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1.4 Environmental occurrence and exposure

1.4.7 Food products

(a) Context and significance

Since the early 1990s, food has been identified as the major route of human exposure to polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD, PCDF) but also to other lipophilic and persistent organochlorines such as PCB. In the population that is not exposed to other known sources, dietary intake contributes about 90–98% of the total daily intake of dioxin-like compounds and of this, food of animal origin contributes about 90% in European countries (Büchert *et al.,* 2001) (Päpke & Fürst, 2003) and is similar in other regions of the world (Schecter et al., 1997; Llobet et al., 2003a,b; Bocio and Domingo, 2005; Charnley and Doull, 2005; Huwe and Larsen, 2005).

Regarding specifically PCBs, for both the six indicator PCB~~non-dioxin-like~~ (PCB_6 ~~ndl-PCB~~[1]) and twelve dioxin-like PCB (dl-PCB) it is generally accepted that ingestion of especially fatty foodstuffs constitutes the major route of exposure (IARC, 1978; IPCS, 1993; EFSA, 2005a; Lindell, 2012), although inhalation can also be an significant source (see Section 1.4.0).

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Human food has become contaminated with PCB by three main routes:

a) uptake from the environment by fish, birds, livestock (via food-chains), and crops;

b) contamination of animal feed by regular practices or by accident;

c) direct contamination of food by accident.

PCB concentrations in food are presented in many different ways, making comparisons difficult. With respect to data reporting, the number of congeners analysed differs be-

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tween studies and often they are summed up according to groups, such as indicator-PCB, dl-PCB, or other number of congeners. When using TEFs, the scheme should be noted. Some studies report TEQ on the basis of bioassays such as CALUX biological equivalents (BEQ). Results have been reported on different reference units (wet weight, dry weight, or lipid weight). Further difficulties arise that different parts of especially fish or seafood were analyzed (muscle, liver, skin, etc.,); some also report on the basis of prepared food (to account for changes by cooking or frying). Finally, the objectives of a study may bias the sampling strategy; often towards higher concentrations.

One category of food is human milk. Due to its fat content, human milk can accumulate a large amount of PCB and thus, makes it an ideal matrix for the determination of PCB (and other lipophilic compounds) by applying a non-invasive sampling technique. In addition, human milk represents a good indicator for lipophilic unmetabolised PCBs stored in the body since fat is mobilised for the production of milk during lactation. Animal studies and mass balances for humans have revealed that large amounts of PCBs can be eliminated through lactation (referenced by Lindell et al. 2012).

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(b) PCB concentrations in food

Global and regional data~~**International and global data**~~

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Several reviews have been undertaken by scientists or governmental authorities to assess the concentrations of PCB in food at global, regional or national level. Some papers also summarize data from certain groups of populations having common characteristics, such as people living in the polar regions or high fish consumers. In addition, an abundance of papers is available that report case studies or single measurement campaigns. This section focuses on

[1] These do not exhibit the characteristics of the 12 dioxin-like PCB that have been assigned a TEF by WHO Expert Group. Often, and especially in the European food and feed legislation they are referred to as "non-dioxin PCB" or "NDL-PCB")

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the broader studies and reviews; thus, the findings are considered representative but far from complete.

Domingo and Bocio (2007) reviewed the concentrations of PCB and PCDD/PCDF in marine species and human intake through fish and seafood consumption (for detailed information, see Table). The studies will be presented in the different region-specific sections.

Hites *et al*. (2004a) analyzed farmed salmon obtained from farms in eight major salmon farming regions of the world, samples of five species of wild Pacific salmon, and retail market samples of farmed salmon obtained in 16 cities in Europe and North America (Hites et al., 2004a,b). PCDD/PCDF and PCB levels in salmon showed notable differences depending on the respective origin. They found that farmed salmon had significantly higher contaminant burdens than wild salmon, whereas farmed salmons from Europe were significantly more contaminated than those from South and North America (Hites et al., 2004a, c). This finding was opposite to the results from the European market (EFSA 2012).

Polar regions

The traditional food of the Arctic is an important source of nutrition and energy to indigenous peoples, besides being of high cultural value. These food items include lipid-rich tissue of high trophic level animals. Following long-range transport and biomagnification of PCBs in the Arctic marine food chain, PCBs accumulate in edible animals like fish, seals and whales (AMAP, 2004). This dietary exposure led to PCB levels in Arctic inhabitants which exceeded those at temperate~~oral~~ latitudes (Dewailly et al., 1993), but have been shown to decrease (AMAP, 2009). Likewise, PCBs in traditional food items have generally decreased (Rigét et al., 2010).

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In an older study on the traditional food of the Inuit of Québec, lipid-rich food (polar bear fat) reached ∑PCB10 concentrations of approximately 7 µg/g lipid, while seal blubber was somewhat lower (1 µg/g lipid) and Arctic char muscle had a concentration of 150 ng/g lipid

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(Dewailly et al., 1993). The female consumers of this food had higher PCB levels in milk than a group in Southern Québec.

Johansen et al. (2004) classified a wide range of traditional food products from West Greenland according to their ∑PCB10 content, which placed minke whale, beluga and narwhal blubber into the "very high" concentration group (> 500 ng/g) and halibut liver, kittiwake liver and muscle, minke whale skin and seal blubber into the "high" concentration group (50-500 ng/g). Compared with the marine animals, concentrations in food sources from the terrestrial environment were characterized as low. Based on these data and information on dietary habits, a daily PCB intake rate of 23 µg/day/person was calculated, which decreased to 3 µg/day/person if blubber food items were excluded from the diet.

Using the grouping presented by Johansen et al. (2004), traditional food items of the Dene/Métis of the Northwest Territory showed a number of foodstuffs in the 50-500 ng/g group, including cooked sucker flesh, raw beluga mattak (skin/blubber) and boiled Canada goose meat (Berti et al., 1998). A mean and median daily PCB intake of 23 ng/day/kg body weight (bw) and 11 ng/day/kg bw, respectively, was calculated and compared with a provisional tolerably daily intake rate of 1 µg/day/kg bw, used by Health Canada.

