has been the subject of several studies. Often individual contaminant groups (e.g. polychlorinated dibenzo-p-dioxins and -furans (PCDD/Fs), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs)) were studied, but in some cases multiple contaminant groups were investigated in food basket studies and studies on individual food items (e.g. shellfish). Many exposure studies point towards fish and shellfish as the important sources for human exposure to contaminants (1,2). In The Netherlands, dairy is the predominant source for exposure to PBDEs and PCDD/Fs and dioxin-like PCBs (dl-PCBs) because dairy consumption is high (3,4). Fish is also an important source of exposure. However, in spite of the presence of these contaminants, fish consumption is promoted because it contains selenium and unsaturated (omega-3 and 6) fatty acids which are believed to be beneficial for human health (5,6).
The fish and shellfish we consume is a very heterogeneous group. They originate from different waters (e.g. freshwater, brackish, marine), have different positions in an aquatic food chain (pelagic or benthic prey or predator) or may be farmed. Fish feeds may contain high amounts of marine proteins and lipids, or may consist of vegetable ingredients. All these variables determine to what extent fish is contaminated with organohalogenated contaminants. In several studies, exposure assessments were carried out on single contaminants (e.g. PBDEs or hexachlorobenzene (HCB) or PCDD/Fs and dl-PCBs (2,4,7,8)), or a few contaminant groups (1,9). In a Swedish study the exposure from fish decreased in the following order: PCBs (349 ng/day), DDTs (256 ng/day), chlordane (87 ng/day), HCB (36 ng/day), hexachlorocyclohexane (35 ng/day), PBDEs (23 ng/day) and total-TEQ (31 pg/day) (1). These exposure calculations were based on 13 fish samples. In the present study the dietary exposure from fish to multiple contaminant groups (i.e. PCDD/Fs, PCBs, HCB, DDTs, PBDEs, hexabromocyclododecane (HBCD) and perfluorooctane sulfonate (PFOS)) was estimated for The Netherlands. In recent Dutch surveys and monitoring studies edible wild fish species such as eel, herring, mussels, cod, plaice, tuna, pike-perch and sole and several farmed species including salmon, eel, trout, pangasius, tilapia and shrimp were investigated for the above mentioned contaminants. The availability of these data allows an integrated exposure analysis of several contaminants based on a diverse group of fish species.

Materials and methods

The human exposure to contaminants is determined by combining information on contaminant concentrations in fish with information on the consumed quantities of fish. The contaminant data was taken from several studies (see Table 4.14). In most studies the same species were analysed. Therefore, we have collected data for all contaminants in cod and related species (Gadidae family), plaice, sole, wild eel and farmed eel, mussels, wild and farmed shrimp, herring, mackerel, sole, plaice, salmon, trout, pangasius and tilapia. Pike-perch and tuna data were not available for all contaminants and were therefore left out in this study. An overview of the contaminants studied is presented in Table 4.14. As many contaminant concentrations were rather low, detection and quantification limits (LOD and LOQ, respectively) could have a substantial influence on the final result. Therefore, the selection of contaminants in this study was limited to those of which considerable numbers of concentrations were above the LOD/LOQ. For the OCPs, the study was limited to HCB (86% >LOD). p,p’-DDE was present in all samples (100% >LOD) and p,p’-DDT and -DDD in the majority of the samples. The selection of the other contaminants was made in a similar way. Additional criteria were applied for PCDD/Fs, PCBs, PBDEs and PFOS (see Table 4.14). This study was limited to the edible parts (i.e. data on livers was excluded). Data on Western Scheldt fish were not considered because only very little fish from
that region is consumed, whereas contaminant concentrations are high due to local pollution. This would have led to an unrealistically high contribution of fish from this area to the contaminant exposure (i.e. PFOS, PBDEs and HBCD).

Table 4.14 Overview of contaminants included in this study.

