

Summary

1 Introduction

Evaluation of the results of a former survey on chemical contamination of the foodstuffs most frequently consumed in Catalonia over the 2000-2002¹ period showed that seafood was an important contributor of environmental contaminants in diet. For this reason, considering the large diversity of fish species consumed in Catalonia, we decided it was important to characterize more precisely contaminant intake through fish and shellfish in diet.

2 Objectives

- To characterize the dietary intake of the following contaminants: arsenic, cadmium, mercury, lead, dioxins and furans, polychlorinated biphenyls, polycyclic aromatic hydrocarbons, hexachlorobenzene, polybrominated diphenyl ethers, polychlorinated diphenyl ethers and polychlorinated naphthalenes; from fish and shellfish, amongst the population of Catalonia
- To identify the edible marine species responsible for the largest intake of contaminants.
- To assess the health risks posed by the intake of these contaminants.
- To monitor food contamination and food safety levels.

3 Materials and methods

3.1 Type of study

As in the 2000-2002 total diet study (TDS), a mixed technique was used. Based on the features of the individual foods analyzed, this technique incorporates certain aspects of the market basket studies to analyze composite samples from the same species.

3.2 Selection of contaminants

The contaminants selected are the same as those analyzed under the 2000 TDS. They are listed in Section 2.

3.3 Selection of fish and shellfish species

The 14 edible marine² species to be analyzed were selected based on the Health Department's 2002-2003 Nutritional Survey of Catalonia (Encat), nutritional survey

on children (enKid) and national studies of food consumption carried out by the Spanish Ministry of Agriculture, Fisheries and Food.

3.4 Sampling and preparation

A total of 840 individual samples were analyzed, including 60 samples from each species purchased in six towns all over Catalonia.

3.5 Analytical methods

Metal determination was carried out by the Spectroscopy Laboratory at the University of Barcelona's Scientific-Technical Services. Organic contaminants (dioxins and furans, polycyclic aromatic hydrocarbons, hexachlorobenzene and polychlorinated naphthalenes) were analyzed at the SGS Control-Co. M.b.H Laboratory in Hamburg (Germany).

3.6 Population groups studied

As in the 2000-2002 TDS and according to the World Health Organization (WHO) guidelines, the age groups studied represent the standard individual among the Catalan population, as well as other groups of population that have, or may have, different patterns of consumption.

3.7 Data on daily fish and shellfish consumption

The data were gathered from the 2002-2003 Nutritional Survey of Catalonia and from national food consumption studies carried out by the Spanish Ministry of Agriculture, Fisheries and Food as compiled in *La alimentación en España*.

3.8 Estimate of daily intake of contaminants through fish and shellfish consumption

The daily intake of a contaminant from seafood in diet was calculated for each fish or shellfish studied by multiplying the concentration found in it by the amount consumed daily and finally adding them together.

3.9 Estimate of results below the limit of detection

Analytical results that were below the analytical limit of detection (LoD) were considered to be half this limit.

3.10 Variation 2000-2005

Average concentrations of each contaminant were compared, taking into account the fact that 5 marine edible species were studied in 2000 whereas there were 14 in 2005. A comparison was also made between the three³

1. Chemical contaminants: a total diet study in Catalonia. Catalan Food Safety Agency. June 2005.

2. Sardine, tuna, anchovy, mackerel, swordfish, salmon, hake, red mullet, sole, cuttlefish, squid, clam, mussel and shrimp.

3. Sardine, hake and mussels.

species common to both studies, by evaluation of the variation observed. The daily intake of each contaminant was estimated according to the methodology described in previous sections.

4 Arsenic

4.1 Results

Table 8 shows the concentrations found in the fish and shellfish analyzed. The highest concentrations corresponded to red mullet, followed by prawns and sole. The lowest levels corresponded to tuna and salmon.

4.2 Estimated daily intake

The estimated intake of arsenic through fish and shellfish consumption was 253.14 µg/day (Table 9). The highest proportion of this intake corresponded to hake, followed by sole and prawns. The lowest proportions of daily intake corresponded to swordfish and clams. White fish contributed 52% of the total intake, whilst oily fish and shellfish contributed 22% and 26% respectively (Figure 1).

