

1. Introduction

Mercury (Hg) is a ubiquitous, persistent and lipophilic contaminant detected worldwide. It is slowly eliminated in vertebrate species, including humans. Natural biomethylation reactions of Hg in freshwater and marine ecosystems results in formation of the organic and most toxic form, methylmercury (MeHg). MeHg is readily taken up by organisms and biomagnifies in the food web (Björnberg et al. 2005), and can thus make up 95% of the total Hg in fish and fishery products (Bloom 1992, Downs et al. 1998, Freije & Awadh 2009, Kannan et al. 1998). Generally, fish is the main source of MeHg exposure in humans (EC 2008, Freire et al. 2010, JECFA 2003).

The major human health effect from exposure to MeHg is neurotoxicity. MeHg readily crosses the blood-brain barrier. Nonspecific cell injury is mediated through binding of MeHg to cytosolic proteins. This seriously harms normal neuronal development and can lead to altered brain architectures and decreased brain size; extreme exposures even leads to coma and death as seen in 1956 in Minamata, Japan (Harada 1972). A mass outbreak of MeHg poisoning in Iraq illustrated that maternal hair mercury concentrations of 10-20 $\mu\text{g/g}$ during pregnancy were associated with the first MeHg-caused symptoms in children, demonstrating the susceptibility of the developing central nervous system; similar effects are seen in adults only with concentrations five times higher (McElhatton 2000, WHO 1990). Exposures of this magnitude are not achievable by consuming fish with current natural concentrations but only through contamination incidents.

Infants and children are particularly vulnerable to mercury intoxication, and should therefore be treated as a separate subgroup when assessing intake of persistent organic pollutants. There are two clear reasons for children-specific intake assessments of environmental contaminants:

- 1) food consumption per weight is higher in young children due to rapid growth and

development; and 2) children are more vulnerable to development-disrupting effects of contaminants due to their still maturing physiology. Although improvements have been made during the past decade in Finland in general population estimates of contaminant intake, it has been widely agreed that more effort should be invested in studying sensitive subgroups and environmental health risk assessment of contaminants. These are demanding tasks that require multidisciplinary approaches, but should be performed to improve risk management and to mitigate food-mediated exposure to contaminants. With children-specific intake assessments these tasks can be adequately fulfilled.

This study was designed to specifically assess the long-term mean daily MeHg intake in susceptible gender-specific age groups of children from 1 to 6 years. We had no previous information whether the intakes could differ between genders. Calculated intakes are compared to international recommendations. Databases for fish consumption and fish concentration were used to estimate the intakes in 1, 3 and 6-year-old Finnish children. Only approximately one third of fish consumed in Finland is of domestic origin, which justifies inclusion of imported fish (RKTL 2008). An effective intake and risk assessment (and management) of contaminants in children should not only pay attention to the long-term mean daily intakes but particularly be able to tackle the highly exposed population subgroups. Presenting fish consumption data as probability distributions allows us to examine the probability range of the long-term mean daily intakes of MeHg from fish, and to determine the portion in the examined age groups which exceeds the provisional tolerable weekly intake (PTWI) by FAO/WHO Joint Expert Committee on Food additives and Contaminants (JECFA 2006) and the tolerable daily intake set by the U.S. Environmental Protection Agency (U.S.EPA 2001).

This study brings new information for assessing health risk of the most sensitive subpopulation of mercury mediated health effects. Due to the country-specific fish

consumption pattern and geographical variation of MeHg found in fish, the scope is local but physiologically the results apply to children globally.

2. Materials and methods

2.1 Study subjects

Our study was possible by using as subjects the participants in a population-based cohort study called the Finnish Type 1 Diabetes Prediction and Prevention Study (DIPP) (Kupila et al. 2001, Kyttälä et al. 2010, Virtanen et al. 2006) for which food consumption data of different children with varying ages was available. Newborn infants were recruited from two urban areas in Finland, Oulu and Tampere. Human leukocyte antigen (HLA) -conferred genetic susceptibility to type 1 diabetes was screened from cord blood and children with HLA genotypes conferring increased risk for type 1 diabetes were invited to take part in the DIPP study (15% of the population screened). The series comprises at-risk children born in 1997-2004. The diets of the subjects were followed at 3 to 12 month intervals by food records and questionnaires. A background questionnaire and structured dietary questionnaires with 3-day food records were collected at ages 1, 3 and 6. Food records were available from groups of 1-year-olds (n=963), 3-year-olds (n=1045) and 6-year-olds children (n=850). All food records had been kept during the years 2003-2005. The study was approved by the local Ethics Committees, and all the families gave their written informed consent at the beginning of the study.

