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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 with the intake of a background population. The median estimated intake for recreational fishermen varies between 18.4 and 237.6 ng iPCBs/kg bw/day, depending on the consumption scenario, while the estimated intake of the background population (consumers only) is 4.3 ng iPCBs/kg bw/day. 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 proportionately higher and reach levels of toxicological relevance. The intake of the seven iPCBs via consumption of self-caught eel in Flanders seems to be at a level of high 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) exist in many different technical mixtures and were 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 considerably restricted in the 1970's. 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, that 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 6 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 and in the contaminated sediment. Consequently, eel is expected to have a large exposure to contaminants and is therefore commonly used as an environmental bio-indicator for a variety of contaminants (Versonnen et al. 2004; Wiesmuller and Schlatterer 1999). Human dietary exposure to

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4 iPCBs might be driven by the consumption of highly-contaminated fishes, at least for a
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6
7 subpopulation of eel consumers (Harrad and Smith 1999).
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11 Since 1994, the Flemish eel pollutant monitoring network monitors about 300 different
12 sites in Flanders (the northern part of Belgium, a region of 13,500 km²) by measuring
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14 contaminants in European eel. The monitoring sites are situated in rivers, canals, polder
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16 waters and closed water bodies. The monitoring program includes PCBs,
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18 organochlorine pesticides (e.g. hexachlorobenzene, lindane, dieldrin, ...),
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20 polybrominated flame retardants (polybrominated diphenyl ethers, ...) and heavy metals
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22 (such as mercury, cadmium, lead, arsenicum, ...) (Goemans et al. 2003; Goemans and
23
24 Belpaire 2004).
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33 The concentrations of the Σ_7 iPCBs measured by this monitoring network are very high:
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35 in 80 % of all sampling sites, the mean concentration in eel exceeds the Belgian PCB
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37 standard for fish (75 ng/g fresh weight) (Goemans and Belpaire, 2004). For this reason,
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39 in 2002, the Flemish authorities have issued a catch-and-release obligation for all fish in
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41 the 5 most polluted waters in Flanders and an overall catch-and-release obligation for
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43 eel in the whole region of Flanders. It has been demonstrated that, in spite of this
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45 restriction, some recreational fishermen still take their eel home, most likely for
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47 consumption (Vandecruys 2004).
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54 The objective of this study was to assess the intake of Σ_7 iPCBs via eel consumption in
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56 this sub-group of recreational fishermen and to compare it to the intake of a Flemish
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58 background population.
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Materials and Methods

In order to estimate the exposure to Σ_7 iPCBs through eel consumption, two approaches were used. For the sub-population of fishermen (and their family), 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).

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 the weight of the 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 9,492. A total number of 3,001 of the 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 the eel they have caught. No information was obtained about what these fishes were used for. Therefore, some assumptions had to be made concerning the consumption of these fishes. However, personal or familial consumption can be

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4 expected based on the small number of eels caught per fishing trip. Based on
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6 extrapolation to all licensed fishermen, the number of people taking home the eel,
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8 caught in Flemish public waters, is estimated to be more than 4,000.
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13 For group A (the group of fishermen always taking home the eel caught), it is calculated
14
15 that an average of 25.88 kg/year of edible eel (or a mean of 498 g/week) is taken home,
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17 based on the number of fishing occasions (average of 41.67 trips/year), the number of
18
19 eels caught per occasion (average of 4.14) and a mean weight of edible portion per eel
20
21 (150 g). For group B, the fishermen stating that they only “sometimes” take home their
22
23 catch, it was assumed that on average one eel out of five caught, is taken home. The
24
25 same calculation has been done (average number of fishing occasions = 42.03/year, the
26
27 number of eels caught per occasion and taken home = 3.12/5, the mean weight of edible
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29 portion per eel = 150 g), resulting in 3.93 kg edible eel per year (76 g/week).
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35 We further considered two different consumption scenarios for both groups:

- 36 - In scenario A1, the fisherman takes home 498 g/week (cf. supra) or 71.14 g/day.
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38 In this worst case scenario, it was assumed that this was consumed by the angler
39
40 himself;
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42
- 43 - In scenario A2, the fisherman takes home the same amount of eel (498 g/week).
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45 Here it was assumed that he eats only half of this amount (i.e. 35.57 g/day). The
46
47 other half could be consumed by friends and/or family;
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49
- 50 - In scenario B1, the fisherman takes home 76 g/week (cf. supra) or 10.86 g/day.
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52 This is consumed by the fisherman himself;
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- 55 - In scenario B2, the fisherman takes home the same amount (76 g/week) and eats
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57 half of it (i.e. 5.43 g/day).
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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). 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, the normal distribution, ...). All these distributions were tested. In this study, the Anderson-Darling test was used in order 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. In order to preclude too high contamination data, the distribution was truncated at the upper level, at twice the maximum value measured during monitoring (13,466 ng/g). Also at the lower end the distribution was truncated (half of the minimum value: 5.5 ng/g).

