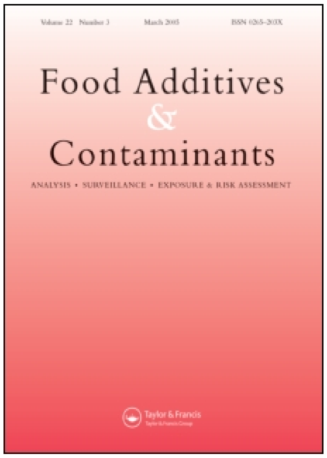

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Probabilistic approach to polychlorinated biphenyl (PCB) exposure through eel consumption in recreational fishermen vs. the general population

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Abstract
Concentrations of the sum of the seven indicator PCBs (Σ_7 iPCBs) measured in non-commercial European eel (*Anguilla anguilla* L.) in Flanders are high: in 80% of all sampled localities, the Belgian PCB standard for fish was exceeded. The objective of this study was to assess the intake of the Σ_7 iPCBs through consumption of eel by recreational fishermen and to compare it to the intake of a background population. The median estimated intake for recreational fishermen varied between 18.4 and 237.6 ng iPCBs kg^{-1} bw day^{-1}, depending on the consumption scenario, while the estimated intake of the background population (consumers only) was 4.3 ng iPCBs kg^{-1} bw day^{-1}. Since the levels of intake via eel for two intake scenarios were, respectively, 50 and 25 times higher than the intake of the background population, the body burden (BB) might be proportionally higher and reach levels of toxicological relevance. The intake of the seven iPCBs via consumption of self-caught eel in Flanders is at a level to cause serious concern. The Flemish catch-and-release obligation for eel, established in 2002, should be maintained and supervised (more) carefully.

Keywords: *Indicator PCB, dietary intake, exposure, recreational fisheries*

Introduction

Polychlorinated biphenyls (PCBs) are found in many chemical mixtures, mainly used in electronic appliances, heat transfer systems and hydraulic fluids, but also in other applications such as paints, coatings and flame retardants. The use of PCBs was restricted in the 1970s; however, most PCB congeners are very lipophilic and persistent and tend to accumulate in the environment and the human food chain. Mixtures of PCBs are generally assessed on the basis of a chemical analysis of the (sum of the) so-called indicator PCBs (Σ_7 iPCBs, i.e. congeners 28, 52, 101, 118, 138, 153, 180). None of these PCB congeners exhibits dioxin-like activity, except for PCB 118, which has a toxic equivalence factor (TEF) value of 0.00003 (van den Berg et al. 2006). They are known to bioaccumulate in the human diet and are assumed to be representative for all PCBs, as they are the predominant congeners in biotic and abiotic matrices (Bakker et al. 2003). The sum of six indicator PCBs (congeners 28, 52, 101, 138, 153 and 180) represents about 50% of the total non-dioxin like PCBs in food (EFSA 2005).

European eel (*Anguilla anguilla* L.) is known to bioaccumulate lipophilic contaminants, such as PCBs and organochlorine pesticides, through carnivorous feeding behaviour. Moreover, eel is a so-called benthic fish, living near or in the contaminated sediment. Consequently, eel is expected to undergo significant exposure to contaminants and is, therefore, commonly used as an

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environmental bio-indicator for a variety of contaminants (Wiesmuller and Schlatterer 1999; Versonnen et al. 2004). Human dietary exposure to iPCBs might be exacerbated by the consumption of highly-contaminated fishes, at least for a sub-population of eel consumers (Harrad and Smith 1999).

Since 1994, the Flemish eel pollutant-monitoring network has monitored about 300 different sites in Flanders (the northern part of Belgium, a region of 13 500 km^2) by measuring contaminants in European eel. The monitoring sites are situated in rivers, canals, polder waters and closed water bodies. The monitoring program includes PCBs, organochlorine pesticides (e.g. hexachlorobenzene, lindane, dieldrin, etc.), polybrominated flame retardants (polybrominated diphenyl ethers, etc.) and heavy metals (such as mercury, cadmium, lead, arsenic, etc.) (Goemans et al. 2003; Goemans and Belpaire 2004).