2.2 Fish consumption estimations

The fish consumption by the children was recorded by the parents and day care personnel using 3-day food records that included 3 consecutive days, 2 weekdays and 1 weekend day (Kyttälä et al. 2010). The families and the day care personnel received written instructions to

record with household measures, the type, brand and preparation method of all the food taken by the child. A trained study nurse reviewed the records item by item for completeness and accuracy, and added information when needed. Food consumption was converted to ingredient level using in-house software and the Finnish Food Composition Databank FINELI[®] (2010). Children for whom the food consumption had been recorded for less than 3 days (n=4.9%) were excluded from the analysis.

2.3 Fish mercury concentrations

The majority of Hg concentration data for Finnish fish originated from a specific research project on Finnish fish, “EU-fish”, in which 135 fish muscle samples without skins were analyzed for Hg using a previously established method (Louie et al. 1985, Venäläinen et al. 2004). The following method used in this study was accredited according to standard EN ISO/IEC 17025 by Finnish Accreditation Service FINAS. The homogenized sample, about 1 g is weighed into an Erlenmeyer and 1 ml of 10 M HCl and 5 ml of 14 M HNO₃ is added and mixed carefully. Then 2 ml of 18 M H₂SO₄ is added. The mixture is left to stand covered with watch glass until the reaction in the vessel is over. Then the mixture is left to stand in a water bath for 1.5 hours. The mixture is cooled down to room temperature and 10 ml of water and 20 ml of hexane is added. The mixture is shaken and let to stand at room temperature for 2 hours until all fat is extracted to hexane. Hexane is extracted away, erlenmeyer is rinsed with water, 25 µl of 5 % KMnO₄ is added and solution is adjusted to 50 ml with water. The Hg concentrations were measured by Atomic Absorption Spectrometry (AAS) using cold vapor technique (Perkin-Elmer 4100+FIAS 200+AS 90 autosampler+ EDL Power Supply). Sample and standard solutions (Reagecon) were treated with 5 % KMnO₄ so that Hg is present in ionic form in the acidic solution. NaBH₄ is used as the reducing agent. Reductant is dispensed into the sample solution by FIAS (flow injection analysis system) where it reacts to liberate hydrogen. Since Hg has an appreciable volatility even at ambient temperature,

metallic Hg vapor is driven out of the sample solution by the hydrogen and transported to the quartz cell where its absorption is measured. The cell is heated to around 100° C to prevent condensation. The measurement was carried out with direct comparison with standard solution in 0.1 M HNO₃ at a wavelength of 253.7 nm.

Certified reference materials (BCR 184 bovine muscle and BCT 422 Cod muscle) were analyzed together with each sample series to check the accuracy of the method. The values corresponded very well to the certified values. The results of certified reference materials were monitored on the control charts. The recovery was studied by adding known amounts of standard solution to samples. The amounts added were selected so that they would be close to the amounts normally found. Hg recoveries in different fish species were 80- 110%. Blank samples were also analyzed with each sample series to check for contamination. Repeatability (CV% =6 %) was tested with replicate analysis of samples. Limit of qualification (0.005 mg/kg) was determined by analyzing the blank samples.

The dataset covered the year 2002, and included ten Finnish fish species of which seven were sampled from inland habitats (number of samples per species: 4-8) and eight from the Baltic Sea (number of samples per species: 1-35): burbot (*Lota lota*), bream (*Abramis brama*), pike-perch (*Sander lucioperca*), perch (*Perca fluviatilis*), pike (*Esox lucius*), vendace (*Coregonus albula*), whitefish (*Coregonus lavaretus*), sprat (*Sprattus sprattus*), Baltic Sea salmon (*Salmo salar*), and Baltic herring (*Clupea harengus membras*).

Hg concentrations in farmed salmon (rainbow trout, *Oncorhynchus mykiss*), imported fish species such as Atlantic salmon (*Salmo salar*), , tuna (Scombrinae) as well as several Gadidae species consumed substantially by the Finnish population were obtained from several other sources (LeBlanc et al. 2006, NIFES web page <http://www.nifes.no/>, Ásmundsdóttir et al.