The background population

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4 For the background population, the most recent data on eel consumption available in
5
6 Belgium were used. Within the context of a large Flemish biomonitoring study, in the
7
8 field of environmental health, a food frequency questionnaire (FFQ) was used to
9
10 estimate the daily consumption of fat-containing food items. This FFQ contained a
11
12 question on the frequency (“how often do you consume eel?” with 7 response
13
14 categories, ranging from “never or less than 1 day a month” to “6 to 7 days a week”)
15
16 and the portion (“how much do you consume on that day?”) of eel consumption. This
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18 FFQ was completed by 1,179 women of childbearing age (18-44 years). The data were
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20 collected between September 2002 and December 2003.
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28 In this study population, a total of 132 women (11.2 %) consumed eel at least once
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30 during the last year. The mean intake among consumers was 2.87 (\pm 1.28) g/day.
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32 Again, BestFit[®]-software was used to determine a distribution describing these
33
34 consumption data. In order to preclude unrealistic consumption data, the distribution
35
36 was truncated at 0.16 g/day (half of the minimal estimated consumption) and at 15
37
38 g/day (double of the maximal estimated consumption).
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45 For this population, contamination data on the Σ_7 iPCBs measured in commercially
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47 available eel in Flanders were used (Belpaire et al. 2000). A total of 81 samples of
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49 commercially available eel was analysed for iPCBs. Again, a distribution was fit on
50
51 these data using BestFit[®]-software. In order to preclude unrealistic contamination
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53 values, the distribution was truncated at both ends: 0.7 ng/g (half of the minimal
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55 contaminant concentration) and 11,472 ng/g (double of the maximum contaminant
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57 concentration).
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7 The consumption and the contamination distributions were combined using a
8
9 probabilistic approach (@Risk[®], Risk Analysis Add-in for Microsoft Excel; Palisade
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11 Corporation, Ivybridge, Devon). The mean body weight (self-reported) of the women
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13 was 64.6 (\pm 11.4) kg.
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15 16 17 18 **Results**

19 20 *Distributions*

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22 For the contamination data of eel (commercially available eel and eel caught by Flemish
23
24 recreational fishermen), two lognormal distributions were chosen. In Figure 1, the
25
26 original contamination data are compared via a Box and Whisker plot. In Figure 2, the
27
28 fitted distributions, based on these contamination data, are shown.
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34 Also for the consumption of the background population, a lognormal distribution was
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36 used.
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42 < insert Figure 1 about here >

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45 < insert Figure 2 about here >
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49 Based on the distribution of the data (see Figure 1), the truncation of the distribution at
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51 the double of the maximum seems reasonable, since the probability of measuring
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53 concentrations higher than twice the maximum is very low.
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58 *iPCB-exposure*
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5 The median intake for recreational fishermen varies between 18.4 ng iPCBs/kg bw/day
6 (scenario B2: consumption of 10.9 g eel/day) and 237.6 ng iPCBs/kg bw/day (worst
7 case scenario A1: consumption of 71.1 g eel/day). At median level, the estimated intake
8 of the background population (consumers only) is 4.3 ng iPCBs/kg bw/day. At the 90th
9 percentile, the estimated intake for the fishermen varies between 86 (consumption
10 scenario B2) and 1118 ng iPCBs/kg bw/day (scenario A1), while the intake for the
11 background population (consumers only) is 42.9 ng iPCBs/kg bw/day. The estimated
12 intakes for the Σ_7 iPCBs are presented in Table I for both the background population
13 and the fishermen. Cumulative distribution functions for the estimated intake of Σ_7
14 iPCBs are shown for the background population and for the different consumption
15 scenarios of the fishermen in Figure 3.
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33 < insert Table I about here >

