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Assessment of mercury intake associated with fish consumption in Serbia

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A b s t r a c t: Nutritional benefits of fish can be attributed primarily to the content of high-quality proteins, vitamins, elements and omega-3 polyunsaturated fatty acids. On the other hand, fish and fishery products are susceptible to contamination by chemicals that have been recognized as ubiquitous environmental pollutants such as toxic elements and polychlorinated organic compounds. Fish consumption could be therefore considered as one of the major sources of human exposure to all above-mentioned environmental contaminants. This paper is focused on mercury (Hg) that enters the environment by both, natural means (such as volcanic activity, erosions and weathering, factors which contribute to the presence of Hg in water, soil and the atmosphere) and human activities – mining, fossil fuels combustion, industrial emissions, direct application of fertilizers and fungicides as well as disposal of solid waste.

Total concentrations of Hg were measured in fish muscle and canned fish products available on Serbian market. Total of 651 samples were analyzed: 350 samples of marine fish (hake, mackerel, sprat, scorpanea, gilthead, salmon), 34 samples of freshwater fish (trout and carp) and 267 samples of canned fish products (tuna and sardines). Data were collected during 2011. For the purpose of intake assessment, we used the data obtained from the GEMS/Food Consumption Cluster Diets database. According to this source, estimated average weekly consumption of marine fish is 106.4 g/week, while freshwater fish and canned fish contribute to the consumption with 29.4 g and 18.2 g/week respectively.

Mercury concentrations in marine fish were in the range of 0.005–0.208 μgg^{-1} (mean 0.040 µgg-1); in freshwater fish 0.005–0.099 μgg^{-1} (mean 0.020 μgg^{-1}) and in canned products they were in the range of 0.005–0.642 μgg^{-1} (mean 0.064 μgg^{-1}). All analyzed samples contained mercury below the maximum level laid down by the European Union and Serbian regulation. The estimated weekly intake for total mercury, based on mean mercury value in fish and average body mass of 70 kg, was 0.095 µg/kg b.w./week.

Based on FAO/WHO recommended safe limit and on obtained results, we can conclude that the intake of mercury in the case of consuming fish and canned fish products is lower than the safe limit.

Key words: *intake, fish, mercury.*

Introduction

Health benefits related to fish consumption are primarily due to the presence of proteins, minerals, vitamins and unsaturated essential fatty acids, especially polyunsaturated fatty acids (PUFAs) like omega-3 PUFAs. In contrast to the health benefits of dietary fish intake, an issue of concern related with frequent fish consumption is the risk derived from exposure to persistent environmental contaminants, both carcinogenic (e.g., dichlorodiphenyltrichloroethane (DDT), polychlorinated biphenyl's (PCBs), dioxin, etc.), and/or non-carcinogenic (e.g., mercury). Although these contaminants are present in the environment at low levels, they could be taken up by aquatic organisms and undergo bioconcentration and bioaccumulation processes, resulting in progressively higher levels of these compounds in the food chain, particularly in the case of longer-living and predatory fish. Based on literature, fish consumption could be considered as one of the major sources of human exposure to the above-mentioned environmental contaminants (*EFSA,* 2005a; *Storelli,* 2008).

Mercury (Hg) enters the environment by both natural sources – volcanic activity, erosions and weathering, factor that contributes to the presence

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of mercury in the water, soil and the atmosphere, and human activities – mining, fossil fuels combustion, industrial emissions, direct application of fertilizers and fungicides as well as disposal of solid waste. Mercury exists in three forms – metallic (elemental), inorganic and organic mercury compounds. The inorganic mercury can be converted to methylated form by microorganisms especially in aquatic systems and this form is predominantly present in fish (*Merritt and Amirbahman*, 2009; *Ersoy and Çelik*, 2010; *Saei-Dehkordi et al.,* 2010). Tuna and swordfish are large predatory species that tend to accumulate relatively high levels of methyl mercury (MetHg). In other foods, mercury is mainly present in inorganic form; however, dietary inorganic mercury is of little toxicological concern.