2005). Differences in MeHg concentrations between fish species were computed by the nonparametric Kruskal-Wallis test, followed by stepwise comparisons (PASW Statistics 18.0). Baltic Sea whitefish included only 1 measurement and it was excluded from the analysis. A p-value of <0.05 was considered significant.

2.4 Methylmercury intake calculations

Long-term mean daily MeHg intakes were calculated for 1-, 3- and 6-year-old children separately for both sexes using the calculated mean daily consumption of the fish containing food items multiplied by the corresponding mean values of MeHg concentrations. Intakes were summed over each study day for all the subjects. In order to remove day-to-day variation and to adjust for bias effects (e.g. day of the week, interview sequence), we used the method of Nusser et al. (1996) to obtain long-term average daily intakes, or usual intakes. These intake distributions were estimated using C-SIDE[®] (version 1.0; Department of Statistics, Center for Agricultural and Rural Development -CARD, Iowa State University, Ames), which implements the Nusser method. Because no individual child weights were available at the time of preparing this paper, we used age- and gender-specific proportional body weights (i.e. mean body weights from age-specific mean-heights) from Finnish child welfare clinics as specified in 1993 to adjust the long-term mean daily intakes to body weights. The used body weights as kg were: 9.9, 14.6, 20.8 and 10.5, 15.0, 21.2 for 1-, 3- and 6-year-old girls and boys, respectively. Fish Hg concentrations were analyzed using both the upper bound and the lower bound concentrations. These were calculated on the assumption that all the values of the individual measurements below the limit of quantification are equal to the limit of quantification or to zero, respectively. The difference between these two quantification methods in the long-term mean daily intakes was negligible (0.9999 ± 0.0001 , mean \pm SD) and we present only the lower bound intakes in this paper. Because more up to 95% of Hg present

in fish is in the form of MeHg (Downs et al. 1998, Freije & Awadh 2008, Kannan et al. 1998, Westöö 1973), total Hg analyzed from both domestic and imported fish was converted to MeHg using a conversion factor of 0.93 (Westöö 1973).

2.5 Comparison to tolerable intakes

The assessed long-term mean daily intake of MeHg was compared with the provisional tolerable weekly intake (PTWI) of 1.6 µg/kg bw set by the FAO/WHO Expert Committee on Food and Contaminants (JECFA, 2003) and with the commonly used reference dose (RfD) of 0.1 µg/kg bw/day set by the U.S.EPA (U.S.EPA 1997, U.S.EPA 2001).

3. Results

3.1 Fish consumption

In the 1-year-old children, rainbow trout (farmed salmon) was clearly the most commonly consumed species followed by Atlantic salmon and Gadiformes (Table 1) although as quantities, usage of pike-perch and other species was significant (Figure 1). Children aged 1 and 3 years showed similar trends in species consumption patterns although the used quantities almost doubled from 1 to 3 years. In the 6-year-old children rainbow trout, Atlantic salmon, Gadiformes and tuna were the species consumed most (Figure 1). Girls in this age group favoured Gadiformes, whitefish and tuna, whereas boys consumed remarkably less whitefish. The consumption of burbot, bream, Baltic Sea salmon and sprat was minor and those species are not shown in Fig 1.

Table 1. Percentage of users of a particular fish species in different age groups. Results are based on 3-d food record periods in Finnish 1 to 6-year-old children. All species = percentage of fish users of the study population.

Figure 1. Fish consumption statistics (min, 1st quartile, median, 3rd quartile, max) in Finnish 1 (A), 3 (B) and 6-year-old (C) children estimated from the 3-d food record study among those who use fish. 7.5% of boys and 27.5% of girls had no fish by the age of 1 year.