4.3 Estimated daily intake by population group

Table 10 shows the estimated intake of total arsenic and inorganic arsenic in the different population groups.

4.4 Risk assessment

For all population groups, estimated inorganic arsenic intake was well below the toxicological safety level of 15 µg/kg/week (Table 11). In order to assess arsenic intake through dietary consumption of seafood, an estimate based on the 2000 TDS was made. This estimate indicates that total dietary intake in 2005 was below the maximum level established by the FAO-WHO Mixed Committee of Experts on Food Additives and Contaminants (JECFA).

4.5 Variation 2000-2005

The average arsenic concentrations found in all fish and shellfish analyzed under this study was double the amount observed in 2000 (Figure 2). By comparing the seafood common to both studies, it is noted that arsenic levels in hake doubled, whilst levels in mussels and sardines did not vary significantly (Figure 3). Total estimated intake of arsenic through fish consumption for a standard individual in Catalonia in 2005 was similar to the intake estimated in 2000.

5 Cadmium

5.1 Results

Table 12 shows the concentrations found in the fish and

shellfish analyzed. The highest cadmium concentrations were found in clams and mussels.

5.2 Estimated daily intake

Estimated intake of cadmium through fish and shellfish consumption was 1.41 µg/day (Table 13). The largest proportion of this intake corresponded to cuttlefish (Figure 4). Crustaceans and mollusks contributed 66% of total intake, followed by oily fish and white fish, with 16% and 18% respectively.

5.3 Estimated daily intake by population group

Table 14 shows estimated cadmium intake in the different population groups.

5.4 Risk assessment

Estimated cadmium intake through fish and shellfish consumption was below the toxicological safety level (Table 15) in all population groups. In order to assess cadmium intake through seafood in diet, an estimate based on the 2000 total diet study was made. This estimate indicates that total dietary intake in 2005 was below the maximum level established by JECFA.

5.5 Variation 2000-2005

Comparison of the latest figures with the results obtained from the 2000 study (Figure 5) show no differences in cadmium concentrations. When seafood common to both studies is compared, a slight increase in cadmium concentration levels in both hake and sardines is observed, whilst there is no significant difference in concentrations found in mussels (Figure 6). Total estimated cadmium intake through dietary consumption of fish and shellfish by a standard individual in Catalonia in 2005 was lower than the intake estimated in 2000.

6 Mercury

6.1 Results

Table 16 shows the concentrations found in the fish and shellfish analyzed. The highest levels corresponded to swordfish and tuna, the lowest to cuttlefish, clams and mussels. All concentration levels, with the exception of those found in swordfish, were below the limits established by the European Community.

6.2 Estimated daily intake

Estimated mercury intake through fish and shellfish consumption was 12.61 µg/day (Table 17). The highest

proportions of this intake corresponded to tuna and hake. Though the highest mercury concentrations were found in swordfish, consumption of this fish species is low (0.06 g/day). This does not, therefore, represent a significant proportion of intake in overall terms. Oily fish, mainly tuna, contributed 58% of total intake, whilst white fish contributed 35% (Figure 7).

6.3 Estimated daily intake by population group

Table 18 shows estimated mercury intake in the different population groups.

6.4 Risk assessment

Table 19 shows estimated mercury and methyl mercury intake through seafood consumption in the different population groups, expressed as a function of body weight. In order to assess total mercury intake through seafood in diet, an estimate based on the 2000 TDS was made. This estimate indicates that total dietary intake of these contaminants in 2005 was below the maximum level established by JECFA. However, in children, the figure was slightly higher than this maximum level. Moreover, methyl mercury intake in children was found to be higher than the maximum level established by JECFA.

6.5 Variation 2000-2005

Average mercury concentrations in all fish and shellfish analyzed under the 2005 survey increased in comparison to those found in the 2000 TDS (Figure 8). This is basically due to the fact that the later survey includes predator fish, which have greater capacity to accumulate mercury. Comparing the species concurrent in both studies, it is noted that the most significant increase corresponds to hake, whilst mussels show no significant variation (Figure 9). Total estimated mercury intake through fish and shellfish consumption for a standard male adult in Catalonia was 12.61 µg/day. This figure is slightly higher than the intake estimated in 2000, which was 8.9 µg/day.