<table>
<thead>
<tr>
<th>Contaminant class</th>
<th>Individual compounds included</th>
<th>Rationale for selection and source of data</th>
</tr>
</thead>
<tbody>
<tr>
<td>PCDD/Fs and dl-PCBs</td>
<td>All 17 WHO PCDD/Fs and 12 WHO dl-PCBs</td>
<td>TEF(^1) values and TDI(^2) values available based on these congeners (10). Source: (11,12).</td>
</tr>
<tr>
<td>PCBs</td>
<td>CB 28, 52, 101, 118, 138, 153 and 180 (7 indicator PCBs)</td>
<td>Selection commonly reported. Source: (11,12).</td>
</tr>
<tr>
<td>HCB</td>
<td>N.a.</td>
<td>In 86% of the samples HCB &gt;LOD. Source: (12,13).</td>
</tr>
<tr>
<td>DDTs</td>
<td>Sum of p,p(^\prime)-DDT, p,p(^\prime)-DDD and p,p(^\prime)-DDE</td>
<td>In 100% of the samples the sum-DDTs &gt; LOD. Source: (12,13).</td>
</tr>
<tr>
<td>PBDEs</td>
<td>BDE 28, 47, 49, 99, 100, 153, 154 and 209</td>
<td>Eight congeners selected by EFSA(^3) for monitoring (14). BDE 183 was excluded because nearly all samples were &lt; LOD. On the contrary, BDE 49 was included because in nearly all samples &gt; LOD. Source: (12,15).</td>
</tr>
<tr>
<td>HBCD</td>
<td>α-HBCD</td>
<td>A large proportion of samples (47%) &gt; LOD. β-HBCD and γ-HBCD were excluded because in nearly all samples &lt; LOD. Source: (12,15).</td>
</tr>
<tr>
<td>Perfluorinated compounds</td>
<td>Perfluorooctane sulfonate (PFOS)</td>
<td>Emerging contaminant and recent risk assessment by EFSA (16). A large proportion of samples (39%) &gt; LOD. Other PFCs were &lt; LOD in nearly all samples. Source: (12,17,18).</td>
</tr>
</tbody>
</table>

N.a  Not applicable

\(^{1}\) TEF: 2,3,7,8-Tetrachlorodibenzo-p-dioxin Equivacency Factor
\(^{2}\) TDI: Tolerable daily intake
\(^{3}\) EFSA: European Food Safety Authority

The daily food consumption data was taken from the Dutch National Food Consumption Survey (DNFCS), which specifies the diet of 6,250 individuals (male, female, age 1-97) over two consecutive days (19). The DNFCS was performed in 1998, and does not include recently introduced species like pangasius and tilapia. In order to come to a reasonable inclusion of these species, sales volumes of these fishes were taken from a recent inventory (20). Based on interviews with commercial fisheries representatives, an estimate was made of the volumes of these fish being sold in The Netherlands to consumers in 2006. Taking into account the number of Dutch citizens (assumed 16,000,000), these data can be transferred into daily consumption data (in g/day). For trout, a correction of this consumption was required as the majority of trout is sold as whole fish (degutted), whereas only the fillets are consumed. It was assumed that the fillets represent 30% of the eight of a whole degutted fish, and therefore a 0.3 correction factor was applied. For farmed shrimp, a 0.5 correction factor was applied as shrimp are often sold with non-edible parts (e.g. tails, heads, and legs) still on (20). It is recognised that the DNFCS data and the fish sales data are obtained through different methods and represent fish consumption (DNFCS) or fish sales to consumers.
Contaminants in fish, shellfish and shrimp from The Netherlands

(20). In the latter case, this concerns mostly raw (uncooked) products, and this may differ somewhat from the consumed products. Furthermore, both studies are from different periods (1998 vs. 2006) and changes in consumption patterns may not be represented very well. However, these differences were accepted considering the aim to compare contaminant exposure from newly farmed species (tilapia, pangasius) and wild species (e.g. herring, eel and plaice).

The contaminant data of all species of the same Gadidae family were combined in one dataset. This included cod, haddock, pollock and hake. These have similar feeding habits and their contaminant levels were similar. Also the consumption data on Gadidae were combined. Consumption data on processed products like e.g. fish fingers, 'lekkerbekje' and 'kibbeling' (battered and fried Gadidae) were added to the Gadidae category.