3.2 Methylmercury concentration in fish

Hg was detected in all analyzed fish species. Three out of six sprat individuals, and two out of 35 Baltic herring individuals showed undetectable concentrations. The mean (\pm standard deviation) FW (freshwater) and BS (Baltic Sea) MeHg-concentrated concentrations were: FW pike: 0.353 (\pm 0.108); BS pike: 0.370 (\pm 0.223); FW perch: 0.128 (\pm 0.077); BS perch: 0.346 (\pm 0.411); FW pike-perch: 0.276 (\pm 0.066); BS pike-perch: 0.102 (\pm 0.057); FW burbot: 0.205 (\pm 0.096); BS burbot: 0.238 (\pm 0.075); FW vendace: 0.078 (\pm 0.040); FW whitefish: 0.075 (\pm 0.013); BS whitefish: 0.017; Baltic Sea salmon: 0.063 (\pm 0.015); FW bream: 0.057 (\pm 0.019); Baltic herring: 0.027 (\pm 0.019); and BS sprat: 0.011 (0.013). MeHg concentration in fish was strongly variable and related to certain fish species (Figure 2; Kruskal-Wallis, $0.001 < p < 0.05$). The highest concentrations were measured in lean predatory fish species, pike and perch. Predatory freshwater pike-perch and burbot also showed high MeHg concentrations. Nonparametric stepwise comparisons revealed homogenous subsets of pike and burbot in both habitats, freshwater pike-perch and Baltic Sea perch (Kruskal-Wallis, $p < 0.05$). A single high concentration measurement (1.26 mg/kg) of the Baltic perch explains the high variation. There were no clear differences between freshwater and Baltic Sea habitats in these species. Concentrations in Baltic perch were double those in freshwater, but

freshwater pike-perch had more than double the concentration of those in the Baltic Sea.

Other homogenous subsets were bream, vendace, whitefish and perch in freshwater habitats together with salmon and pike-perch in the Baltic (Kruskall-Wallis, $p < 0.05$), and also Baltic Sea sprat (only one sample, thus no test statistics computed) and Baltic herring (one sample).

Figure 2. MeHg (mg/kg fresh weight) in various Finnish fish species (muscle without skin) from freshwater (FW) and the brackish Baltic Sea (BS) environments. Vertical bars indicate standard error and dots min and max values.

3.3 Methylmercury intakes

In 1-, 3- and 6-year-old boys the long-term mean daily MeHg intake estimates from fish ranged between 0-0.125, 0.004-0.197 and 0.003-0.262 $\mu\text{g}/\text{kg}$ bw, respectively (Table 2). In the girls aged 1, 3 and 6 years the minimum and maximum long-term mean intake levels were 0-0.170, 0.003-0.142 and 0.004-0.331 $\mu\text{g}/\text{kg}$ bw/d, respectively (Table 2). The long-term median daily intake of MeHg for boys aged 1 year was 0.020 (95% CI: 0-0.076), for boys aged 3 years 0.031 (95% CI: 0.008-0.112), and for boys aged 6 years 0.045 (95% CI: 0.007-0.164) $\mu\text{g}/\text{kg}$ bw. The corresponding intakes for the girls were 0.020 (95% CI: 0-0.097), 0.022 (95% CI: 0.005-0.081) and 0.044 (95% CI: 0.008-0.183) $\mu\text{g}/\text{kg}$ bw, respectively. A systematic increasing trend in the long-term mean daily intake of MeHg corrected for body weight with age was seen for boys but not as clearly for girls (Figure 3). Those girls in our study population who ate fish at the age of 1 seemed to have higher long-term mean daily MeHg intakes than the 3-year-old girls (Figure 3).

Table 2. Gender-specific long-term mean daily MeHg intake statistic parameters ($\mu\text{g}/\text{kg}$ bw/d) in Finnish 1, 3 and 6 year-old children estimated from the 3-d food record study. Probability

to exceed RfD and PTWI stands for probability of exceeding the current U.S.EPA reference dose of 0.1 $\mu\text{g}/\text{kg bw}/\text{d}$ and JECFA provisional tolerable weekly intake of 1.6 $\mu\text{g}/\text{kg bw}$ ($=0.229 \mu\text{g}/\text{kg bw}/\text{d}$), respectively. SD = standard deviation.

Figure 3. The cumulative probability density of gender-specific long-term mean daily MeHg intake ($\mu\text{g}/\text{kg bw}/\text{d}$) in Finnish 1, 3 and 6 year-old boys (A) and girls (B) based on a 3-d food record study. U.S.EPA RfD for MeHg is 0.10 $\mu\text{g}/\text{kg bw}/\text{d}$ and JECFA PTWI 0.23 $\mu\text{g}/\text{kg bw}/\text{d}$ (see text for further information).