7 Lead

7.1 Results

Table 20 shows the lead concentrations found in the different fish and shellfish species analyzed.

7.2 Estimated daily intake

Estimated lead intake through fish and shellfish consumption was 2.55 µg/day (Table 21). By far the largest proportion of this intake corresponded to hake, whilst

the lowest amounts corresponded to swordfish. White fish contributed 53% of total intake, whilst oily fish and shellfish contributed 26% and 21% respectively (Figure 10).

7.3 Estimated daily intake by population group

Table 22 shows estimated intake of lead in the different population groups.

7.4 Risk assessment

Table 23 shows estimated daily lead intake through fish and shellfish consumption in the different population groups, expressed as a function of body weight. In order to assess lead intake through dietary consumption of fish, an estimate based on the 2000 TDS was made. This estimate shows that total dietary intake in 2005 was below the maximum level established by JECFA.

7.5 Variation 2000-2005

Average overall lead concentrations in the fish and shellfish analyzed under this study was slightly lower than those found in the 2000 survey (Figure 11). Comparing the species concurrent to both studies, no variation in lead concentrations in mussels is found, whilst significant increases are found in hake and sardines compared to the 2000 TDS (Figure 12). Total estimated lead intake through fish consumption for a standard individual in Catalonia was lower in 2005 than that estimated in 2000.

8 Dioxins, furans and polychlorinated biphenyls

8.1 Dioxins and furans (PCDD/PCDF)

8.1.1 Results

Table 24 shows the concentrations of congeners found in the seafood analyzed. The highest levels corresponded to red mullet, followed by salmon, mackerel and sardines. Generally speaking, higher concentrations were found in oily fish than in white fish, crustaceans or shellfish.

8.1.2 Estimated daily intake

Daily intake of dioxins and furans through seafood consumption was 7.68 pg WHO-TEQ/day (Table 25). The highest levels corresponded to tuna, sardine and sole. The highest proportion (63%) of the total TEQ (toxicity equivalence) ingested corresponded to oily fish, whilst white fish contributed 24% (Figure 13).

8.1.3 Estimated daily intake by population group

Table 26 shows estimated daily intake of dioxins and furans in the different population groups.

8.1.4 Risk assessment

Estimated daily intake of dioxins and furans (Table 27) in an adult male through dietary consumption of fish and shellfish is 0.11 pg WHO-TEQ/kg. This figure lies within the lower ranges of the amount established by the WHO as the tolerable daily intake for dioxins and furans and dioxin-like polychlorinated biphenyls (1-4 pg WHO-TEQ/kg/day). In order to assess dioxin intake through dietary consumption of fish, an estimate based on the 2000 TDS was made. This estimate indicates that total dietary intake in 2005 was below the maximum level recommended by the WHO.

8.1.5 Variation 2000-2005

Average dioxin concentrations found in fish and shellfish overall decreased by 50% compared to the concentrations found in 2000 (Figure 14). This trend is maintained when the average concentrations for the three species common to both studies are compared (Figure 15). For an adult male, estimated daily intake of dioxins and furans through fish and shellfish consumption in 2000 was 28.74 pg WHO-TEQ/day, whilst the 2005 figure falls to 7.68 pg WHO-TEQ/day.

8.2 Polychlorinated biphenyls (PCBs)

8.2.1 Results

Table 28 shows the PCB concentrations found in the seafood samples analyzed. Of the 18 congeners analyzed, 12 are currently considered dioxin-like. The results, expressed in ng WHO-TEQ/kg of fresh fish, correspond to concentrations of dioxin-like polychlorinated biphenyls. The highest concentrations found corresponded to red mullet, sardines and anchovies, whilst the lowest levels of these contaminants were found in prawns and cuttlefish. Although it is amongst those found in the lowest concentrations, PCB#126 is the contaminant that contributes the highest proportion of total TEQ, as its toxic equivalence factor is higher than the others. Figure 16 shows the contribution of dioxin-like polychlorinated biphenyls to the total TEQ calculated, expressed as a percentage.