Africa

Loufty et al. (2007) determined the concentrations of PCDD/PCDF and dioxin-like PCBs in samples of fish and seafood (mullet fish, bolti fish, bivalves and crab) randomly acquired in local markets from Ismailia city, Egypt. The upper bound levels for PCDD/PCDF and dioxin-like PCBs ranged from 0.12 (crab) to 0.40 (mullet) pg WHO-TEQ/g ww, and from 0.14 (bivalves) to 0.76 (mullet) pg WHO-TEQ/g ww, respectively.

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North America

Hayward et al. (~~in press~~2007) reported the levels of PCDD/PCDF and PCBs in commercially wild caught and farm-raised fish (bluefish, "rockfish", wild caught Alaska King, Sockeye and Coho salmon, and farm-raised salmon) fillets purchased from markets in Maryland, Washington, DC, and North Carolina during April–August 2004. The highest and lowest PCB concentrations were found in a wild bluefish fillet (800 ng/g ww) and in a wild Coho salmon fillet (0.35 ng/g ww), respectively.

In domestic meat and poultry surveys conducted by the US. Department of Agriculture (USDA) in the mid-1990s, 2002–2003, and again in 2007–2008 (Huwe, 2010), mean concentrations of non-ortho-PCB appear to have declined in the 5 year span, except for WHO_{2005}-TEQ from *non*-ortho PCB in turkeys. The increase in turkeys is most likely due to a cluster of turkeys from California (USA) which had *non*-ortho PCBs TEQs ranging from 0.74 pg g^{-1} lipid to 4.42 pg g^{-1} lipid (for detail, see Table S1).

Schecter et al. (2010) detected six of seven non-dioxin-like PCB congeners in salmon and canned sardines, with PCB-153 and PCB-138 at highest levels (salmon: PCB-153, 1.2 ng/g ww; PCB-138, 0.93 ng/g ww; canned sardines: PCB-153 and PCB-138, 1.8 ng/g ww each). We measured PCB-153 and PCB-180 in hamburger meat at 1.2 and 0.21 ng/g ww, respectively. PCB-180 was present in peanut butter (0.43 ng/g ww) and ice cream (0.091 ng/g ww).

Brown et al. (2006) measured the levels of PCDD/ Fs and coplanar PCBs in 65 samples of a variety of fish collected from California coastal waters in 2000–2001. For all species from all sampling areas, the mean I-TEQ was 33.1 pg/g lipid for PCDD/PCDF and 109 pg/g lipid for thre~~the 3~~e *non*-ortho ~~coplanar~~ PCBs (77, 126, 169)[2]. In turn, Rawn et al. (2006) also analyzed the levels of PCDD/PCDF and PCBs in fish products available on the Canadian retail

[2] Sic : I-TEFs do not include PCB, it is assumed that WHO_{1998}-TEFs were used to calculate the TEQ

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market in 2002. The geometric means WHO-TEQ (pg/g whole weight) ranged from 0.06 (shrimp) and 0.08 (tilapia) to 0.92 (salmon).

Oceania (Australia/New Zealand)

During 2000-2001, 168 samples of 22 foods collected for the Australian Total Diet Survey were analyzed for dioxin-like PCB congeners (Table 1.mnA). The Australia-New Zealand agency compared these concentrations with those from other areas of the world (Table 1.mnB) (Food Standards Australia New Zealand (2004).

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Recent data are available from Padula et al. (2012) who reported PCB concentrations from composite samples of Australian farmed Yellowtail Kingfish (Seriola lalandi) (YTKF) (n = 27), Mulloway (Argyrosomus hololepidotus) (n = 6) and manufactured feed (n = 5). The mean concentration of PCBs (analytes included PCB-1, 3, 4, 15, 19, 28, 37, 44, 49, 52, 54, 70, 74, 99, 101, 104, 110, 138, 153, 155, 170, 180, 183, 187, 188, 194, 196, 199, 202, 205, 206, 208 and 209) in YTKF was 21 μg kg^{-1} (range 8.6–29) and in Mulloway was 5.4 μg kg^{-1} (mean 4.7– 6). The mean concentration of dioxin-like PCBs in YTKF was 2.1 pg TEQ g^{-1} (range 1.2– 2.8) and in Mulloway was 0.51 pg TEQ g^{-1} (range 0.41– 0.61).

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Asia

For Japan, ~~only~~ dl-PCB have been reported together with PCDD, PCDF on TEQ basis. A study sponsored by the Ministry of Health and Welfare of human~~breast~~ milk samples stored in Osaka Prefecture also showed a more than 50% decrease in PCDD/PCDF and three non-ortho substituted~~co-planar~~ PCBs between 1973 and 1996 (MoE/MoH 1999). The daily intake for the general population in Japan of 2.60 pg TEQ/kg b.w./day has been derived from a survey of food items on the market and typical consumption patterns as shown in Figure X. Of these, the vast majority, 2.41 pg TEQ/kg b.w./day is from ingestion of food whereas inhalation and

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soil ingestion contribute only with 0.19 pg TEQ/kg b.w./day. A "typical" Japanese person receives 120.7 pg TEQ *per* day through food consumption of fish/shellfish followed by meat/eggs.

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Food (Survey on the State of Food Contamination by Dioxins and Related Compounds, by the Ministry of Health and Welfare, 1997)

Food Group	Daily Consumption (National Nutrition Survey) A (g)	Dioxin Concentration B (pg/g)		Dioxin Intake C (pg)		Dioxin Daily Intake per kg Body Weight (pg/kg)
Fish and shellfish	97.0	0.776	A X B →	75.28	C / 50 kg →	1.506
Meat and eggs	129.0	0.174		20.87		0.417
Milk and dairy products	133.9	0.070		9.42		0.188
Green vegetables	98.8	0.050		4.94		0.099
Cereals, potatoes	166.2	0.025		4.21		0.084
Nonessential foods	182.4	0.007		1.31		0.026
Other vegetables, seaweeds	205.0	0.006		1.23		0.025
Rice	166.5	0.007		1.18		0.024
Sugar and sweets	34.2	0.020		0.70		0.014
Oils and fats	16.9	0.031		0.53		0.011
Processed foodstuffs	5.5	0.073		0.40		0.008
Beans and processed beans	72.3	0.006		0.43		0.008
Fruits	118.6	0.002		0.24		0.004
Water	600.0	0.00003		0.02		0.0004
Total	2,017.4			120.7		**2.41**

Intake of TEQs in Japan from sources other than food:

Environment

			Dioxin Daily Intake per kg Body Weight (pg/kg)
Atmosphere	• Atmosphere concentration: 0.55 pgTEQ/m³ "Monitoring of Hazardous Air Pollutants by Local Governments in 1997" • Daily intake by respiration: 15 m³	→	0.17
Soil	• Soil concentration: 22 pgTEQ/g "Comprehensive Pilot Study on Dioxins in 1997" • Oral daily intake of soil: (Children / Adults) : (150/50) to (260/100) mg/day • Additional exposures via lungs and skin are also considered	→	0.0024-0.021
			0.19
		Total Dioxin Intake	**2.60 pg/kg/day**

Fig. 1 Daily Intake of Dioxins and Related Compounds in Japan

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Liu et al. (2011) determined seven indicator PCB (PCB-28, 52, 101, 118, 138, 153, and 180) in marine fish from four areas of China (South China Sea, Bohai Sea, East China Sea, and Yellow Sea) using GC/NCI–MS and GC/ITMS, respectively. The sum for PCB_7 ranged from 0.3 ng g^{-1} ww to 3.1 µg g^{-1} ww, with median and mean values of 6.4 ng g^{-1} ww and 398 ng g^{-1} ww, respectively. High contributions of PCB-138 (32%) and PCB-153 (25%) were found in all samples. It was noted that the concentrations found in this study were higher than in other parts of the world. Interestingly, in all fish PCB_7 was strongly correlated with the sum of PBDE (r = 0.907, n = 44). Table Liu (Section TABLES)

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Yim et al. (2005) determined the levels of 22 PCB congeners in the muscles of sport and market fish in Korea's coastal waters. Mean (and range) concentrations of sport and market fish were 23.0 (4.48–95.6) and 8.91 (2.96–68.2) ng/g ww, respectively.

Moon and Ok (2006) found concentrations from 0.008 to 0.6 pg WHO-TEQ/g ww of dioxin-like PCBs in 40 species of marine organisms from Korean coastal waters. The estimated intake of dioxin-like PCBs was estimated to 0.40 pg WHO-TEQ/kg body weight/day. The contribution of dioxin-like PCBs through consumption of five fish and seafood groups to the total intake from all organisms (0.68 pg WHO-TEQ/kg body weight/ day) averaged 60%, being the relative contribution of each organism group in descending order: fish, crustaceans, cephalopods, bivalves and gastropods.

Nakata et al. (2002) found relatively high PCB concentrations in fish and seafood) collected in China in 2000–2001: from 0.20 (shrimp and mussel) to 2.5 (mackerel) ng/g ww. Dioxin-like PCB in various species of bivalves and gastropods collected from eight sampling sites along the Bohai Sea coastline (NE China) ranged from 0.87 to 7.71 pg/g lipid weight for bivalves and from 0.53 to 7.9 pg/g lw for gastropods, (Zhao et al., 2005).

In a survey by Yang *et al.* (2006), PCB concentrations in a wide variety of fish and shellfish collected from local supermarkets from three Chinese cities (Dalian, Tianjin and Shangai) showed that in most samples, PCB-138 and PCB-153 were dominant, followed by PCB-101 and PCB-180. The average PCB concentration was 3.60 ng/g ww, with the highest values corresponding to flathead flounder (8.04 ng/g ww) and white sardine from Shangai (5.60 ng/g ww), and the lowest to Pacific cuttlefish (0.83 ng/g ww) and white sardine from Dalian (1.11 ng/g ww). The estimated daily intake of PCBs through fish and shellfish, considering a body weight of 60 kg and an average consumption of 30.5 g/person/day, was 1.83 ng/kg body weight.

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In a total diet study in metropolitan Tokyo, Japan – years between 1999 and 2004 - Sasamoto et al. (2006) estimated the dietary daily intake of PCDD/PCDF and dioxin-like PCBs by analyzing 14 food groups (including fish and shellfish): WHO-TEQ$_{PCDD/PCDF/PCB}$ in fish and shellfish ranged from 0.98 to 0.91 pg WHO-TEQ/g in 1999 and 2004, respectively. PCB 126, followed by PCB 118, was the highest contributing congener, and accounted for over 40% of the daily intake of PCDD/PCDF and PCBs from fish and shellfish. In relation to the total dietary intake of all food groups, this amounts to a mean percentages of 90%.

Europe

For the 27 EU member States and Switzerland and~~d~~ Norway, the report "Scientific Opinion on the presence of dioxins (PCDD/PCDF) and dioxin-like PCBs (DL-PCBs) in commercially available foods for infants and young children" (EFSA CONTAM, 2012) was published by EFSA. Taking all food groups together, the P50, P90 and P95 were < 0.005 [< 0.005], 0.02 [0.01] and 0.03 [0.01] pg WHO_{2005}-TEQ g^{-1} wet weight (w.w.) for PCDD/PCDF. For the total TEQ, including PCDD, PCDF and dl-PCB, the P50, P90 and P95 for all food groups based on UB (LB) concentrations were 0.01 (< 0.005), 0.04 (0.02) and 0.07 (0.04) pg WHO_{2005}-TEQ g^{-1} w.w. The maximum UB concentrations in this data set were 0.08 pg WHO_{2005}-TEQ g^{-1} w.w and 0.19 pg WHO_{2005}-TEQ g^{-1} w.w for PCDD, PCDF, and dl-PCB, respectively.

Infant formula showed UB concentrations up to 0.06 pg WHO_{2005}-TEQ g^{-1} w.w. The highest concentrations, up to 0.19 pg WHO_{2005}-TEQ g^{-1} w.w. for the sum of PCDD/PCDF and dl-PCB, were found in ready-to-eat meals containing fish or meat.

The UB concentrations based on all accepted data were below the current MLs for foods for infants and young children of 0.2 pg WHO_{2005}-TEQ g^{-1} w.w. for the sum of PCDD/PCDF and dl-PCB, which are in force since 1 January 2012.

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The most recent, comprehensive assessment of PCB in food was undertaken by the European Food Authority (EFSA) in 2012 (EFSA, 2010; EFSA, 2012).

A total of 13 797 samples collected between 1995 and 2010 were available for dl-PCB and 19 181 samples for ~~ndl-PCB~~PCB6. More precisely, there were 10 468 food samples covering dl-PCB and 17 127 covering ~~ndl-PCB~~PCB6, and 3329 feed samples covering dl-PCB and 2054 covering ~~ndl-PCB~~PCB6.