3.4 Comparison to tolerable intakes

One percent of boys aged 1 year, 5% in the 3-year-old boys and 15% in the 6-year-old boys exceeded the U.S.EPA reference dose (Table 2, Figure 3). The corresponding exceeding in the girls were 2.5%, 2.5% and 15%. One percent of the 6-year-old boys and 2.5% of the girls at the same age exceeded the JECFA provisional tolerable weekly intake of MeHg (Table 2, Figure 3).

4. Discussion

4.1 Methylmercury in fish and fish consumption

MeHg concentrations were approximately ten times higher in pike, perch, pike-perch and burbot than in those with the lowest concentrations. Despite the lower consumption rate of pike, pike together with rainbow trout and tuna appear to be the biggest source of MeHg from the fish species consumed in Finland, these three species each responsible for 20% of the total

MeHg exposure in Finland (Leino et al 2011). In addition, there is a clear correlation between the size of the fish and MeHg concentration (Venäläinen et al. 2004) thus potential MeHg risks are typically due to very specific fish consumption habits.

Finnish freshwater fish species have been reported to contain higher MeHg concentrations than marine fish (Oulun kaupunki 2004, Nakari et al. 2009), but the results presented in this paper show no clear evidence for that. Nevertheless, this might be the case in freshwater areas with acidic highly organic soils, such as newly built reservoirs on swampy soil. These water areas are under special surveillance by the local environmental agencies (Nuotio, 2008).

MeHg concentrations were similar in freshwater whitefish, but lower in freshwater pike and perch with those measured in a small Finnish lake more than a decade ago (Rask and Verta, 1995). Although the MeHg concentration data in fish in this study was quantitatively scarce (n=1-8; except for Baltic herring, n=35), we assume the data to be representative. Indeed MeHg concentrations in vendace, freshwater pike and perch, pike caught from sea regions and Baltic herring in a follow-up study by Nakari et al. (2009) covering years 2006-2007 were similar to the corresponding concentrations in our study.

4.2 Methylmercury intakes

Due to higher food consumption in relation to body weight, children ingest higher doses of environmental contaminants than adults. Because of rapid growth and development, exposure may vary significantly between children and adults but also between children in different age groups. Our results showed that generally the long-term mean daily intake of MeHg per kg body weight was at its highest in 6-year-old boys and girls and lower in younger age groups. The long-term mean daily MeHg intake in the children was also higher than that estimated for the general population in Finland (Evira 2010).

The data showed a slightly increasing trend in the estimated long-term mean daily MeHg intake with age up to 6 years. No such trend was seen in the MeHg intakes from food in Swedish children aged 4, 8-9 and 11-12 years (Concha et al. 2006). From that report it appears that the mean daily body weight adjusted MeHg intakes of 0.036 μg in German children aged 1 to 6 years and of 0.026-0.034 μg in Swedish 4-12 years-old boys and girls were similar to the means between 0.028-0.032 μg observed in 1-3 year-old Finnish girls and 1-year-old Finnish boys, but lower than the means between 0.045-0.068 μg in 3-year-old boys and 6-year-old boys and girls in our study (Table 2). Concha et al. (2006) reported the 95 percentile to be 0.11 and 0.05-0.09 $\mu\text{g}/\text{kg}$ bw/d for German and Swedish children, respectively, whereas in our study they were 0.076, 0.112 and 0.164 $\mu\text{g}/\text{kg}$ bw/d in 1, 3 and 6-year-old boys, and 0.097, 0.081 and 0.183 $\mu\text{g}/\text{kg}$ bw/d in 1, 3 and 6-year-old girls. The slight differences in the MeHg intake estimates between these children can be due to generally slightly higher MeHg concentrations in some Finnish species compared to those in Concha et al. (2006) covering years 2000-2002, consumption of species with high MeHg content as well as different calculation methods between the studies to assess the usual long-term intakes. No conversion from total Hg to MeHg was made in the study of Concha et al. (2006). Ortega-García et al. (2009) found much higher MeHg intake estimations of 0.37 (95% CI: 0.30-0.44) and 0.38 (95% CI: 0.32-0.43) $\mu\text{g}/\text{kg}$ bw/d from fish and shellfish consumption in 1-5 and 6-10-year-old children from the Murcia region, a Mediterranean part of Spain. The main exposure in their study was related to bluefin tuna and swordfish, which are species that contain high concentrations of MeHg and to which Finnish children are not generally exposed. In addition it seems that the variability in the intakes in their study population was much lower than in ours. Average weekly body weight-adjusted MeHg intakes of 1.96 $\mu\text{g}/\text{kg}$ (=0.28 $\mu\text{g}/\text{kg}$ bw/d) from fish and seafood by Spanish children from the Catalonia region

(Martí-Cid et al. 2007), and of 2.37 $\mu\text{g}/\text{kg}$ ($=0.34 \mu\text{g}/\text{kg}$ bw/d) from fish and meat among Inuit preschool children in Nunavut, Canada (Tian et al. 2011) results in tenfold higher intakes when compared to the children in the current study.