Concentrations of the Σ_7 iPCBs measured by this monitoring network are very high: in 80% of all sampling sites, the mean concentration in eel exceeds the Belgian PCB standard for fish (75 ng g^{-1} fresh weight) (Goemans and Belpaire 2004). For this reason, in 2002, the Flemish authorities have issued a catch-and-release obligation for all fish in the five most polluted waters in Flanders and an overall catch-and-release obligation for eel in the whole region. It has been demonstrated that, in spite of this restriction, some recreational fishermen still take their eel home, most likely for consumption (Vandecruys 2004).

The objective of this study was to assess the intake of Σ_7 iPCBs via eel consumption in this sub-group of recreational fishermen and to compare it to the intake of a Flemish background population.

Materials and methods

To estimate the exposure to Σ_7 iPCBs through eel consumption, two approaches were used. For the sub-population of fishermen (and their families), a simple distribution approach was used in which a point-estimate for eel consumption was combined with a contaminant distribution, based on the available data for iPCB contamination of eel (Lambe 2002). On the other hand, for the background population (eel consumers only), two distributions were combined in a full probabilistic model (Cullen and Frey 1999): a distribution for eel consumption and a distribution for PCB contamination (using @Risk® 4.5 for Excel®, Palisade Corporation, Ivybridge, Devon, UK).

Recreational fishermen

In 2003, 61 245 individuals in Flanders had a fishing license for public waters. A survey on specific aspects of recreational fisheries, including the issue of taking home a catch, was carried out (Vandecruys 2004). The survey included questions on the fish species caught and taken home, as well as the number and weight of fish caught and taken home. A systematic random sampling of the dataset of anglers on public waters was carried out and 10 000 entries were selected. After omitting foreign anglers and undelivered mail, the real sample size was 9492. A total of 3001 licensed anglers completed this questionnaire about recreational fishing. Respectively, 1.9 and 5.3% of these anglers indicated that they "always" (group A) or "sometimes" (group B) take home eels caught. No information was obtained about what the fish were used for. Therefore, some assumptions had to be made concerning the consumption of these fishes. However, personal or familial consumption can be expected, based on the small number of eels caught per fishing trip. Extrapolating to all licensed fishermen, the number of people taking home eels, caught in Flemish public waters, is estimated to be more than 4000.

For group A (the group of fishermen always taking home eels caught), it is calculated that an average of 25.88 kg year^{-1} of edible eel (or a mean of 498 g week^{-1}) is taken home, based on the number of fishing occasions (average of 41.67 trips year^{-1}), the number of eels caught per occasion (average 4.14) and a mean weight of edible portion per eel (150 g). For group B, the fishermen stating that they only "sometimes" take home their catch, it was assumed that, on average, one eel of five caught is taken home. The same calculation has been done (average number of fishing occasions = 42.03 per year, the number of eels caught per occasion and taken home = 3.12/5, the mean weight of edible portion per eel = 150 g), resulting in 3.93 kg edible eel per year (76 g week^{-1}).

We further considered two different consumption scenarios for both groups:

- In scenario A1, the fisherman takes home 498 g week^{-1} (see above) or 71.14 g day^{-1}. In this worst-case scenario, it was assumed that it was consumed by the angler himself.
- In scenario A2, the fisherman takes home the same amount of eel (498 g week^{-1}). Here, it was assumed that he eats only half of this amount (i.e. 35.57 g day^{-1}). The other half could be consumed by friends and/or family.
- In scenario B1, the fisherman takes home 76 g week^{-1} (see above) or 10.86 g day^{-1}. This is consumed by the fisherman himself.

– In scenario B2, the fisherman takes home the same amount ($76\,g\,week^{-1}$) and eats half of it (i.e. $5.43\,g\,day^{-1}$).

Fishermen were assumed to have a mean body weight (bw) of 70 kg.

Data on the iPCB contamination of eel in the Flemish water-bodies were based on the eel pollutant-monitoring network in Flanders 1994–2001 (Goemans et al. 2003; Goemans and Belpaire 2004). The concentration of iPCB was analysed in 261 samples. Length of sampled eels varied between 30 and 50 cm. The sampling sites are spread over Flanders.