The detailed assessment includes the following conclusions (EFSA, 2012):

- At least one quantified congener was found in almost each of a total of 13 797 samples analysed for dl-PCB, whereas at least one of the six indicator ~~ndl-PCB~~PCB6 was quantified in respectively 68.4% and 82.6% of the 2054 feed and 17 127 food samples analysed for their presence.
- Feed and food from animal origin contained higher concentrations of PCDD/PCDF and dl-PCB than foods from plant origin. The highest average concentrations were found for the food groups "meat from eels" and "Fish liver and derived products".
- The non-*ortho* PCB was the main contributor to the total toxicity equivalents (WHO_{2005}-TEQ_{total}) of dl-PCB, representing 21.0-74.5% of the total WHO_{2005}-TEQ_{total} (for details see Table S2 ~~and~~ Table S3 ~~in the Appendix~~).
- Concerning the ~~ndl-PCB~~PCB6, the PCB-153, followed by PCB-138 and PCB-180 represented altogether 36.9%-97.8% of the sum of the six ~~ndl-PCB~~PCB6, depending on the food and feed group (for details see Table S4). Graphic representation for dl-PCB and ~~ndl-PCB~~PCB6 is available in Fig. S1 and Fig. S2, respectively.
- The concentration of PCDD/PCDF and dl-PCB exceeded the permitted maximum level (ML) in 9.7% of the food samples and 2.3% of the feed samples. The concentration of the ~~six indicator ndl-PCB~~PCB6 exceeded the ML in 3.0% of the food samples and 2.4% of the feed samples.

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• For time trend analysis, three food groups were available: "Raw milk and dairy products," "Hen eggs and egg products" and "Muscle meat from fishes other than eels." A decrease in the concentrations of dl-PCB was observed over the years in all of them.

• The ~~six indicator ndl-PCB~~PCB6 were found to represent around 50% of the sum of the ~~ndl-PCB~~PCB6 which were measured in the food samples. In all food and feed groups, the concentrations of the ~~six indicator ndl-PCB~~PCB6 and dl-PCB were positively correlated. The correlation depended on the food groups and sometimes was influenced by outliers.

With respect to food categories, it was found that (EFSA, 2012):

• Meat from sheep contained on average less PCB than meat from bovine animals.

• Eggs coming from battery rearing contained significantly less PCB than those coming from free range, organic and outdoor growing production.

• Farmed salmon and trout contained on average less PCB than wild caught salmon and trout. Although in contrast to results from the U.S.A., this finding is well documented and can be explained by the fact that stringent legislation on maximum concentrations in feedingstuff placed on the European market is enforced since a decade; thus minimizing the intake of dl-PCB in farmed fish.

• Herring, salmon and trout coming from the Baltic region were more contaminated by PCB than those coming from other regions.

• Milk at farms had higher concentrations of PCDD/PCDF and dl-PCB than milk from bulk, whereas the inverse was observed for the indicator ~~ndl-PCB~~PCB6.

With respect to dietary exposure, it was found that (EFSA, 2012):

• The major contributor to total human exposure was milk and dairy products for almost all infant and toddler groups, whereas it was fish and seafood products for most of the ado-

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lescents, adults, elderly and very elderly groups. Meat and meat products also contributed significantly to total exposure.

• A general decrease in exposure to the sum of PCDD/PCDF and dl-PCB was observed between 2002–2004 and 2008–2010, estimated to be between 16.6% and 79.3% according to the different population groups.

• A decrease in exposure to the sum of the ~~six indicator ndl-PCB~~PCB6 was observed between 2002–2004 and 2008–2010 in most but not all population groups, estimated to be between 2.0% and 75.6%.

• The estimated decrease over time is most likely an effect of the European risk management measures to reduce the exposure of the European population, but could to some extent also be due to improvements in the analytical methods used and changes to sampling design of the monitoring programs throughout the years.

• The current results should be interpreted with some caution. On the one hand, food contamination and population exposure may have been overestimated due to targeted sampling. On the other hand, not all foods have been taken into account in the exposure assessment, which may have lead to an underestimation of total exposure in some population groups. Finally, regional variations have not been considered as all occurrence data were merged to represent an overall European situation.

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(b) Human Milk

Because of the importance of breast feeding for infants, contamination of human milk is of specific public concern. In 1984, the first measurements for background contaminated milk were reported for Sweden and Germany. The World Health Organization (WHO) introduced worldwide large measurement campaigns to determine the exposure of infants to dl-PCB (UNEP, 2001). To date, five rounds of exposure studies have been concluded (starting from 1987). The Stockholm Convention on Persistent Organic Pollutants (POPs) uses human milk

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and human blood as a matrix for human exposure to POPs. The objective is to determine changes in time to document the effectiveness of the implementation of the obligations under the Convention. Often, human milk from primaparae mothers (for detail, see UNEP 2012a, b) is preferred towards human blood since samples can be taken by non-invasive methods and the concentrations of PCB are easier to detect (due to the higher lipid content). It should be noted that the accessible donors are restricted to mothers and other populations cannot be included.

Worldwide exposure studies

The guidance document as relevant for the effectiveness evaluation of the Stockholm Convent has been applied – with slight changes – for five rounds of the WHO study. Here we list the results from samples in the period 2001-2011 (3rd to 5th rounds) for this global study. The results of the national pools that have been analysed by the WHO/UNEP reference laboratory for POPs in human milk are included in the next section. The results for China and Japan are shown separately because their samples are not included in the global studies.

For global assessment, the last three rounds of the UNEP/WHO survey on mothers' milk (Malisch *et al.*, 2013) had 84 data sets for assessment on national basis (one country, one pooled sample prepared from usually 50 individual mothers having delivered the first child per round). The following figures detail regional differences in the concentrations of mothers' milk for dl-PCB (Fig. 2) and indicator PCB (expressed as ΣPCB_6, Fig. 3). It should be noted that depending on the TEF scheme used, the concentrations for dl-PCB on TEQ basis may differ by 30% (lower if WHO_{2005}-TEFs are applied in contrast to WHO_{1998}-TEFs; range from 2% to 60%, see Table 1). For dl-PCB (on TEQ basis) and indicator PCB, the concentrations in Europe are considerably higher than in other regions of the world, indicating legacies from past exposures.