Two categories of shrimp were distinguished, i.e. farmed shrimp (Penaeus monodon, Penaeus vannamei, Litopenaeus vannamei) and wild shrimp (Crangon crangon). For farmed shrimp, consumption data of the 2006 sales inventory was used, whereas for wild shrimp (mainly North Sea), the DNFCS consumption data was used. The DNFCS data of wild shrimp was corrected by a factor 0.2 as farmed shrimp have become much more popular in recent years than wild shrimp. All contaminant concentrations are expressed on a wet weight (ww) basis as the objective of the study is to study the dietary exposure and to enable comparison with e.g. TDI values.

The exposure calculation is based on the daily fish consumption multiplied by the mean lowerbound concentrations (per species) as in

$$ DE = \Sigma (Q_a \cdot C_a + Q_b \cdot C_b + \ldots + Q_z \cdot C_z) $$

in which $DE = \text{Daily Exposure}, Q = \text{Quantity of daily consumption of fish species } a, b \text{ etc. and } C = \text{mean lowerbound contaminant concentration in fish species } a, b \text{ etc.}$. The mean contaminant concentrations are based on lowerbound data, except for total-TEQ values.

**Results and discussion**

The bottom panel of Figure 4.14 shows that herring and salmon are the predominant contributors to the exposure of the lipophilic contaminants like PCDD/Fs, PCBs, DDTs, HCB, PBDEs and HBCD. This is caused by the relative high daily consumption (Figure 4.14, top panel) combined with the relatively high contaminant levels in these species (Figure 4.14, middle panel). Wild eel has high contaminant levels, but due to the low consumption, the contribution to the exposure is minimal. The farmed species tilapia and pangasius had very low exposure levels because of their low contaminant concentrations, being approx. 100-fold lower (mean) than those in e.g. salmon and herring (Figure 4.14, top). Cod (and other Gadidae) show concentrations in between salmon/herring and sole/plaice/pangasius etc.
Figure 4.14 Fish consumption / sales (top panel), the mean contaminant concentrations (middle panel), and mean dietary exposure (third panel). *Data for Total-TEQ is expressed in pg TEQ/g ww (second panel) and pg TEQ / day (bottom panel). The legend in the bottom panel also applies to the middle panel.
The contaminant concentrations were low but the exposure from Gadidae is nevertheless substantial (i.e. for PCBs and PFOS) due to the high Gadidae consumption (both from fish fillet and popular processed fish such as fish fingers, “kibbeling” and “lekkerbekje”). Exposure from wild and farmed shrimp and from mussels is low (except for PFOS in wild shrimp).

PFOS shows a different exposure pattern. PFOS is a surfactant and because it mostly occurs in the ionic state, it has a different accumulation mechanism compared to the neutral lipophilic contaminants (21). PFOS was not found in any of the salmon samples (<0.6 ng/g ww). This is somewhat surprising as salmons are fed with feeds containing fish meal and fish oil. There is no information on PFOS levels in fish meal, nor in fish oil. Herring stands out for the high contribution to the PFOS exposure. Feeding habits may play a role. Herring feeds predominantly on zooplankton and in other studies it was shown that zooplankton contains PFOS in the low ng/g ww range (22). Finally, plaice stands out for the relatively high PFOS exposure. This is caused by the relatively high PFOS concentrations (higher than lipophilic compounds), in combination with the consumption data. Sole showed PFOS concentrations comparable to those in plaice, which is explained by similar feeding habits of these species, but due to a lower consumption, the exposure contribution of sole is low.

The data is presented as lowerbound data (except for PCDD/Fs and dl-PCBs), in order to minimise the role of LODs on the result. Upperbound data did not differ much (data not presented), because only contaminants were selected for which a large proportion of the samples had >LOD concentrations. Means of all contaminant concentrations per species were calculated. For comparison, the median was also calculated, but the results showed no large differences (data not shown).