Exposure to high levels of mercury can cause permanent damage to the brain, kidneys, and developing fetus (*WHO*, 1990; *Schantz et al.,* 2003; *Hightower and Moore*, 2003; *Hites et al.,* 2004). Effects on brain functioning may manifest as irritability, tremors, changes in vision or hearing, and memory problems. Vomiting, diarrhea, increases in blood pressure or heart rate, skin rashes, and eye irritation can also occur. Young children are more sensitive to mercury than adults. Mercury in the mother's body is transferred to the fetus and may accumulate there. It can also pass to a nursing infant through breast milk. Children poisoned by mercury may develop conditions of nervous and digestive systems, as well as kidney damage. Adequate data on carcinogenicity of Hg are currently unavailable for some of its forms. The U.S. EPA (United States Environmental Protection Agency) has determined that mercury chloride and methyl mercury are possible human carcinogens (*Risher and DeWoskin,* 1999).

Global fish consumption varies significantly from one country to another depending primarily on geographic position, tradition, economic development, dietary habits etc.

Average annual fish consumption in Serbia is about 5 kg per capita (Baltic et al., 2009), which is significantly lower compared to the global consumption (16.4 kg) or EU average of 11 kg (in Austria) up to 57 kg (in Portugal) (*Lekic-Arandjelovic et al.,* 2008).

Recently, fish consumption has increased in Serbia primarily due to its health benefits (*Ćirković et al.,* 2011; *Trbović et al.,* 2011). Therefore, the aim of this work is quantitative evaluation of the intake of mercury, and assessment of potential health risk related to fish consumption among Serbian population.

Materials and methods

Contamination data

Total concentrations of Hg were measured in fish and canned fishery products available on Serbian market. Total of 651 samples were analyzed: 350 samples of marine fish (hake-*Merluccius merluccius,* mackerel-*Scomber scombrus*, sprat-*Sprattus sprattus,* scorpanea-*Scorpaena scrofa,* gilthead-*Sparus aurata,* salmon-*Salmo salar*), 34 samples of freshwater fish (trout-*Salmo irideus* and carp-*Cyprinus carpio*) and 267 samples of canned fish products (tuna and sardines). These data were collected during 2011. Determination was carried in compliance with ISO standard 17025.

Fish was kept frozen at -20 °C before analysis. Edible parts were chopped into 2-3 cm thick portions and homogenized. Samples for Hg analysis were prepared by microwave digestion (ETHOS Milestone). Analyses were carried out on atomic absorption spectrometer Varian "SpectrAA 220" with VGA 77 hydride system. Cold vapor technique was applied. The limit of quantification was 5 ng/g. Analytical quality control was achieved using certified reference material BCR 185R. Replicate analyses were in the range of certified values.

Intake assessment

Total diet study has not been undertaken in Serbia, so far. Instead of such comprehensive data base, for the purpose of intake assessment, we used the only available surrogate taken from the GEMS/Food Consumption Cluster Diets database (*FAO/WHO*, 2006). According to this source, estimated average weekly consumption of marine fish is 106.4 g/week, while freshwater fish and canned fish contribute to the consumption with 29.4 g and 18.2 g/week respectively.

The following formula was used for calculation of intake assessment expresed as weekly intake (WI) in μg/kg b.w.:

$$WI = \frac{\text{Weekly consumption data x Concentration of compound}}{\text{Body weight}}$$

In order to calculate Hg intake, we have developed four different scenarios, obtained using 2×2 design. Namely, two concentration levels of certain contaminants were chosen: an average and a maximal value combined with two body mass values were used: body mass of 70 kg that represents 50[th] percentile of population and body mass of 51 kg that represents 5[th] percent of total population of 808 adult healthy volunteers (395 female and 413 male), included in national survey from the Department of endocrinology of Clinical center of Novi Sad (*Srdić,* 2002).

For each of four chosen scenarios, hazard index (HI) was calculated based on formula given below:

$$H = \frac{\text{calculated weekle intake for mercury}}{\text{provisional tolerable weekly intake}}$$

Descriptive statistics was carried out using ORIGIN software (version 7.1).

Results and discussion

Contents of Hg in fish and fishery products are given in table 1.

In order to protect public health, maximum levels of Hg in fishery products are laid down by the Commission Regulation (EC) No 1881/2006 of 19 December 2006 (*European Commission,* 2006) and Serbian legislation. Hg limit for fishery products in general is 500 ng/g fresh weight, (1000 ng/g fresh weight for anglerfish, Atlantic catfish, bass, blue ling, bonito, eel, halibut little tuna, marlin, pike, plain bonito, Portuguese dogfish, rays, redfish, sail fish, scabbard fish, shark, snake, mackerel, sturgeon, swordfish and tuna). These limits were not exceeded in any of the analyzed samples.