A distribution of iPCB concentrations in eel was fit, using BestFit® software (BestFit Probability Distribution Fitting for Windows; Palisade Corporation, Ivybridge, Devon, UK). BestFit® determines the optimal distribution and the optimal parameters for each data set, performing three standard tests to determine the goodness of fit: Chi-squared, Anderson–Darling and Kolmogorov–Smirnov. The probability distributions evaluated by BestFit include 28 possible distributions (e.g. binomial, exponential, gamma, logistic, log–logistic, lognormal, normal distribution, etc.). All these distributions were tested. In this study, the Anderson–Darling test was used to determine the optimal distribution: this test focuses on the differences between the tails of the fitted distribution and input data, rather than on the center of the distribution. To preclude excessively high contamination data, the distribution was truncated at the upper level, at twice the maximum value measured during monitoring ($13\,466\,ng\,g^{-1}$). The distribution was also truncated at the lower end (half of the minimum value: $5.5\,ng\,g^{-1}$).

Background population

For the background population, the most recent data on eel consumption available in Belgium were used. Within the context of a large Flemish biomonitoring study, in the field of environmental health, a food frequency questionnaire (FFQ) was used to estimate the daily consumption of fat-containing food items. This FFQ contained a question on the frequency ("how often do you consume eel?" with seven response categories, ranging from "never or less than 1 day a month" to "6 to 7 days a week") and the portion ("how much do you consume on that day?") of eel consumption. This FFQ was completed by 1179 women of childbearing age (18–44 years). The data were collected between September 2002 and December 2003.

In this study population, a total of 132 women (11.2%) consumed eel at least once during the last year. The mean intake among consumers was 2.87 ($\pm$1.28) $g\,day^{-1}$.

Again, BestFit® software was used to determine a distribution describing these consumption data. To preclude unrealistic consumption data, the distribution was truncated at $0.16\,g\,day^{-1}$ (half of the minimal estimated consumption) and at $15\,g\,day^{-1}$ (double of the maximal estimated consumption).

For this population, contamination data on the Σ_7 iPCBs, measured in commercially available eel in Flanders, were used (Belpaire et al. 2000). A total of 81 samples of commercially available eel was analysed for iPCBs. Again, a distribution was fit to these data using BestFit® software. To preclude unrealistic contamination values, the distribution was truncated at both ends: $0.7\,ng\,g^{-1}$ (half of the minimal contaminant concentration) and $11\,472\,ng\,g^{-1}$ (double of the maximum contaminant concentration).

The consumption and the contamination distributions were combined using a probabilistic approach (@Risk®, Risk Analysis Add-in for Microsoft Excel; Palisade Corporation). The mean body weight (self-reported) of the women was 64.6 ($\pm$11.4) kg.

Results

Distributions

For the contamination data of eel (commercially available eel and eel caught by Flemish recreational fishermen), two lognormal distributions were chosen. In Figure 1, the original contamination data are compared via a Box and Whisker plot. In Figure 2, the fitted distributions, based on these contamination data, are shown.

Also, for the consumption of the background population, a lognormal distribution was used.

Based on the distribution of the data (Figure 1), the truncation of the distribution at double of the maximum seems reasonable, since the probability of measuring concentrations higher than twice the maximum is very low.

iPCB exposure

The median intake for recreational fishermen varies between 18.4 ng iPCBs $kg^{-1}\,bw\,day^{-1}$ (scenario B2: consumption of $10.9\,g\,eel\,day^{-1}$) and 237.6 ng iPCBs $kg^{-1}\,bw\,day^{-1}$ (worst case scenario A1: consumption of $71.1\,g\,eel\,day^{-1}$). At median level, the estimated intake of the background population (consumers only) is 4.3 ng iPCBs $kg^{-1}\,bw\,day^{-1}$. At the 90th percentile, the estimated intake for the fishermen varies between 86 (consumption scenario B2) and 1118 ng iPCBs $kg^{-1}\,bw\,day^{-1}$

(scenario A1), while the intake for the background population (consumers only) is 42.9 ng iPCBs kg^{-1} bw day^{-1}. The estimated intakes for the Σ_7 iPCBs are presented in Table I for both the background population and the fishermen.