The total exposure per contaminant (Figure 4.15) shows that the seven indicator PCBs and PFOS represent approx. 70% of the exposure to all contaminants. The exposure of the general Dutch citizens (i.e. exposure averaged over all Dutch citizens, including non-consumers) to the seven indicator-PCBs was 77 ng/day (1.1 ng/kg bw per day). No TDI exists for the indicator PCBs. A re-evaluation of the toxicity of non dioxin-like PCBs (ndl-PCBs) is currently ongoing in the framework of the EU project Athon (23). The ndl-PCB concentrations in all fish samples were (far) below the current Dutch maximum level (ML) (see (11)). A Swedish study showed higher PCB exposures (349 ng/day, sum of 23 PCBs, 79 ng/day for CB 153 only) (1).

The PFOS exposure is 72 ng/day (1.0 ng PFOS/kg bw per day). The EFSA recently published a human risk evaluation of PFOS. They determined a TDI of 150 ng PFOS/kg bw per day (16). In that study, an exposure of 58 ng PFOS/kg bw per day was determined for the Dutch population (from fish). This is considerably higher than found in the present study because EFSA (i) used much higher PFOS concentrations in fish (68 ng/g ww for all fish) and (ii) they used consumer only fish consumption data, meaning a 5-fold overestimation of the fish consumption compared to the general Dutch population. A more
thorough discussion on the causes of these differences can be found in Chapter 4.3. Based on the exposure estimated in the present study, it is assumed that the average consumer will not exceed the PFOS-TDI from fish consumption only. Data on other food items should also be considered, but data is still lacking. Within the framework of the EU Perfood project (2009-2012) data will be produced EU-wide that will enable an exposure assessment including several food items, drinking water, beverages and other exposure pathways.

![Figure 4.15](image)

**Figure 4.15**  
Summarised exposure to different contaminants (ng/day) from all fish, and relative importance of different contaminant groups. Legenda: Σ denotes sum (e.g. Σ7-PCB means the sum of 7 PCB congeners).

The exposure to DDTs in this study is lower than in Sweden (256 ng/day for the sum of four DDTs, 164 ng/day for p,p’-DDE) (1). The dietary exposure to DDT (sum of 6 isomers) of secondary school students in Hong Kong amounted 145-291 ng/kg bw per day and was predominantly caused by seafood consumption (24). This is much higher than found in the Swedish study and in this study.

The exposure to PBDEs is lower than in Sweden (fish only, 23 ng/day for 5 PBDEs, 17 ng/day for BDE 47) (1). The exposure for a 70 kg man from Catalonia, Spain was estimated at 27 ng/day (fish only, sum of six PBDEs) (2). In both studies, fish was the predominant contributor to the exposure.

The exposure to α-HBCD is low (4 ng/day) and far below benchmark doses (1.6-3.0 mg/kg bw per day) obtained from toxicity evaluations (25,26). The mean total-TEQ was 18 pg/day, which corresponds to an exposure of 0.26 pg/kg bw per day. This is in the same order of magnitude as reported by de Mul et al. (4). This contribution is well below the TDI of 2 pg/kg bw per day as set by the Scientific Committee on Food (27). When other foods are included, the exposure approaches the TDI (0.8 pg/kg bw per day) (4). The exposure used in this study is lower than reported by Darnerud et al. (2006) for Sweden (30 pg/day) (1).

The HCB exposure is well below the suggested guidance value of 160 ng/kg bw per day (28) and was 6-fold lower than in Sweden (1).
Conclusions

Herring and salmon are the most important fish species that contribute to the exposure of Dutch consumers when considering all contaminants studied. The exposure resulting from wild and farmed shrimp, sole and trout and the newly farmed species pangasius and tilapia was low. The exposure was dominated by PFOS and the sum of the 7 indicator PCBs (approx. 70% of the total). PFOS showed a deviating exposure pattern mainly because no exposure through salmon was observed, whereas salmon is an important contributor for other contaminants. PFOS exposure through plaice was substantial, whereas plaice was not relevant for the other contaminants. Exposure to HCB and α-HBCD was low due to low contaminant concentrations in all fish species.

The exposure to dioxins and dl-PCBs was the low (18 pg total-TEQ/day, or 0.26 pg total-TEQ/kg bw per day). Nevertheless, the exposure was only 8-fold below the TDI, which is a small margin. Safety margins were higher for the other contaminants.
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