8.2.2 Estimated daily intake

Table 29 shows estimated daily intake of dioxin-like polychlorinated biphenyls in an adult male. Estimated intake through dietary consumption of fish and shellfish was 49.37 pg WHO-TEQ/day in 2005. Tuna contributed the largest proportion of this total intake, followed by hake and sardines. Figure 17 shows the percentage contribution made to the overall figure for congeners by

dioxin-like polychlorinated biphenyls. It is noted that the highest proportion, 69%, corresponded to PCB#126. Oily fish, particularly tuna, contributed 66% of dioxin-like polychlorinated biphenyl intake, whilst white fish contributed 26% (Figure 18).

8.2.3 Estimated daily intake by population group

Table 30 shows the estimated daily intake of dioxin-like polychlorinated biphenyls in the different population groups, by age and sex.

8.2.4 Risk assessment

Table 31 shows the estimated daily intake of dioxin-like polychlorinated biphenyls through dietary consumption of fish and shellfish in the different population groups, expressed as a function of body weight. For an adult male, intake is estimated at 0.71 pg WHO-TEQ/kg/day. To assess dietary intake through seafood, an estimate based on the 2000 TDS was made. This estimate indicates that total dietary intake in 2005 was 1.67 pg WHO-TEQ/kg/day.

8.2.5 Variation 2000-2005

A comparison was made of the figures for the congeners analyzed in the two surveys (Figure 19). The levels observed in the latest study were slightly higher than in 2000. However, taking into account only the species common to both, a slight decrease in concentrations was observed in 2005 compared to the earlier study. Figure 20 shows the concentrations observed for each species.

In 2000, intake of dioxin-like polychlorinated biphenyls through seafood consumption was 40.80 pg WHO-TEQ/day, whilst in 2005 the figure stood at 82.9 pg WHO-TEQ/day.

8.3 Dioxins and furans, and dioxin-like polychlorinated biphenyls. Overall assessment

8.3.1 Combined concentrations

Table 32 shows the total concentrations of dioxins, furans and polychlorinated biphenyls found. Concentrations in all samples analyzed were below the maximum limits established by law.

8.3.2 Estimated combined daily intake

Table 33 shows the estimated daily intake for an adult male to be 57.05 pg WHO-TEQ. Dioxin-like polychlorinated biphenyls make up 87% of this intake. The highest concentrations correspond to oily fish, followed by white fish.

8.3.3 Estimated combined daily intake by population group

Figure 21 shows the levels found for combined intake in

the different population groups, as well as the contribution of dioxins and furans and dioxin-like polychlorinated biphenyls to daily intake in each type of fish. The figure clearly shows that dioxin-like polychlorinated biphenyls account for a large proportion of total intake.

8.3.4 Combined risk assessment

Table 34, containing figures on estimated intake in the different population groups, shows that children represent the highest intake group. To assess intake of these contaminants through seafood in diet, an estimate based on the 2000 TDS was made. This estimate indicates that total dietary intake in 2005 was 2.73 pg WHO-TEQ/kg/day.

8.3.5 Variation 2000-2005

Taking into account only concentrations of congeners common to both studies, a decrease from 1.23 to 0.97 ng WHO-TEQ/kg is observed. Comparing the species concurrent to both studies, a significant decrease in concentrations of both mussels and sardines is found. Combined estimated intake in 2005 was 48.48 (57.05) pg WHO-TEQ/day, whilst in the 2000 survey this figure was 111.61 pg WHO-TEQ/day.

9 Polycyclic aromatic hydrocarbons (PAH)

9.1 Results

Table 35 shows the concentrations of polycyclic aromatic hydrocarbons found in the fish and shellfish analyzed. The highest levels corresponded to mussels, clams and prawns, whilst the lowest concentrations were found in white fish, cuttlefish and squid.

Naphthalene and phenanthrene were the hydrocarbons found in the highest levels, whilst the lowest concentrations in most species analyzed corresponded to dibenzo(a,h)anthracene (Figure 22). Compounds considered potentially carcinogenic contributed 21.8% of the polycyclic aromatic hydrocarbons found (Figure 23).

9.2 Estimated daily intake

Table 36 shows estimated daily intake of polycyclic aromatic hydrocarbons in an adult male. Total estimated intake through fish and shellfish consumption was 0.342 µg/day. The largest proportion of this consumption corresponded to prawns and hake.