The highest average mercury concentration was found in canned tuna – 0,075 $\mu g g^{-1}$. Mean mercury content in marine fish – 0,040 $\mu g g^{-1}$ was two times higher than mean mercury content in freshwater fish – 0,020 $\mu g g^{-1}$.

Weekly intake of Hg through fish consumption among Serbian population has been calculated by deterministic model using fixed mean or maximum values for contaminants concentrations. The estimated weekly intakes of contaminants by adult male of 70 kg (50th percentile) and of 51 kg body (5th percentile) weights are summarized in table 2.

Table 1. Structure and contents of Hg in a composite fish dish

Tabela 1. Struktura i sadržaj Hg u kompozitnom ribljem obroku

Fish / Riba	**Intake (g/week) / Unos (g/nedelja)**	**Intake (%) / Unos (%)**	**Hg (ng/g)**		
			min	**max**	**mean**
Marine fish / Morska riba	106.4	69.09	5	208	40
Freshwater fish / Slatkovodna riba	29.4	19.09	5	99	20
Canned products / Riblje konzerve	18.2	11.82	5	642	64
Composite fish dish / Kompozitni riblji obrok					
Σ	**154**	**100**	**5**	**279.4**	**43.1**

Table 2. Weekly intakes of Hg via fish consumption

Tabela 2. Nedeljni unos Hg preko konzumirane ribe

Weekly intake of Hg (μg/kg b.w.) / Nedeljni unos Hg (μg/kg telesne težine)	
Mean concentration / Srednja koncentracija	Maximum concentration / Maksimalna koncentracija
50th percentile – 70 kg	
0.095	0.615
5th percentile – 51 kg	
0.130	0.844

In the period 2004-2007, several expert opinions concerning human dietary exposure to Hg were issued *(EFSA*, 2004, 2005a; *UK-COT*, 2004, 2007; *Japan FSC,* 2005; *Canada BCS,* 2007). All these documents indicate that fish and seafood are the major source of Hg intake in humans. Depending on species, MetHg accounts for 70-100% total Hg in fish (*EFSA,* 2005a). However, for conservative assessment purposes, it is generally assumed that 100% of the Hg found in fish and shellfish is MetHg. According to European Food Safety Authority (EFSA), the range of average fish consumption is from 10 to 80 g per day for six European countries, corresponding to Hg weekly intake from 1.3 to 92 µg, per person (*EFSA,* 2004). This is similar to the intake values calculated in our study (6.6 – 43 µg, per person). Substantially higher values were reported for Faroe Islands (average 252 µg/week), while in the Seychelles the daily Hg intake was estimated to be 103 µg, assuming annual consumption of fish of 75 kg (205 g per day) per capita (*Robinson and Shroff,* 2004). The estimated average weekly intake of Hg by the French population is 68 µg for adults aged 15 years or more (corresponding to 1.1 µg/kg b.w. per week for a 60 kg person) and 55 µg for children aged 3-4 years *(Leblanc et al.,* 2005). Estimated weekly intake of total Hg in the population from Catalonia (*Bocio et al.,* 2005) is 148 µg, corresponding to 2.1 µg/kg b.w. per week, and is principally due to the high consumption of fish in this region.

For the purpose of evaluating the health risk, the estimated dietary exposures were compared to the corresponding health based guidance values. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a provisional tolerable weekly intake (PTWI) of 1.6 µg MetHg/kg b.w. (i.e. 0.228 µg/kg b.w./day) based on epidemiological studies that investigated the relationship between maternal exposure to Hg and impaired neurodevelopment in their children (*FAO/WHO*, 2003).

Calculated HIs, for all four scenarios for Hg are given in table 3 below.

HI values have shown that in all four anticipated scenarios, there is no risk of overexposure to Hg due to fish consumption. In general, human health risk assessment of individual contaminants can be relatively simple procedure, due to establishment of various health based guidance values determined by national and international authorities. However, humans are exposed to a mixture of chemicals at any given time. It should be mentioned that other chemicals such as cadmium, lead, organochlorine insecticides (other than DDT), polychlorinated dibenzo dioxines (PCDDs) and polychlorinated dibenzo furanes (PCDFs) etc. may be also present in fish. It is however, difficult to predict impact to human health from the exposure to all these toxic compounds, particularly in view of the other exposure patterns.