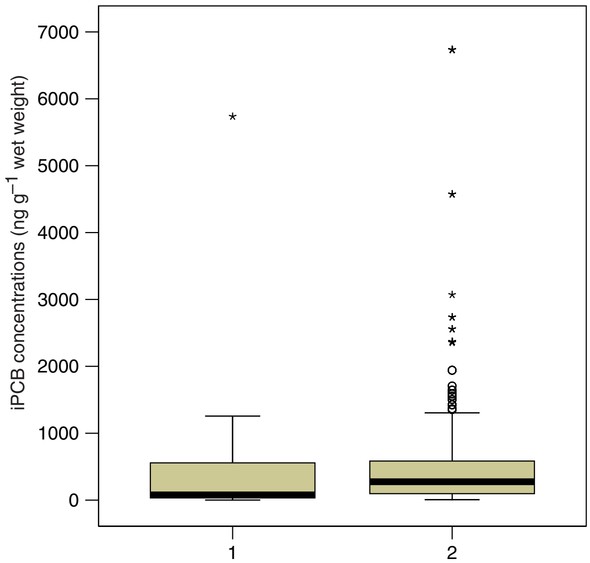

Figure 1. Box and Whisker plots for concentrations of the Σ_7 iPCBs (ng g^{-1} wet weight), analysed in (1) commercially available eel ($n = 80$) and (2) eel in Flemish waterbodies ($n = 261$). Each box represents the interquartile range (P25–P75). The bold line expresses the median value. The whiskers extend from the boxes and indicate the upper and lower values not classified as statistical outliers or extremes. Stars are statistical outliers (i.e. cases with values between 1.5 and 3 times the interquartile range). Open circles are statistical extreme values (i.e. cases with values more than 3 times the interquartile range).

Cumulative distribution functions for the estimated intake of Σ_7 iPCBs are shown for the background population and for the different consumption scenarios of the fishermen in Figure 3.

It should be noted that the results presented in this study (Table I and Figure 1) are based on eel consumers only: 7.2% of the recreational fishermen consume self-caught eel, while 11.2% of the background population are eel consumers. When extrapolating these results to an intake assessment for the population at large (consumers and non-consumers together), the assessed intakes of this study would be situated at the higher end of the overall distribution.

On the other hand, only the intake via eel is taken into account. Also other food items, such as other fish and food items containing animal fat, will contribute to the overall PCB intake. In a previous dietary intake assessment of polychlorinated dibenzodioxins/furans (PCDD/Fs) and dioxin-like PCBs in Belgium, Vrijens and co-workers (2002) reported that fish remains an important source of dioxin-like contaminants for the higher percentiles of the population. At the 90th percentile, fish becomes the greatest contributor to dietary PCB exposure (Vrijens et al. 2002).

Discussion

The intake of iPCBs via eel consumption was estimated using a probabilistic model, based on Monte Carlo techniques, for a population that could be at risk, i.e. eel fishermen, and compared to a