Estimated intake of benzo(a)pyrene is 7.791 ng/day. Figure 24 shows the contributions of the different species to estimated daily intake of this contaminant as a per-

centage. The highest proportions of estimated daily intake of polycyclic aromatic hydrocarbons corresponded to crustaceans and shellfish (40%, Figure 25).

9.3 Estimated daily intake by population group

Table 37 shows estimated intake of polycyclic aromatic hydrocarbons in the different population groups.

9.4 Risk assessment

Compounds for which reference doses are established were assessed separately and individually. The results show daily intake well to be below the established limits for all population groups (Table 38). To assess intake of benzo(a)pyrene equivalent through dietary consumption of fish, an estimate based on the 2000 TDS was made. This estimate indicates that total dietary intake in 2005 was 0.23 µg/day.

9.5 Variation 2000-2005

The overall average concentration of polycyclic aromatic hydrocarbons found in the fish and shellfish analyzed under this study is practically unchanged compared to that observed in the 2000 survey (Figure 26). Comparing the species common to both studies (Figure 27), however, a significant increase is noted in concentrations found in mussels, whilst a decrease of nearly 50% is observed in sardines. The total estimated intake of polycyclic aromatic hydrocarbons through fish consumption for a standard individual in Catalonia 2005 was 0.342 µg/day, compared to 0.73 µg/day in 2000.

10 Hexachlorobenzene

10.1 Results

Table 40 shows the concentrations observed in the different fish and shellfish species analyzed. The highest levels corresponded to salmon and mackerel, whilst the lowest levels corresponded to cuttlefish and mussels.

10.2 Estimated daily intake

Table 41 shows that estimated daily intake of hexachlorobenzene for an adult male through fish and shellfish consumption was 14.64 ng/day. The largest proportion of this intake corresponded to salmon and sole. Oily fish contributed 53%, and white fish 43%. Crustaceans and shellfish contributed 4% of total hexachlorobenzene intake (Figure 28).

10.3 Estimated daily intake by population group

Table 42 shows the estimated intake of hexachlorobenzene in the different population groups.

10.4 Risk assessment

Table 43 shows estimated daily intake of hexachlorobenzene through dietary consumption of fish and shellfish in the different population groups, expressed as a function of body weight. To assess hexachlorobenzene intake through dietary consumption of fish, an estimate based on the 2000 total diet study was made. This estimate shows that total dietary intake in 2005 was well below the reference dose established by the American Environmental Protection Agency (EPA).

10.5 Variation 2000-2005

The average overall concentration of hexachlorobenzene in the fish and shellfish analyzed as part of this study is 330 ng/kg of fresh fish, compared to 256 ng/kg of fresh fish found in the 2000 survey (Figure 29). Comparing the species common to both studies, we note a huge decrease in hexachlorobenzene levels in sardines. Levels in hake are higher, whilst there is no significant variation in the case of mussels (Figure 30). Total estimated hexachlorobenzene intake through fish consumption for a standard individual in Catalonia in 2005 was 14.70 ng/day. This figure is considerably lower than estimated intake in 2000, which was 23.58 ng/day.

11 Polybrominated diphenyl ethers (PBDE)

11.1 Results

Table 44 shows the concentrations of polybrominated diphenyl ethers found in the fish and seafood sampled. The highest concentrations of polybrominated diphenyl ethers corresponded to salmon and mackerel. The predominant homologues were tetra-BDE and penta-BDE.

11.2 Estimated daily intake

Table 45 shows that daily intake of polybrominated diphenyl ethers for an adult male is 26.47 ng/day. The largest proportion of polybrominated diphenyl ethers consumed through diet corresponded to tuna, followed by salmon and hake. Figure 31 shows the distribution of the different homologues in the total intake of polybrominated diphenyl ethers through dietary consumption of fish and shellfish. The largest proportion correspon-

ded to tetra-BDE, which contributed 54% of total intake. Figure 32 shows the contributions made by the different types of fish to this daily intake of polybrominated diphenyl ethers. Amongst white fish, particular mention should be made of red mullet; although it is the species with the highest concentration of these compounds, it does not make a particularly significant contribution to daily intake, as consumption levels of this fish are low.

11.3 Estimated daily intake by population group

Table 46 shows the estimated daily intake of polybrominated diphenyl ethers through fish and shellfish consumption in diet by the different population groups.