34 < insert Figure 3 about here >

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39 It should be noted that the results, presented in this study (Table I and Figure 1), are
40 based on eel consumers only: 7.2 % of the recreational fishermen consume their self
41 caught eel, while 11.2 % of the background population are eel consumers. When
42 extrapolating these results to an intake assessment for the population at large
43 (consumers and non-consumers together), the assessed intakes of this study would be
44 situated at the higher end of the overall distribution.
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56 On the other hand, only the intake via eel is taken into account. Also other food items,
57 such as other fish and food items containing animal fat, will contribute to the overall
58 PCB intake. In a previous dietary intake assessment of polychlorinated
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4 dibenzodioxins/furans (PCDD/Fs) and dioxin-like PCBs in Belgium, Vrijens and co-
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6 authors reported that fish remains an important source of dioxin-like contaminants for
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8 the higher percentiles of the population. At the 90th percentile, fish becomes the greatest
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10 contributor to dietary PCB exposure (Vrijens et al. 2002).
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13 14 15 16 **Discussion**

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18 The intake of iPCBs via eel consumption was estimated using a probabilistic model,
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20 based on Monte Carlo techniques, for a population that could be at risk, i.e. eel
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22 fishermen, and compared with a background population. Large differences of estimated
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24 intake have been found between the different scenarios.
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30 31 *Methodological considerations*

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33 Probabilistic techniques such as Monte Carlo analysis have been used since about 1990
34
35 to characterize the health risks of populations exposed to various chemicals (Carrington
36
37 et al. 1996; McKone 1994). Many papers have been published showing that
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39 probabilistic methods represent a significant improvement over deterministic
40
41 approaches (Finley and Paustenbach 1994; Finley et al. 1993; Thompson 2002). As in
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43 deterministic techniques, however, the quality of the output depends largely on the
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45 quality of the input data.
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52 The available information on consumption for the population of recreational fishermen
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54 is rather elusive and several assumptions had to be made: fishermen stated that they take
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56 home the fish they have caught, still it is not known who is consuming this eel. We have
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58 chosen to consider four different scenarios, as a reflection of a range of true variation. In
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4 the worst case scenario the mean intake is 498 g eel/week. Other available consumption
5 data from Flanders (a seven day food record, 341 adolescents, 12-18 years old, 1997)
6
7 (Matthys et al. 2003), showed that a consumption of 500 g fish/week corresponds to the
8
9 97th percentile of the distribution for total fish consumption for adolescents. Our worst
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11 case scenario, therefore, seems not to be exceptional, as compared to the general
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13 population. It is perhaps not unrealistic to assume that at least some anglers are among
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15 the highest consumers of fish in the population.
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23 Considering the background population, it could be stated that women of childbearing
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25 age (18 – 44 years) are not a representative group for the general population in order to
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27 assess the consumption of eel. It is clear that there are differences in consumption
28
29 between men and women and between different age groups. Nevertheless, these data
30
31 were used because no other, recent consumption data on eel were available for Belgium
32
33 or Flanders. The FFQ used, focussed on consumption during the last year.
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40 Concerning the contamination data, two different data sets were used since the
41
42 contamination of eel commercially available on the Belgian market (exposure for the
43
44 background population) is known to be different from the contamination of eel caught
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46 in public waters in Flanders (exposure for the recreational fisherman). Contamination
47
48 levels can be influenced by several factors. It is possible and even probable that some
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50 individuals of the background population, consuming eel in a restaurant, are served eel
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52 from an unofficial circuit. This eel might be caught in private waters. PCB levels of
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54 those eels are unknown, but suspected to be in the range of the eels living in public
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56 waters in Flanders. This can be a reason for an underestimation of exposure of the
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4 background population. Secondly, it is known that consumers can reduce the
5
6 contaminant level by removing the skin and fat from fish before cooking them (Sidhu
7
8 2003). Also, other processing or cooking procedures will influence the contaminant
9
10 level. Furthermore, the dataset of contaminants in feral eel from Goemans et al. (2003)
11
12 are originating from eels of a specific length class (30-50 cm). Many eels caught and
13
14 consumed by fishermen are larger, and therefore containing higher contaminant levels.
15
16 In this way, our calculation of PCB exposure might be biased and data presented here
17
18 might be an underestimation. From the dataset it is obvious that regional variations in
19
20 PCB contamination throughout Flanders are important (Goemans et al., 2003). Refined
21
22 analysis of intake levels from heavily contaminated eels in specific areas might point
23
24 towards more severe risks.
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33 *Available data on intake of iPCBs in other countries*