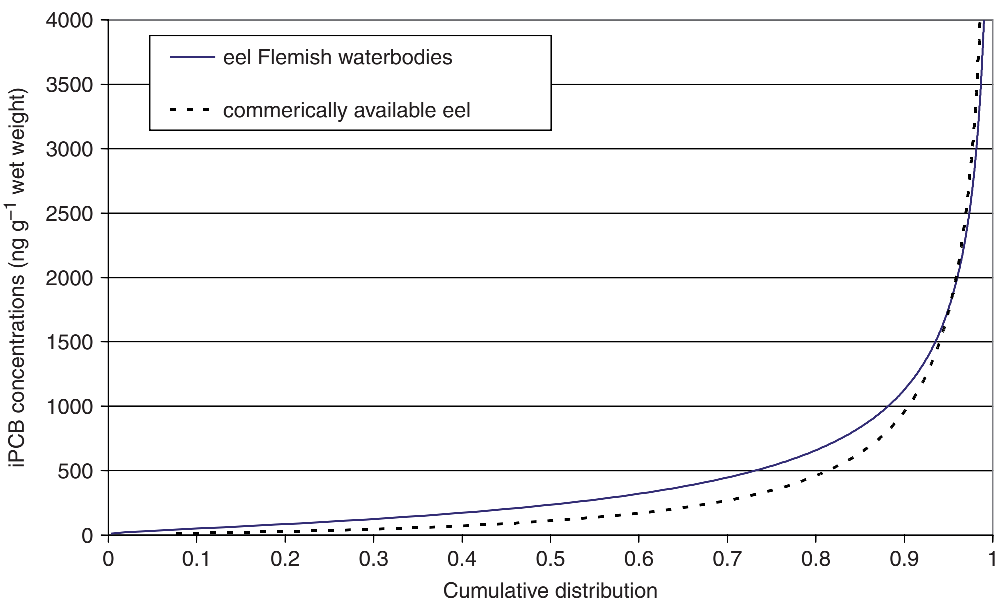

Figure 2. Fitted cumulative distribution functions for the concentration of Σ_7 iPCBs (ng g^{-1} wet weight) for eel in Flemish waters and commercially available eel.

background population. Large differences in estimated intake have been found between the various scenarios.

Methodological considerations

Probabilistic techniques, such as Monte Carlo analysis, have been used since the 1990s to characterize the health risk of populations exposed to various chemicals (McKone 1994; Carrington et al. 1996). Many papers have been published showing that probabilistic methods represent a significant improvement over deterministic approaches (Finley et al. 1993; Finley and Paustenbach 1994; Thompson 2002). As in deterministic techniques, however, the quality of the output depends largely on the quality of the input data.

The available information on consumption for the population of recreational fishermen is rather tenuous and several assumptions had to be made: fishermen stated that they take home the fish they have caught, yet it is not known who is consuming this eel. We have chosen to consider four different scenarios, as a reflection of a range of true variation. In the worst case scenario, the mean intake is 498 g eel week^{-1}. Other available consumption data from Flanders (a seven-day food record, 341 adolescents, 12–18 years old, 1997) (Matthys et al. 2003), showed that a consumption of 500 g fish week^{-1} corresponds to the 97th percentile of the distribution for total fish consumption for adolescents. Our worst case scenario, therefore, seems not to be exceptional, compared to the general population. It is perhaps not unrealistic to assume that at least some anglers are among the highest consumers of fish in the population.

Considering the background population, it could be stated that women of childbearing age (18–44 years) are not a representative group for the general population in assessing the consumption

Table I. Estimated intake of the Σ7 iPCBs (ng kg^{-1} bw day^{-1}) for the background population and recreational fishermen. Estimates for the fishermen are presented for various consumption scenarios (A1, A2, B1, B2).

		Recreational fishermen			
Percentile	Background	A1	A2	B1	B2
5	0.2	31.9	16.8	5.2	2.5
25	1.3	105.2	52.8	16.1	8.1
50	4.3	237.6	118.6	36.7	18.4
95	80.4	1727.8	861.6	285.2	140.3
97.5	135.2	2513.1	1282.7	425.0	203.9
99	238.2	4032.2	1946.0	647.9	296.7
99.9	707.9	8582.8	4181.3	1362.3	656.0