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It is noted that in Europe the concentrations in WHO-TEQ_{PCB} are significantly higher than in other regions. Whereas in Europe, the dl-PCB have a significant contribution to the WHO-TEQ_{total} their contribution is less in other regions. For ΣPCB_6 in Europe, the median of 115.3 ng g^{-1} lipid for is between 3.8-times and 10.7-times higher than in other regions, the maximum concentration was 502 ng g^{-1} lipid up to 14.9-times higher. The median in other regions is between 10.8 ng g^{-1} lipid and 30.7 ng g^{-1} lipid; maxima in other regions are between 37.1 ng g^{-1} lipid and 65.8 ng g^{-1} lipid.

The 2003 IPCS document (WHO, 2003) reports that average PCB concentration in human milk fat ranges from 0.5 mg kg^{-1} to 4 mg kg^{-1}. The mean concentration of PCB in whole breast milk in Canadian women steadily increased from 6 µg kg^{-1} in 1970 to 12 µg kg^{-1} in 1975 to 26 µg kg^{-1} in 1982, before declining to 6 µg kg^{-1} in 1986. Norén and Meironyté (referenced in (WHO, 2003) reported a steady decrease (from 910 to 324 ng g^{-1} lipid) of total PCB concentrations in the breast milk of Swedish women for the period 1967–1997. In Japan, breast milk samples were analysed for PCB content to determine their yearly trend; the average PCB concentration increased from 1972 (1.302 µg g^{-1} lipid basis) to its highest in 1974 (1.514 µg g^{-1} lipid basis), and then decreased to about 13% of that concentration (0.200 µg g^{-1} lipid basis) in 1998. Daily intake of PCB from breast milk was estimated to decrease from 22.3 µg g^{-1} d^{-1} to 0.31 µg g^{-1} d^{-1} during this period. [This trend reflects a change in concentrations of PCB in food consumed (both a decrease in contamination and more dependence on imported foods, which were less contaminated than domestic ones) (WHO, 2003)) and is consistent with a decline in the concentrations of PCB in both the environment and human tissues.

Germany

A declining trend in Germany can also be observed for the ~~indicator~~ PCB_6~~PCB~~ s in humans (Fürst, 2001). Fig 1 shows the mean values for the congeners PCB-138, PCB-153 and

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PCB-180 in more than 1800 individual milk samples from Germany collected and analysed between 1984 and 2000. Concentrations for these three compounds were almost equal in the 1980s, the results from samples analysed in the 1990s also showed a declining trend. Compared to 1984, the concentrations in samples from 2000 were approximately 60–70% lower. It was assumed that this decrease is the result of the ban of PCB usage in open systems and the strict regulations on the use of PCB in closed systems as well as on their disposal.

Asia

Time trends of TEQs (PCDD/PCDF, PCB) in human milk in Japan (MoE/MoH 1999) are shown below.

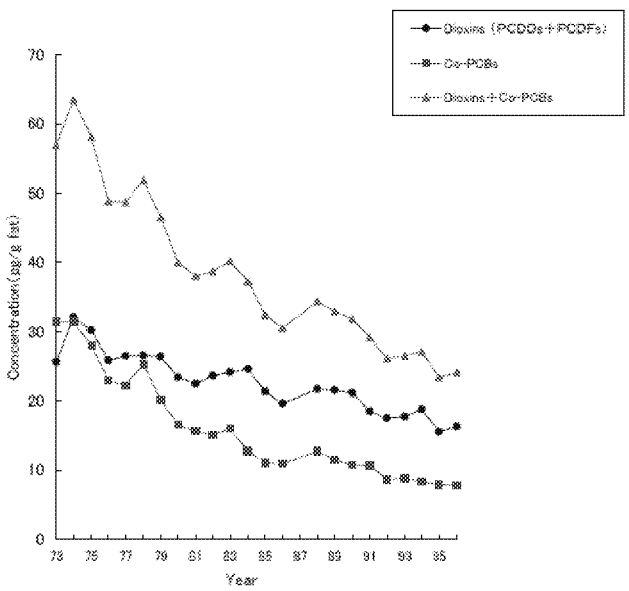

Fig.2 Dioxin Concentration in Breast Milk
Source: Health and Welfare Scientific Study Project in Fiscal Year 1997 Studies on Dioxins in Breast Milk[12] (Ministry of Health and Welfare)

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In China, a national investigation comprising 1237 individual human milk samples from 12 provinces in China was performed (Li *et al.* 2009). The population of the provinces accounts for approximately 50% of the total Chinese population. The range of upper-bound total-TEQ in samples was from 2.59 pg TEQ g^{-1} lipid to 9.92 pg TEQ g^{-1} lipid with a mean of 5.42 pg TEQ g^{-1} lipid and a median of 5.11 pg TEQ g^{-1} lipid. PCDD/PCDF-TEQ and total-TEQ in human milk from rural areas were lower than those from urban areas. The positive correlations were found between total-TEQ level in human milk and the consumption of aquatic food and meat respectively. The mean estimated daily intake of PCDD/PCDF and dl-PCBs by infants was 28.0 pg TEQ kg^{-1} bw per day with a range from 14.2 pg TEQ kg^{-1} bw per day to 48.6 pg TEQ kg^{-1} bw per day.

1.4.8 Estimated daily intakes

In Europe, more than 90% of the PCB exposure in the general population is via food (EFSA, 2005c). Average daily dietary intakes of total ~~ndl-PCB~~PCB6 can be estimated to be in the range of 10–45 ng kg^{-1} body weight (b.w.) *per* day. Limited exposure data for young children, up to six years of age, indicates that the average intake (breastfeeding excluded) of total ~~ndl-PCB~~PCB6 is about 27–50 ng kg^{-1} b.w. d^{-1}. However, where data on both adults and children within a specific population were available, in general children had exposure concentrations 2.5-fold higher than adults.