11.4 Risk assessment

Table 47 shows estimated daily intake of polybrominated diphenyl ethers through seafood consumption in the different population groups, expressed as a function of body weight. Intake is estimated to be 0.38 ng/kg per day in an adult male. To assess polybrominated diphenyl ether intake through dietary consumption of fish, an estimate based on the 2000 TDS was made. This estimate indicates that total dietary intake in 2005 was below the reference dose established by the EPA.

11.5 Variation 2000-2005

The average concentration of polybrominated diphenyl ethers found in all the fish and shellfish analyzed under this study was 563.9 ng/kg, compared to 333.9 ng/kg in the 2000 survey (Figure 33). This difference is reduced when comparison is restricted to the three fish species concurrent in both studies (Figure 34). Concentrations found in sardines show a decrease compared to the earlier survey, whilst levels were found to have increased for hake and mussels. Dietary intake of polybrominated diphenyl ethers through fish was 30.7 ng/day in 2000 and 26.5 ng/day in 2005.

12 Polychlorinated diphenyl ethers (PCDE)

12.1 Results

Table 48 shows the concentrations of polychlorinated diphenyl ethers found in the fish and shellfish analyzed. The highest concentration corresponded to red mullet, though it should be noted that, generally speaking, higher concentrations were found in oily fish than in either white fish or shellfish. The predominant homologues in nearly all samples were hexa-CDE and hepta-CDE,

whilst tetra-CDE were the lowest contributors to the overall presence of PCDE. In clams and mussels, the homologue profile is clearly different from other samples studied, as in these two species tetra-CDE account for the highest concentrations.

12.2 Estimated daily intake

Estimated daily intake of PCDE in an adult male was 50.24 ng/day (Table 49). The largest proportion of this intake corresponded to tuna, followed by hake and sardines. Figure 35 shows the proportions of polychlorinated diphenyl ether intake corresponding to the different homologues. The predominant homologues are hexa-CDE and hepta-CDE. Oily fish contributed 64% of total intake of polychlorinated diphenyl ethers, whilst swordfish contributed a very small proportion of the total, as both concentrations of polychlorinated diphenyl ethers in this species and daily consumption levels are low. White fish contributed 27% of total intake (Figure 36).

12.3 Estimated daily intake by population group

Table 50 shows estimated daily intake of polychlorinated diphenyl ethers through dietary consumption of fish and shellfish the different population groups.

12.4 Risk assessment

Table 51 shows estimated daily intake of polychlorinated diphenyl ethers through fish and shellfish consumption per kilogram of body weight in the different population groups. For an adult male, daily intake is estimated to be 0.72 ng/kg. Due to the lack of data on the toxicity of these contaminants, safety levels have not yet been established. It is, therefore, not possible to assess the risk to human health posed by dietary intake of polychlorinated diphenyl ethers.

12.5 Variation 2000-2005

The overall average concentration of polychlorinated diphenyl ethers in fish and shellfish analyzed under this study was 1,094.7 ng/kg, whilst for the earlier survey the figure was 417.7 ng/kg (Figure 37). No appreciable differences are found when the species common to both studies are compared, therefore it is assumed that the other species are responsible for the difference observed (Figure 38). This is demonstrated in Table 48, which shows relatively significant concentrations in the other oily fish as well as in red mullet. No significant changes are observed in estimated intake of polychlorinated diphenyl ethers through fish and shellfish consumption, which was 38.4 ng/day in 2000, compared to an estimated figure of 50.24 ng/day in the more recent study.

13 Polychlorinated naphthalenes (PCN)

13.1 Results

Table 52 shows the concentrations of polychlorinated naphthalenes found in the fish and shellfish analyzed. The highest concentration of these contaminants corresponded to salmon, followed by mackerel. The lowest levels were found in cuttlefish and prawns. By congener group, pentaCN contributed the largest proportion (60%), followed by tetraCN (33%) (Figure 39).

13.2 Estimated daily intake

Table 53 shows estimated daily intakes. In adult males, estimated intake of polychlorinated naphthalenes through seafood in diet is 1.95 ng/day. The largest proportion of daily intake corresponded to salmon, followed by sole and tuna. The highest intake of polychlorinated naphthalenes corresponded to oily fish, which contributed 60% of the total, whilst white fish contributed 33% and shellfish 7% (Figure 40).