4.3 Comparison to tolerable values

Because fish is clearly the only significant source of MeHg from food for most populations (excluding some rare cases; JECFA 2003), MeHg intake can be diminished by reducing consumption of those species most contaminated with MeHg, such as pike, perch, pike-perch and burbot (its biological half life is 45-70 d).

To assess a possible health risk, MeHg intakes were compared to the RfD of the USEPA and the provisional tolerable weekly intake (PTWI) of the JECFA. The RfD by the U.S.EPA (1997) of 0.1 $\mu\text{g}/\text{kg}$ /bw uses MeHg-induced neurotoxicity (fetal and postnatal periods) over lifetime as a sensitive endpoint. A safety factor of up to ten at its highest, one by one, has been incorporated in the RfD to account for the following uncertainties: 1) extrapolation from animal to human data; 2) extrapolation of data to sensitive subpopulations; 3) lack of chronic data and use of a LOAEL (lowest-observed-adverse-effect-level) in the absence of NOAEL (no-observed-adverse-effect-level); and 4) lack of some other critical (e.g. toxicokinetic) data (NRC 2000; Rice 2004). The provisional tolerable weekly intake (PTWI) of 1.6 $\mu\text{g}/\text{kg}$ bw, proposed by the JECFA, is based on the most sensitive toxicological end-point, developmental neurotoxicity of children, in the most susceptible species (humans). Despite recognizing that adults may be less sensitive to the adverse effects of MeHg, the Committee could not identify a level of intake higher for adults than the existing PTWI for children.

Leino et al. (2011) analyzed MeHg intake of maternal fish consumption during pregnancy in relation to net health effects of the child by Monte Carlo analysis, using intelligence quotient as a composite index. Results showed net health effects varying between slightly beneficial to slightly adverse, according to type of fish consumed (fatty fish or lean fish, respectively).

Therefore, in our opinion the current long-term mean daily MeHg intake in children does not pose appreciable risk to the cognitive development in Finnish children. Indeed, Clewell et al. (1999) and Stern (1997) have proposed a MeHg RfD of 0.1-0.3 $\mu\text{g}/\text{kg}$ bw/d when only the uncertainties in the dose conversion factor from hair mercury concentration to chronic MeHg ingestion rate (this conversion was used in the analysis by Leino et al. 2011) were analyzed using Monte Carlo analyses. An observational cohort study in the USA by Hibbeln et al. (2007) also indicates that limiting maternal seafood consumption during pregnancy to reduce MeHg risk to the child could actually be detrimental to optimal fetal neurodevelopment. However, just as the U.S.EPA (1997) itself states “the risk following exposures above the RfD is uncertain, but risk increases as exposures to MeHg increase”, the MeHg intake in Finnish children surely needs to be carefully monitored and should be diminished.

Particularly, as it appears from our results that MeHg intake increases with age at least from 1 to 6 years although it will decrease again at later ages. According to our results the long-term mean daily MeHg intakes exceeded the U.S.EPA RfD 1.3-3.3 times at their highest. For the Finnish adult population with a fully developed nervous system MeHg risks at current intake level are negligible.