In the USA, the dietary intake of PCB for adults continually decreased from 1978 until 1986–1991. In 1978, the estimated dietary intake of PCB by adults in the USA was 0.027 µg/kg body weight per day, but it declined to 0.0005 µg/kg body weight per day in 1982.1984 and < 0.001 µg/kg body weight per day for the period 1986.1991 (WHO/CICAD, 2003). The mean daily intake was < 0.001 µg kg^{-1} body weight *per* day (ng kg^{-1} b.w. d^{-1}) for individuals of ages 6 months and 25 years–30 years and 0.002 µg kg^{-1} body weight *per* day for 2-year-olds between 1986 and 1991 (based on average total diet composition). In a study on dietary

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intake during 1991–1997, the decreasing trend did not continue, and the dietary exposure was 3 ng kg^{-1} b.w. d^{-1}–5 ng kg^{-1} b.w. d^{-1} for adults and 2 ng kg^{-1} b.w. d^{-1}–12 ng kg^{-1} b.w. d^{-1} children of different ages (based on calculated intakes from 265 different food items) (WHO CICAD, 2003).

Dietary intake of PCB ~~levels~~ have been calculated. The concentrations of PCDD/PCDF and dl-PCBs in four food groups of animal origin were determined in 12 provinces in China. The concentration of PCDD/PCDF ranged from 0.02 to 0.28 pg TEQ g-1. Using food consumption data from a 3-day household dietary survey, the daily dietary intake of dioxin-like compounds from foods of animal origin in China was estimated to be in the range of 0.09 to 0.59 pg TEQ kg-1 body weight day-1 , which is lower than that in some developed countries. (Liu et al. 2013)

In specific subpopulations with high dietary PCB exposure such as Baltic Sea fishermen the daily intake from fish of the sum of the six ~~ndl-PCB~~PCB6 could be about 40 ng kg^{-1} b.w. d^{-1}, corresponding to an intake of total ~~ndl-PCB~~PCB6 of 80 ng kg^{-1} b.w. d^{-1} before taking into account the rest of the diet. Breastfed infants are a group of high ~~ndl-~~PCB_6~~PCB~~ intake which might be two orders of magnitude higher than adult exposure (Lindell *et al.*2012).

Commented [A5]: The ref provided for the Great Lakes does not provide usable PCB concentrations or characteristics.

Breastfed infants are a special group with high intake of ~~ndl-PCB~~PCB6. From the results of the most recent WHO exposure study, a mean intake of about 1600 ng kg^{-1} b.w. d^{-1} (range: 230 ng kg^{-1} b.w. d^{-1}-7300 ng kg^{-1} b.w. d^{-1}) can be calculated for total ~~ndl-PCB~~PCB6. Thus, the exposure of infants to ~~ndl-PCB~~PCB6 (and dl-PCB) through human milk is about two orders of magnitude higher than the average daily intake by adults.

Usually ~~ndl-PCB~~PCB6 from air (ambient, indoor), dust and soil contribute only a few percent to the body burden of ~~ndl-PCB~~PCB6. However, depending on the source of contamination, situations exist where a contribution from indoor air, particularly of the lower chlorinated congeners, could be considerable.

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For Europe, graphic representation of PCDD/PCDF and dl-PCB on TEQ-basis from the EFSA report 2012 (EFSA, 2012) is shown in Fig. S1, which illustrates the relative contribution of PCDD, PCDF, mono-*ortho* PCB and non-*ortho* PCB, expressed in WHO_{2005}-TEQ, to the lower and upper bound estimates of the total WHO_{2005}-TEQ of PCDD/PCDF and dl-PCB in food and feed. Overall, the main contributors were the non-*ortho* PCB, representing between 21.0% and 74.9% of the WHO_{2005}-TEQ_{total} of PCDD/PCDF and dl-PCB in food, followed by the PCDD and PCDF which together represented between 12.4% and 73.2% of the WHO_{2005}-TEQ_{total}. The mono-*ortho* PCBs did not represent more than 12% of the WHO_{2005}-TEQ_{total}. When looking at the most contaminated samples, the relative contribution of the non-*ortho* PCB increased, from 34.2% to 86.1% depending on the food group. The food groups with the highest concentrations of non-*ortho* PCBs (more than half the total contamination) were products from aquatic animals and from ruminants.

The contamination profile of the feed groups was quite similar to what was observed in food, except for four of the feed groups. As observed in the food groups, the most contaminated feed group samples had an increased relative contribution of non-*ortho* PCB to the WHO_{2005}-TEQ_{total}, up to two times higher than the average contribution.

Fig. S2 illustrates the average relative contribution of each individual ~~indicator~~ congener to the sum of the ~~six ndl-PCB~~PCB6 for the lower and upper bound estimates in food and feed.

References

<conf>Büchert A, Cederberg T, Dyke P *et al.* (2001). ESF Workshop on Dioxin Contamination in Food. ESPR – Environ. Sci. & Pollut. Res. **8**:84–88.</conf>

<eref>BVL (no year). Bundesamt für gesundheitlichenVerbraucherschutz, Germany; http://www.bvl.bund.de/SharedDocs/Downloads/01_Lebensmittel/unerwuenschteStoffe/4_Aus-wertung_Dioxin_PCB.html</eref>

<eref>Codex alimentarius (2006). *Code of Practice for the Prevention and Reduction of Dioxin and Dioxin-like PCB Contamination in Food and Feeds*. Available at: http://www.codexalimentarius.org/download/standards/10693/CXP_062e.pdf</eref>

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<eref>EFSA European Food Safety Authority (2005a). Opinion of the Scientific Panel on Contamination in the Food Chain on a Request from the Commission Related to the Presence of Non-dioxin-like Polychlorinated Biphenyls (PCB) in Feed and Food (Question N° EFSA-Q-2003–114), Adopted on 8 November 2005. The EFSA Journal (2005) 284, 6 pages. http://www.efsa.eu.int</eref>

<eref>EFSA European Food Safety Authority (2005b). Annex I to the Opinion on Non-dioxin-like Polychlorinated Biphenyls (PCB) in Feed and Food. The EFSA Journal (2005) 284, Annex I, 1 – 48. Available at: http://www.efsa.eu.int</eref>

<eref>EFSA European Food Safety Authority (2005c). Opinion of the Scientific Panel on Contamination in the Food Chain on a Request from the Commission Related to the Presence of Non-dioxin-like Polychlorinated Biphenyls (PCB) in Feed and Food (Question N° EFSA-Q-2003–114), Adopted on 8 November 2005. The EFSA Journal (2005) 284, 137 pages. Available at: http://www.efsa.europa.eu/en/efsajournal/doc/284.pdf</eref>