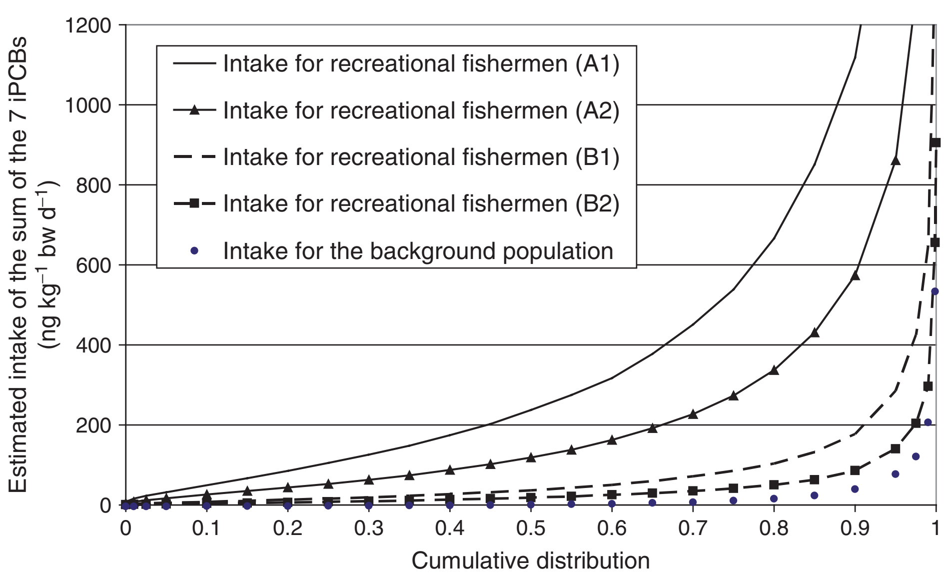

Figure 3. Cumulative distribution functions of the estimated intake of Σ_7 iPCBs (ng kg^{-1} bw day^{-1}) for the background population and recreational fishermen. The results for the fishermen are presented for various consumption scenarios (A1, A2, B1, B2).

of eel. It is clear that there are differences in consumption between men and women and between different age groups. Nevertheless, these data were used because no other recent consumption data on eel were available for Belgium or Flanders. The FFQ used focussed on consumption during the last year.

Concerning contamination data, two different data sets were used since the contamination of eel, commercially available on the Belgian market (exposure for the background population), is known to differ from the contamination of eel caught in public waters in Flanders (exposure for the recreational fisherman). Contamination levels can be influenced by several factors. It is possible and even probable that some individuals of the background population, consuming eel in a restaurant, are served eel from an unofficial source. This eel might be caught in private waters. PCB levels of those eels are unknown but suspected of being in the range of the eels living in public waters in Flanders. This could be a possible reason for an underestimation of exposure in the background population. Secondly, it is known that consumers can reduce the contaminant level by removing the skin and fat from fish before cooking (Sidhu 2003). Other processing or cooking procedures will also influence the contaminant level. Furthermore, the dataset of contaminants in feral eel from Goemans et al. (2003) originated from eels of a specific length class (30–50 cm). Many eels caught and consumed by fishermen are larger and, therefore, contain higher contaminant levels. Thus, our calculation of PCB exposure may be biased and data presented here may be an underestimation. From the dataset, it is obvious that regional variations in PCB contamination throughout Flanders are important (Goemans et al. 2003). Refined analysis of intake levels from heavily contaminated eels in specific areas could reveal more severe risks.

Available data on intake of iPCBs in other countries

Comparable literature data are scarce for several reasons (Wilhelm et al. 2002; Bakker et al. 2003; Baars et al. 2004; Fattore et al. 2005). The most important reason is the use of different methodologies, such as (1) a different number of congeners (e.g. Σ_3 PCBs, Σ_6 PCBs, Σ_7 PCBs, Σ_{10} PCBs) taken in account, (2) intake via total diet vs. via specific food groups or food items, (3) total population vs. consumers only, (4) different age groups, etc. Nevertheless, a limited number of intake estimates from other countries are presented here.

In Italy, the intake of Σ_6 iPCBs (PCB 28, 52, 101, 138, 153 and 180) was estimated based on a food diary of 3–7 consecutive days, completed by 1940 subjects (age 0–94 years) (Fattore et al. 2005). The estimated intake for adolescents and adults (13–94 years) was 5.9, 10.9 and 23.8 ng kg^{-1} bw day^{-1} for the 5th percentile, mean and 95th percentile, respectively. On average, 42% could be attributed to fish and fish products. This means that, on average, 4.6 ng kg^{-1} bw day^{-1} (Σ_6 iPCBs) is due to the consumption of fish and fish products.

A Dutch intake assessment of Σ_7 iPCBs via the whole diet resulted in following estimated median intake: 4.8 ng iPCBs kg^{-1} bw day^{-1} (Bakker et al. 2003; Baars et al. 2004). At the 90th percentile, an intake of 8.6 ng iPCBs kg^{-1} bw day^{-1} was estimated. In France, the average intake of Σ_7 iPCBs among French high seafood consumers (Calipso Study) was estimated to be 57 ng kg^{-1} bw day^{-1} through seafood consumption only (Sirot et al. 2006). Recent European studies estimated the average daily intake of total non dioxin-like PCBs for adults to be in the range 10–45 ng kg^{-1} bw day^{-1} (EFSA 2005).