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35 Comparable data in literature are scarce, due to several reasons (Baars et al. 2004;
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37 Bakker et al., 2003; Fattore et al. 2005; Wilhelm et al. 2002). The most important
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39 reason is the use of different methodologies, such as (1) a different number of congeners
40
41 (e.g. Σ_3 PCBs, Σ_6 PCBs, Σ_7 PCBs, Σ_{10} PCBs) that are taken in account, (2) intake via
42
43 total diet versus via specific food groups or food items, (3) total population versus
44
45 consumers only, (4) different age groups, etc. In spite of this, a limited number of
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47 intake estimates from other countries are presented here.
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54 In Italy, the intake of Σ_6 iPCBs (PCB 28, 52, 101, 138, 153 and 180) was estimated
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56 based on a food diary of 3 to 7 consecutive days, completed by 1940 subjects (age 0-94
57
58 years) (Fattore et al., 2005). The estimated intake for adolescents and adults (13-94
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4 years) varied from 5.9 over 10.9 to 23.8 ng/kg bw/day for the 5th percentile, mean and
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6
7 95th percentile respectively. On average, 42 % could be attributed to fish and fish
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9 products. This means that on average 4.6 ng/kg bw/day (Σ_6 iPCBs) is due to the
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11 consumption of fish and fish products.
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16 A Dutch intake assessment of Σ_7 iPCBs via the whole diet resulted in following
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18 estimated median intake: 4.8 ng iPCBs/kg bw/day (Baars et al., 2004; Bakker et al.,
19
20 2003). At the 90th percentile, an intake of 8.6 ng iPCBs/kg bw/day was estimated. In
21
22 France, the average intake of Σ_7 iPCBs among French high seafood consumers (Calipso
23
24 Study) was estimated to be 57 ng/kg bw/day through seafood consumption only (Sirot et
25
26 al. 2006). Recent European studies estimated the average daily intake of total non
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28 dioxin-like PCBs for adults to be in the range of 10-45 ng/kg bw/day (EFSA, 2005).
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34 35 *Risk evaluation*

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37 Non dioxin-like PCBs are less toxic than PCDD/Fs and dioxin-like PCBs. Nevertheless,
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39 it is recommended that the intake is as low as possible. Unlike for dioxin-like
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41 substances (Tolerated Daily Intake (TDI) = 1 - 4 pg TEQ/kg bw/day)(Scientific
42
43 Committee on Food 2001) or total PCBs (TDI = 20 ng/kg bw/day, in Aroclor
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45 Equivalent)(WHO 2003), no specific health based guidance value (e.g. a tolerated daily
46
47 or weekly intake, TDI or TWI), has been proposed for the non-dioxin like PCBs only
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49 (EFSA, 2005). The major problem encountered was that it is very difficult to distinguish
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51 between effects of non dioxin-like PCBs and effects of dioxin-like PCBs and PCDD/Fs
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53 that may be part of PCB mixtures. No definite relationship, however, has been found
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60 between levels of non dioxin-like PCBs and levels of dioxin-like PCBs and PCDD/Fs in

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4 these mixtures. Only occasionally a certain relationship could be found, e.g. in the PCB
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6 animal feed contamination case in Belgium in 1999 or in geographically defined
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8 sampling areas (EFSA, 2005; Vrijens et al., 2002).
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14 The WHO (2003) proposed a TDI for total PCBs, expressed in Aroclor equivalent, of
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16 20 ng/kg bw/day, while Sirot et al. (2006) stated that the concentration of Σ_7 iPCBs
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18 must be multiplied by two to be expressed in Aroclor equivalent. If our calculated
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20 exposure (the exposure of Σ_7 iPCBs multiplied by two) is compared with the TDI, it can
21
22 be seen that more than 30 % of the eel consumers of the background population exceeds
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24 this TDI, without taking in account other PCB sources. In comparison: between 70 %
25
26 and 99 % of the recreational fishermen exceed this TDI, depending on the consumption
27
28 scenario used.
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36 In a recent publication, a statistically significant relationship has been observed between
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38 individual dioxin-like PCBs and total PCBs, measured in a number of fishes, caught
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40 mainly in Canada and Northern America (Bhavsar et al. 2007). This correlation can be
41
42 an interesting application for risk assessment estimations executed in that region.
43
44 However, it has not been demonstrated that this relationship is also valuable in other
45
46 geographical regions. In contrast, clear spatial and temporal variations have been
47
48 observed in the ratio of PCB118 to the sum of the remaining 6 iPCBs in eel in Flemish
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50 water bodies (Goemans and Belpaire 2005). Therefore, this extrapolation has not been
51
52 used in the current estimation, since this paper handles the intake of eel, locally caught
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54 in Belgium.
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4 EFSA concluded that the margin of body burden (MoBB) – which was calculated by
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6 comparing the body burden (BB) in the rat at the no observed adverse effect level
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8 (NOAEL) of 500 µg/kg bw (liver and thyroid toxicity) with the estimated median
9
10 human BB for total non dioxin-like PCBs (48 µg/kg bw) in the general population – was
11
12 about 10. We do not know how much PCBs the fishermen ingest via the total diet, but
13
14 since the levels of intake via eel in scenario A1 and A2 are respectively 50 and 25 times
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16 higher than the intake of the background population, BB might be quite higher and
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18 reach levels that become toxicologically relevant.
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26 Since other animal based food items are very likely to contain some concentration of
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28 iPCBs, it, therefore, remains advisable to maintain the catch-and-release obligation for
29
30 eel and to sensitize the recreational fishermen about the contamination problem of eel in
31
32 the Flemish waters.
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38 Attention has to be paid to the background population too, since high eel consumers
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40 might also be at risk. In other countries, e.g. the USA, advisories on fish consumption
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42 were formulated, especially focussing on pregnant women, young children (under 15)
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44 and women of childbearing age. (MDCH Environmental and occupational
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46 epidemiology division 2004; Scientific Advisory Committee on Nutrition and Food
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48 Standard Agency 2004; US EPA 2005; US EPA and US FDA 2004). Also, the Swedish
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50 National Food Administration has recommended pregnant and lactating women to
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52 refrain from eating some predatory species, including eel (Bjornberg et al. 2005).
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59 **Conclusion**