Conclusion

Contaminant levels of Hg are sufficiently high in some fish to cause adverse human health effects in people consuming large quantities. Although all calculated values of HIs related to fish consumption are below the critical value of 1, without reliable data for dietary exposure, the credible conclusion regarding safe level in Serbian population cannot be drawn. Taking into consideration global decline in contamination levels due to the public awareness on health impacts of these contaminants and consequent restrictions in their production and utilization, the situation in which dietary intake would result in immediate risk, is not likely to occur. Therefore, the current level of total Hg in fish and fishery products available at the Serbian market does not pose a threat to consumers' health. This synergistic effect of risk decline on one side and preservation of well-known benefits from fish consumption on the other, opens a wide array of possibilities for fishery industry to offer higher quantities of reasonably safe and nutritionally benefitial products.

Table 3. Hazard indexes for Hg intake

Tabela 3. Indeksi opasnosti za unos Hg

Scenario	**Mean 50th**	**Max 50th**	**Mean 5th**	**Max 5th**
HI	0.059	0.384	0.081	0.528

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Procena unosa žive kroz konzumaciju ribe u Srbiji

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R e z i m e: Nutritivna korist od konzumacije ribe ogleda se, pre svega u sadržaju visoko vrednih proteina, vitamina, makro i mikroelemenata i omega-3 polinezasićenih masnih kiselina. Sa druge strane, riba i proizvodi od ribe su u značajnoj meri podložni hemijskoj kontaminaciji ubikvitarnim zagađivačima kao što su teški metal i polihlorovana organska jedinjenja. Zbog svega ovoga, konzumacija ribe se smatra jednim od najznačajnijih izvora izloženosti navedenim kontaminentima. U radu je ispitivana kontaminacija živom koja se u spoljašnjoj sredini može naći kao posledica prirodnih procesa (vulkanska aktivnost, erozija tla i klimatska dešavanja koja doprinose prisustvu žive u vodi, zemljištu i atmosferi), kao i aktivnosti čoveka – eksploatacija ruda, sagorevanje fosilnih goriva, emisija industrijskih gasova, direktna primena veštačkih đubriva i fungicida, kao i neadekvatno odlaganje čvrstog otpada.

Sadržaj ukupne žive je određivan u svežoj i konzervisanoj ribi sa srpskog tržišta. Ispitan je 651 uzorak: 350 uzoraka morske ribe (oslić, skuša, sardela, škarpina, orada i losos), 34 uzorka slatkovodne ribe (šaran i pastrmka) i 267 uzoraka konzervisane ribe (tuna i sardela). Svi uzorci su analizirani tokom 2011. godine.

Za procenu unosa korišćeni su podaci iz „GEMS/Food Consumption Cluster Diets database“. Prema ovom izvoru, procenjena prosečna nedeljna konzumacija morske ribe iznosi 106,4 g, slatkovodne 29,4 g dok je ova vrednost za konzervisane proizvode od ribe 18,2 g.

Sardžaj žive u morskoj ribi se kretao u opsegu od 0,005 do 0,208 μgg^{-1} (srednja vrednost 0,040 μgg^{-1}); u slatkovodnoj ribi 0,005–0,099 μgg^{-1} (srednja vrednost 0,020 μgg^{-1}), dok se u konzervisanim ribljim proizvodima sadržaj žive kretao od 0,005 do 0,642 μgg^{-1} (srednja vrednost 0,064 μgg^{-1}). Nivo žive u svim ispitanim uzorcima je bio ispod maksimalno dozvoljenih vrednosti propisanih relevantnom legislativom EU kao i domaćim propisima. Procenjeni nedeljni unos žive baziran na srednjoj vrednosti žive u konzumiranoj ribi i prosečnoj telesnoj težini od 70 kg, bio je 0,095 μg/kg telesne mase nedeljno.

Na osnovu preporuka FAO/WHO i dobijenih rezultata, može se zaključiti da je unos žive pri konzumaciji morske i slatkovodne ribe i konzervisanih ribljih proizvoda, niži od preporučenih graničnih vrednosti.

Ključne reči: *unos žive, riba.*

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