<eref>EFSA European Food Safety Authority (2008). Statement of EFSA on the risks for public health due to the presence of dioxins in pork from Ireland (Question No EFSA-Q-2008–777) Issued on 10 December 2008. *The EFSA Journal* (2008) 911, 1–15. Available at: http://www.efsa.europa.eu/cs/BlobServer/Statement/contam_ej_911_dioxins_rev2.pdf?ssbinary=true</eref>

<eref>EFSA European Food Safety Authority (2010). Results of the monitoring of dioxin levels in food and feed. EFSA Journal 2010; 8(3):1385 [36 pp.]. doi:10.2903/j.efsa.2010.1385. Available at: www.efsa.europa.eu</eref>

<eref>EFSA European Food Safety Authority (2012). Update of the monitoring of dioxins and PCBs levels in food and feed. EFSA Journal 2012; 10(7):2832. [82 pp.] doi:10.2903/j.efsa.2012.2832. Available at: www.efsa.europa.eu/efsajournal</eref>

<eref>EFSA CONTAM (2012). Scientific Opinion on the presence of dioxins (PCDD/Fs) and dioxin-like PCBs (DL-PCBs) in commercially available foods for infants and young children. EFSA Journal 2012;10(12):2983 [28 pp.]. doi:10.2903/j.efsa.2012.2983. Available at: http://www.efsa.europa.eu/en/efsajournal/pub/2983.htm</eref>

<eref>EU SANCO – D1 (2008) D/412398. Summary Minutes of the Standing Committee on the Food Chain and Animal Health, Brussels, 10 December 2008. Available at: http://ec.europa.eu/food/committees/regulatory/scfcah/toxic/summary10122008_en.pdf</eref>

<eref>European Commission (no year). Health and Consumers – Food, Available at: http://ec.europa.eu/food/food/chemicalsafety/contaminants/dioxins_en.htm</eref>

<eref>HBM (no year).Available at: http://www.eu-humanbiomonitoring.org/</eref>

<jrn>Huwe JK (2010). New and Old POPs (PCDD/Fs, PCBs, and PBDEs) in U.S. Foods Levels, Trends, and Implications for Human Exposure. *Organohalogen Compd*. 72:108–111.</jrn>

Formatted: Superscript

<jrn>IARC (1978). Polychlorinated biphenyls and polybrominated biphenyls. *IARC Monogr Eval Carcinog Risk Chem Hum*, 18:1–124. PMID:215509</jrn>

<jrn>IARC (1997). Polychlorinated dibenzo-para-dioxins and polychlorinated dibenzofurans. *IARC Monogr Eval Carcinog Risks Hum*, 69:1–631. PMID:9379504</jrn>

<eref>IPCS (1993). Polychlorinated biphenyls and terphenyls, 2nd ed. Geneva, World Health Organization, International Programme on Chemical Safety (Environmental Health Criteria 140). Available at: http://www.inchem.org/documents/ehc/ehc/ehc140.htm</eref>

<eref>Lindell B (2012). 146. Polychlorinated biphenyls (PCBs). The Nordic Expert Group for Criteria Documentation of Health Risks from Chemicals, nr 2012; 46(1). University of Gothenburg, Sweden, ISBN 978–91–85971–35–0, ISSN 0346–7821. Available at: www.nordicexpertgroup.org</eref>

<other>Malisch R, Kotz A, Tritscher A *et al.* (2013). WHO/UNEP-Coordinated Global Surveys on Concentrations of Persistent Organic Pollutants in Human Milk – Part 3.1: Levels of PCDD, PCDF, dioxin-like PCBs and non dioxin-like PCBs. (Manuscript in preparation)</other>

<other>NATO/CCMS (1988). International Toxicity Equivalency Factor (I-TEF) Method of Risk Assessment for Complex Mixtures of Dioxins and Related Compounds. Pilot Study on International Information Exchange on Dioxins and Related Compounds, Report Number 176, August 1988, North Atlantic Treaty Organization, Committee on Challenges of Modern Society.</other>

<edb>Päpke O, Fürst P (2003). Background Contamination of Humans with Dioxins, Dioxin-Like PCBs and Other POPs. Chapter 10 in: The Handbook of Environmental Chemistry Vol. 3, Part O, Persistent Organic Pollutants. Fiedler H, editor. Berlin Heidelberg: Springer-Verlag, pp. 271–295.</edb>

<eref>SCOOP (2000). Scientific co-operation on questions relating to food "Assessment of dietary intake of dioxins and related PCBs by the population of EU Member States" Task 3.2.5 – Final Report – 7 June 2000, pp. 141. http://ec.europa.eu/food/fs/sc/scf/out78_en.pdf</eref>

<eref>UNEP (2001). Text of the Stockholm Convention on Persistent Organic Pollutants and implementation activities; amended 2009. Available at: www.pops.int</eref>

<jrn>Van den Berg M, Birnbaum L, Bosveld AT *et al.* (1998). Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. *Environ Health Perspect*, 106:775–792. http://dx.doi.org/10.1289/ehp.98106775 PMID:9831538</jrn>

<jrn>Van den Berg M, Birnbaum LS, Denison M *et al.* (2006). The 2005 World Health Organization reevaluation of human and Mammalian toxic equivalency factors for dioxins and dioxin-like compounds. *Toxicol. Sci.* 93:223–241. http://dx.doi.org/10.1093/toxsci/kfl055 PMID:16829543</jrn>

Formatted: Superscript

<jrn>van Larebeke N, Hens L, Schepens P *et al.* (2001). The Belgian PCB and dioxin incident of January-June 1999: exposure data and potential impact on health. *Environ Health Perspect*, 109:265–273. http://dx.doi.org/10.1289/ehp.01109265 PMID:11333188</jrn>

<eref>WHO (2003). Polychlorinated biphenyls: Human heath aspects. Concise International Chemical Assessment Document 55. Published under the joint sponsorship of the United Nations Environment Programme, the International Labour Organization, and the World Health Organization, and produced within the framework of the Inter-Organization Programme for the Sound Management of Chemicals (IOMC), Geneva, 2003. Available at: http://www.inchem.org/documents/cicads/cicads/cicad55.htm#6.2</eref>