Risk evaluation

Non dioxin-like PCBs are less toxic than PCDD/Fs and dioxin-like PCBs. Nevertheless, it is recommended that intake be as low as possible. Unlike dioxin-like substances (Tolerated Daily Intake (TDI) = 1–4 pg TEQ kg^{-1} bw day^{-1}) (Scientific Committee on Food 2001) or total PCBs (TDI = 20 ng kg^{-1} bw day^{-1}, in Aroclor equivalent) (WHO 2003), no specific health-based guidance value (e.g. a tolerated daily or weekly intake, TDI or TWI), has been proposed for non-dioxin-like PCBs only (EFSA 2005). The major problem is the difficulty in distinguishing between effects of non dioxin-like PCBs and effects of dioxin-like PCBs and PCDD/Fs, which may be part of PCB mixtures. No definite relationship, however, has been found between levels of non-dioxin-like PCBs and levels of dioxin-like PCBs and PCDD/Fs in these mixtures. Occasionally, a specific relationship could be found, e.g. in the PCB animal-feed contamination case in Belgium in 1999 or in geographically defined sampling areas (Vrijens et al. 2002; EFSA 2005).

WHO (2003) proposed a TDI for total PCBs, expressed in Aroclor equivalent, of 20 ng kg^{-1} bw day^{-1}, while Sirot et al. (2006) stated that the concentration of Σ_7 iPCBs must be multiplied by 2 to be expressed in Aroclor equivalents. If our calculated exposure (the exposure of Σ_7 iPCBs multiplied by 2) is compared with the TDI, it can be seen that more than 30% of the eel consumers in the background population exceeds this TDI, without taking into account other PCB sources. In comparison, between 70 and 99% of recreational fishermen exceed this TDI, depending on the consumption scenario used.

In a recent publication, a statistically significant relationship was observed between individual dioxin-like PCBs and total PCBs, measured in a number of fishes caught mainly in Canada and Northern America (Bhavsar et al. 2007). This correlation could be an interesting application for risk assessment estimations in that region. However, it has not been demonstrated that this relationship is applicable to other geographical regions. In contrast, clear spatial and temporal variations have been observed in the ratio of PCB118 to the sum of the remaining six iPCBs in eel in Flemish waters (Goemans and Belpaire 2005). Therefore, this extrapolation has not been used in the current estimation as this paper only handles the intake of eel, locally caught in Belgium.

EFSA (2005) concluded that the margin of body burden (MoBB), calculated by comparing the body burden (BB) in the rat at the no-observed adverse effect level (NOAEL) of 500 $\mu g\,kg^{-1}$ bw (liver and thyroid toxicity) with the estimated median human BB for total non-dioxin-like PCBs (48 $\mu g\,kg^{-1}$ bw) in the general population, was about 10. We do not know the PCBs concentration ingested by fishermen via the total diet but, since the levels of intake via eel in scenario A1 and A2 are, respectively, 50 and 25 times higher than the intake of the background population, BB may be significantly higher, reaching levels that become toxicologically relevant.

Since other animal-based food items are likely to contain some level of iPCBs, it is advisable, therefore, to maintain the catch-and-release obligations and to educate recreational fishermen about the health risk of eel in the Flemish waters.

The background population has to be considered as high eel consumers may also be at risk. In other countries, e.g. USA, advisories on fish consumption have been formulated, especially focussing on pregnant women, young children (under 15) and women of childbearing age (MDCH Environmental and occupational epidemiology division 2004; Scientific Advisory Committee on Nutrition and Food Standard Agency 2004; US EPA/US FDA 2004; US EPA 2005). In addition, the Swedish National Food Administration has recommended that pregnant and lactating women refrain from eating some predatory fish species, including eel (Bjornberg et al. 2005).

Conclusion

In conclusion, the intake of the Σ_7 iPCBs via the consumption of self-caught eel is a cause of serious concern. Further monitoring seems appropriate. Although risk assessment would be easier if, in analogy with PCDD/Fs and dioxin-like PCBs, a reference TDI or TWI could be established for the Σ_7 iPCBs only, the possibility is unlikely in the near future (EFSA 2005). In the meantime, it is advisable to maintain the public health measure of preventing fishermen from consuming their self-caught fish. The catch-and-release obligation should be maintained and supervised more vigilantly.

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