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5 In conclusion, the intake of the Σ_7 iPCBs via the consumption of self-caught eel seems
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7 to be at a level of high concern. Further monitoring seems appropriate. Although risk
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9 assessment would be easier if, in analogy with PCDD/Fs and dioxin-like PCBs, a
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11 reference TDI or TWI could be established for the Σ_7 iPCBs only, it is very unlikely that
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13 this will be possible in the near future (EFSA, 2005). In the meantime, it should be
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15 advised to maintain the public health measure of preventing fishermen from consuming
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17 their self-caught fish. The catch-and-release obligation should be maintained and
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19 supervised (more) carefully.
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38 Technology in Flanders (IWT-Vlaanderen).
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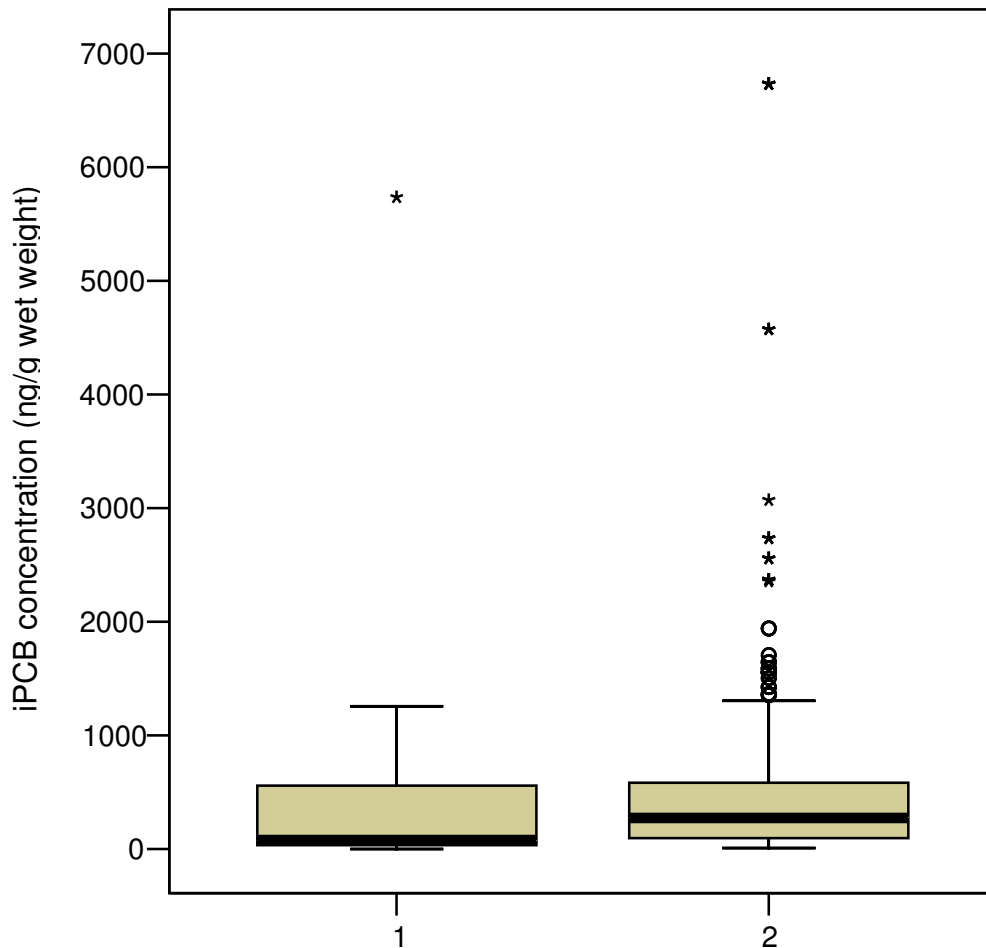
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Table 1. Estimated intake of the Σ_7 iPCBs (ng/kg bw/d) for the background population and the recreational fishermen. The estimates for the fishermen are presented for the different consumption scenarios (A1, A2, B1, B2).