<eref>WHO CICAD (2003). Concise International Chemical Assessment Document 55 Polychlorinated Biphenyls: Human Health Aspects. World Health Organisation CICAD Nr. 55. Available at: whqlibdoc.who.int/publications/2003/9241530553.pdf</eref>

G. Liu, M. Zheng, Z. Cai; Y. Wu and J. Jiang (2013): Dioxin Analysis in China. Trends in Analytical Chemistry (2013), in press

Jingguang Li , Lei Zhang, Yongning Wu, Yinping Liu, Pingping Zhou, Sheng Wen, Jiaying Liu, Yunfeng Zhao, Xiaowei Li (2009): A national survey of polychlorinated dioxins, furans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs) in human milk in China. Chemosphere 75, 1236–1242

J.L. Domingo and A. Bocio (2007): Levels of PCDD/PCDFs and PCBs in edible marine species and human intake: A literature review. Environment International 33,397–405

Yin-ping Liu, Jing-guang Li, Yun-feng Zhao, Sheng Wen, Fei-fei Huang, Yong-ning Wu (2011) Polybrominated diphenyl ethers (PBDEs) and indicator polychlorinated

biphenyls (PCBs) in marine fish from four areas of China. Chemosphere 83, 168–174

Yim UH, Hong SH, Shim WJ, Oh JR. Levels of persistent organochlorine contaminants in fish from Korea and their potential health risk. Arch Environ Contam Toxicol 2005;48:358–66

Moon HB, Ok G. Dietary intake of PCDDs, PCDFs and dioxin-like PCBs, due to the consumption of various marine organisms from Korea. Chemosphere 2006;62:1142–52.

Loufty N, Fuerhacker M, Tundo P, Raccanelli S, El Dien AG, Tawfic Ahmed M. (2006): Dietary intake of dioxins and dioxin-like PCBs, due to the consumption of dairy products, fish/seafood and meat from Ismailia city, Egypt. Sci Total Environ 2006;370:1–8.

Loufty N, Fuerhacker M, Tundo P, Raccanelli S, Tawfic Ahmed M. Monitoring of polychlorinated dibenzo-p-dioxins and dibenzofurans, dioxin-like PCBs and polycyclic aromatic hydrocarbons in food and feed samples from Ismailia city, Egypt. Chemosphere 2007;66:1962–70.

Zhao X, Zheng M, Liang L, Zhang Q, Wang Y, Jiang G. Assessment of PCBs and PCDD/Fs along the Chinese Bohai Sea coastline using mollusks as bioindicators. Arch Environ Contam Toxicol 2005;49:178–85.

Formatted: Superscript

Rawn DFK, Forsyth DS, Ryan JJ, Breakell K, Verigin V, Nicolidakis H, et al. PCB, PCDD and PCDF residues in fin and non-fin fish products from the Canadian retail market 2002. Sci Total Environ 2006;359:101–10

Hites RA, Foran JA, Carpenter DO, Hamilton MC, Knuth BA, Schwager SJ. Global assessment of organic contaminants in farmed salmon. Science 2004a;303:226–9.

Hites RA, Foran JA, Schwager SJ, Knuth BA, Hamilton MC, Carpenter DO. Global assessment of polybrominated diphenyl ethers in farmed and wild salmon. Environ Sci Technol 2004b;38:4945–9.

RA Hites, JA Foran, et al. (2004c): Global assessment of organic contaminants in farmed salmon. Science 303, 226-229

Arnold Schecter, Justin Colacino, Darrah Haffner, Keyur Patel, Matthias Opel, Olaf Päpke, and Linda Birnbaum (2010): Perfluorinated Compounds, Polychlorinated Biphenyls, and Organochlorine Pesticide Contamination in Composite Food Samples from Dallas, Texas, USA. Environmental Health Perspectives 108, 976 ff

UNEP 2012a mothers' milk
http://www.chem.unep.ch/Pops/GMP/Mothers%20milk%20guide%20POPs.pdf
UNEP 2012b : mothers' milk
http://www.chem.unep.ch/Pops/GMP/Mothers%20milk%20guide%20POPs_Annexes.pdf

Fürst P (2001) Organochlorine pesticides, dioxins, PCB and polybrominated biphenyl ethers in human milk from Germany in the course of time. Organohalogen Compounds 52:185–188

Douglas Hayward, Jon Wong, Alexander J. Krynitsky (2007): Polybrominated diphenyl ethers and polychlorinated biphenyls in commercially wild caught and farm-raised fish fillets in the United States. Environmental Research 103, 46-54

AMAP, 2004. AMAP Assessment 2002. Persistent Organic Pollutants in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

AMAP, 2009. AMAP Assessment 2009. Human Health in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

Berti, P.R.; Receveur, O.; Chan, H.M.; Kuhnlein, H.V. (1998). Dietary exposure to chemical contaminants from traditional food among adult Dene/Métis in the Western Northwest Territories, Canada. Environ. Res. 76, 131-142.

Dewailly, E.; Ayotte, P.; Bruneau, S.; Laliberte, C.; Muir, D.C.G.; Norstrom, R.J. (1993). Inuit exposure to organochlorines through the aquatic food chain in Arctic Quebec. Environ. Health Persp. 101 (7), 618-620.

Johansen, P.; Muir, D.; Asmund, G.; Riget, F. (2004). Human exposure to contaminants in the traditional Greenland diet. Sci. Total Environ. 331, 189-206.

Rigét, F.; Bignert, A.; Braune, B.; Stow, J.; Wilson, S. (2010). Temporal trends of legacy POPs in Arctic biota, an update. Sci. Total Environ. 408, 2874-2884.

Formatted: Superscript

Food Standards Australia New Zealand (2004) Dioxins in Food: Dietary Exposure Assessment and Risk Characterization. Technical Report Series No 27

Padula DJ, Madigan TL, Nowak BF, 2012: Australian farmed Yellowtail Kingfish (Seriola lalandi) and Mulloway (Argyrosomus hololepidotus): residues of metallic, agricultural and veterinary chemicals, dioxins and polychlorinated biphenyls. Chemosphere 86:709-717. doi: 10.1016/j.chemosphere.2011.10.044

Carrie Tomasallo, Henry Anderson, Mary Lee Haughwout, Pamela Imm, Lynda Knobeloch (2012): Mortality among frequent consumers of Great Lakes sportfish. Environmental Research 110, 62–69