We lack means to achieve the long-term goals for the intakes other than 1) a total ban on fishing and usage of the most MeHg-contaminated fish species such as pike, perch, pike-perch and burbot as food sources, and 2) fish consumption recommendations. Because of the many beneficial health effects of fish consumption (Mozaffarian and Rimm 2006, Tuomisto et al. 2004, Tuomisto and Pekkanen 2005), the Finnish authorities (Evira; National Institute for

Health and Welfare, THL) have concluded to turn the balance in favour of recommending fish consumption. However, specific recommendations were given that 1) pregnant and breastfeeding women should not consume pike at all, 2) children, adolescents and people of fertile age could eat sea or lake caught pike only 1-2 times a month and 3) people eating inland fish daily should reduce their consumption of MeHg-accumulating fish such as perch, pike-perch and burbot. These recommendations, together with the general instruction to eat fish at least twice a week with varying species, are in line with protecting public health and individuals, and they rely on the current knowledge of relevant dose responses and toxicokinetics of MeHg. Whether they are practicable may be questioned. A limitation of the current guidelines is that while they reach those citizens who are conscious about food-mediated risks and benefits, they require a great awareness from those who have to cope with other issues in their every-day lives. Indeed, according to the latest surveys by Evira (2007), Finnish youth has poor awareness of the restrictions given for fish consumption. We highlight that intake of fish oil supplements or similar health products instead of fish itself is not a resolution to the problem because the wide variety of beneficial effects from fish consumption cannot be achieved through only one health product.

4.4 Uncertainties and suggestions for further work

Our study had its limitations. Using food records to estimate food (here fish) consumption is generally accepted as the best means to collect information on the average individual diet, especially in children. The method involves, however, the possibility of both conscious and unconscious over- or underreporting that can add inaccuracy in the estimated intake levels into both directions (over- and underestimation). Notably, the records used included 3 consecutive days, 2 weekdays and 1 weekend day. Additionally, day-to-day variation and nuisance effects (e.g. day of the week, interview sequence) were controlled for by the method

of Nusser et al. (1996) to obtain the long-run average daily intakes, or usual intakes. EFSA (2009) recommends using 2 non-consecutive days for children up to 10 years in food consumption estimates in order to have data on independent days and a better estimate of the existing intra-individual variability than consecutive records. Some precision on the estimates could have been reached by using 2 non-consecutive days. A third limitation is that MeHg was not measured directly but derived from total Hg measurements with a single averaging conversion factor. There are indications that more than 95% of total Hg present in edible fish muscle is in the organic form, MeHg (Bloom 1992, Downs et al. 1998, Freije & Awadh 2009, Kannan et al. 1998). Our conversion factor of 0.93 was based mainly on the study of Westöö (1973) because it specifically concerns fish species in the Baltic Sea region. Our calculation method was deterministic in a sense that the whole MeHg concentration variation in each species was not included in the estimation. As we however examined usual long-term intakes, it is generally assumed that variations in concentrations are levelled out on the long-run. A more precise estimate for the usual MeHg intakes presented in this study could be achieved by using individual body weights rather than mean population-based weights from 1993. As Finnish children today weigh more than in the 1990s (Virtanen, unpublished data), the long-term mean daily intakes we present can be overestimates. Specifically, individual body weight adjustments could specify the minimum and maximum intakes reported. The study subjects were recruited from two urban areas: Oulu on sea coast and Tampere in inland, and we assume them to represent fairly well the fish consumption habits in Finnish children, including rural regions. We assumed the net absorption efficiency of MeHg from fish to be 100%.

Further population-based estimations of the long-term mean daily MeHg intake from fish in Finland should aim at assessing gender-specific intakes in infants, preschoolers, school children, adolescents, adults and elderly people. This would provide firmer grounds for

lifetime exposure estimations and comparisons to the limit values set by international expert bodies. Because safety margins in relation to the threshold levels are still narrow, calculations could be performed on how different children-specific fish consumption scenarios influence the net benefits or likelihood of net benefits of fish consumption. As in the paper by Leino et al. (2011), these calculations should be implemented as quantitative benefit-risk comparisons if feasible, to include in the analysis any possible food-related beneficial aspects such as the omega-3 polyunsaturated fatty acids in fish that may outweigh the potential contaminant risks. These would be a useful corrective means to enable successful risk management based on the net public health benefit. Currently, food-mediated contaminant risks can be managed successfully only as trade-offs, especially in the case of fish.

5. Conclusions

MeHg long-term mean daily intakes among Finnish children varied greatly between individuals and ages. In each age group of the study population there were a proportion of children with their long-term mean daily intake exceeding considered safe limits set by U.S.EPA or JECFA. Further, estimated long-term mean daily MeHg intakes increased mostly along with older age. These facts demand for an in-depth quantitative benefit-risk assessment of fish consumption in children of all age groups.

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Conflict of Interest

The authors declare there are no conflicts of interests.

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