Estimated iPCB intake (ng/kg bw/d)						
Percentile	Background	Recreational fishermen				
		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 1. Box and Whisker plots¹ for concentrations of the Σ_7 iPCBs (ng/g wet weight), analysed in (1) commercially available eel (n= 80) and (2) eel in Flemish waterbodies (n= 261).



¹ 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).

Figure 2. Fitted cumulative distribution functions for the concentrations of the Σ_7 iPCBs (ng/g wet weight) for eel in Flemish waterbodies and commercially available eel.

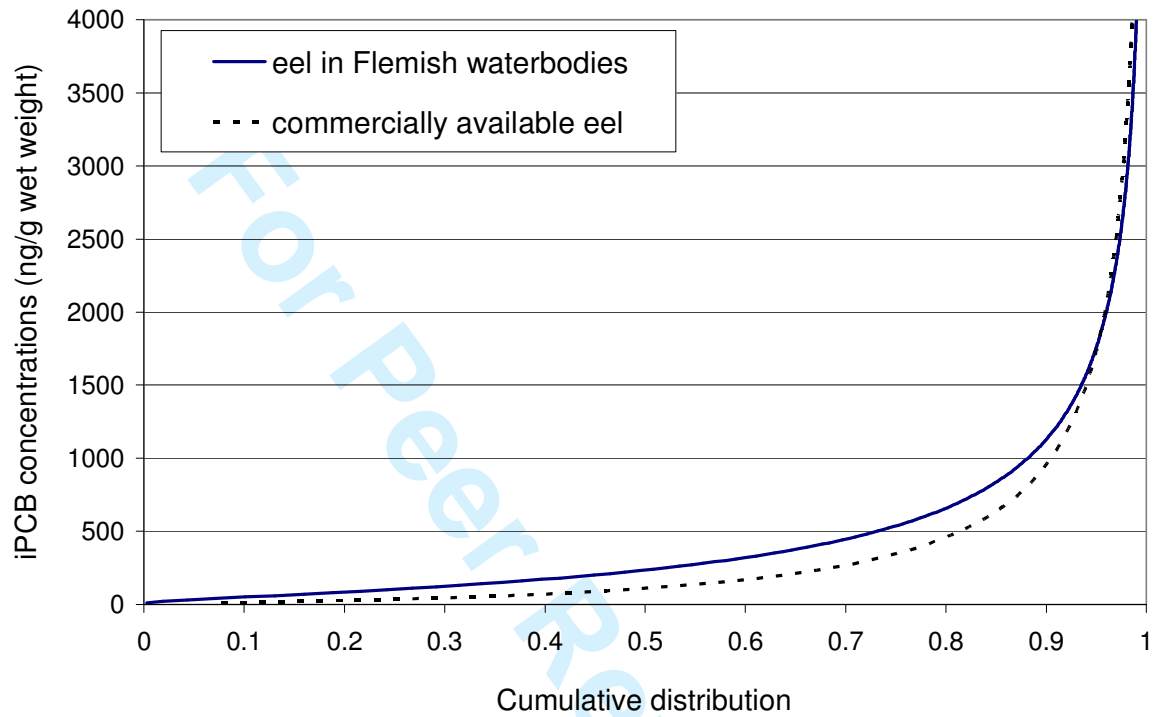


Figure 3. Cumulative distribution functions of the estimated intake of the Σ_7 iPCBs (ng/kg bw/d) for the background population and the recreational fishermen. The results for the fishermen are presented for the different consumption scenarios (A1, A2, B1, B2).