13.3 Estimated daily intake by population group

Table 54 shows estimated intake of polychlorinated naphthalenes in the different population groups.

13.4 Risk assessment

Table 55 shows estimated daily intake of polychlorinated naphthalenes through fish and shellfish consumption as a function of body weight in the different population groups studied. Due to the lack of data on the toxicity of these contaminants, safety levels have not yet been established for these contaminants. It is, therefore, not possible to assess the health risks posed by the dietary intake of polychlorinated naphthalenes.

13.5 Variation 2000-2005

No significant differences are found between the two studies as regards overall average concentrations of polychlorinated naphthalenes in fish and shellfish (Figure 41). Comparing the species common to both studies, a considerable decrease is noted in concentrations found in sardines, whilst much less significant differences are found between the two other species (Figure 42). A significant decrease in concentration levels between the two studies is noted: estimated intake of polychlorinated naphthalenes through dietary consumption of fish and shellfish in 2000 was 3.6 ng/day, compared to 1.95 ng/day in the latest study.

14 Conclusions

14.1 Concentrations of contaminants in seafood

Generally speaking, we should emphasize the higher concentrations of mercury, polychlorinated biphenyls found, whilst also noting, within the white fish group, the relatively high levels of contamination found in red mullet and the high mercury levels found in swordfish.

Regarding arsenic, cadmium, lead and polycyclic aromatic hydrocarbons, the higher concentrations found in crustaceans and shellfish should be noted, as well as the fact that, in the case of arsenic, the highest concentrations correspond to red mullet, whilst salmon is the most important contributor to lead intake through diet. On the other hand, concentrations are, generally speaking, lower in oily fish.

14.2 Estimated daily intake

By way of summary, in the 2005 study the seafood responsible for the highest contribution to daily intake of contaminants, corresponds to tuna for many contaminants, followed by hake for most of them and sole for some others.

It is noted that, for most contaminants or groups of contaminants, oily fish accounts for the highest proportion of intake, followed by white fish and crustaceans and shellfish.

14.3 Estimated daily intake by population group

It must be noted that the groups with the highest intake of contaminants per day through seafood consumption are adult men and women, and men aged over 65 years.

14.4 Risk assessment

Arsenic, cadmium, lead, polycyclic aromatic hydrocarbons (benzo(a)pyrene equivalent), hexachlorobenzene, dioxins and furans, dioxin-like polychlorinated biphenyls, dioxins and furans plus dioxin-like polychlorinated biphenyls, and polybrominated diphenyl ethers

Concentrations found were well below recommended reference values, both for intake from seafood consumption analyzed under this study and for the recalculation of total diet in 2000, with the sole exception of benzo(a)pyrene equivalent, which levels continue to be high, though fish is not the main contributor to this.

Mercury and methylmercury

Intake of mercury through fish consumption was below the toxicological safety level for all population groups. The highest concentrations intake of these contaminants

through seafood in diet were found in children, with 2.23 $\mu\text{g}/\text{kg}/\text{week}$. Considering this figure within the results obtained in the 2000 total diet study, we estimate a total dietary intake of 5.65 $\mu\text{g}/\text{kg}/\text{week}$. As regards methylmercury intake, children exceeded the tolerable intake level, established at 1.6 $\mu\text{g}/\text{kg}/\text{week}$, with intake estimates of 2.01 and 1.65 $\mu\text{g}/\text{kg}/\text{week}$ respectively.

14.5 Variation 2000-2005

Compared to the 2000 study, a decrease was observed in overall concentrations of dioxins and dioxins added to dioxin-like polychlorinated biphenyls. There were no significant variations in lead, cadmium and polycyclic aromatic hydrocarbon concentrations compared to those found in 2000. Concentrations of polychlorinated biphenyls, hexachlorobenzene and polychlorinated naphthalenes increased slightly compared to 2000. However, levels of arsenic, mercury and polybrominated and polychlorinated diphenyl ethers increased significantly, by approximately 50%.

Estimated intake of arsenic, mercury and polybrominated diphenyl ethers by an adult male also increased slightly, whilst the intake of other contaminants decreased.