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Organotin intake through fish consumption in Finland

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ABSTRACT

Background: Organotin compounds (OTCs) are a large class of synthetic chemicals with widely varying properties. Due to their potential adverse health effects, their use has been restricted in many countries. Humans are exposed to OTCs mostly through fish consumption.
Objectives: The aim of this study was to describe OTC exposure through fish consumption and to assess the associated potential health risks in a Finnish population.
Methods: An extensive sampling of Finnish domestic fish was carried out in the Baltic Sea and freshwater areas in 2005–2007. In addition, samples of imported seafood were collected in 2008. The chemical analysis was performed in an accredited testing laboratory during 2005–2008. Average daily intake of the sum of dibutyltin (DBT), tributyltin (TBT), triphenyltin (TPhT) and dioctyltin (DOT) (ΣOTCs) for the Finnish population was calculated on the basis of the measured concentrations and fish consumption rates.
Results: The average daily intake of ΣOTCs through fish consumption was 3.2 ng/kg bw day^{-1}, which is 1.3% from the Tolerable Daily Intake (TDI) of 250 ng/kg bw day^{-1} set by the European Food Safety Authority. In total, domestic wild fish accounted for 61% of the ΣOTC intake, while the intake through domestic farmed fish was 4.0% and the intake through imported fish was 35%. The most important species were domestic perch and imported salmon and rainbow trout.
Conclusions: The Finnish consumers are not likely to exceed the threshold level for adverse health effects due to OTC intake through fish consumption.

1. Introduction

Organotin compounds (OTCs) are a large class of synthetic compounds with widely varying chemical properties (Champ and Seligman, 1996; Hoch, 2001). Due to their strong biocidal activity, the trisubstituted OTCs, tributyltin (TBT) and triphenyltin (TPhT), have been used extensively in antifouling paints and in agriculture. However, TBT and TPhT have been shown to be very toxic to marine organisms (McAllister and Kime, 2003) and the use of TBTs and TPhTs is currently restricted in many countries. In the European Union, the use of TBT is currently banned in all marine vessels trafficking in EU waters, as well as in plant protection agents (EC, 2003). Di- and monosubstituted OTCs, which are not as toxic, are still used in plastics industry as stabilizers of polyvinyl chloride and as catalysts in the production of polyurethane foams and silicones. Further restrictions on the use of the trisubstituted OTCs as well as dibutyltin (DBT) and dioctyltin (DOT) will be effective during 2010–2015 (EC, 2009).

Due to decades of leaching from antifouling paints applied on boat hulls and other underwater structures, TBT and TPhT have caused serious environmental problems in many harbors and dockyards worldwide (Antizar-Ladislao, 2008). OTCs have a tendency to bioaccumulate in the marine ecosystem (Shawky and Emons, 1998), and the consumption of contaminated fish poses a potential health risk for humans. In spite of the recent restrictions on the use of OTCs, concentrations in the marine environments may not be immediately reduced, due to the long persistence time of these compounds in sediments. For example, under unfavorable conditions in northern dark, cold and anoxic sediments, the half-time of TBT may reach several decades (Viglino et al., 2004). Moreover, mono- and di-substituted OTCs are still widely permitted in various consumer products and water pipes. From them, OTCs may be leached and carried into the water systems (Fent, 1996).

Abbreviations: GC/MS, gas chromatography/mass spectrometry; LOQ, limit of quantification; *m/z*, mass-to-charge ratio; EFSA, European Food Safety Authority; TDI, tolerable daily intake; OTC, organotin compound; MBT, monobutyltin; DBT, dibutyltin; TBT, tributyltin; MPhT, monophenyltin; DPhT, diphenyltin; TPhT, triphenyltin; DOT, dioctyltin; fw, fresh weight; bw, body weight

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Humans are exposed to OTCs mostly through food, and especially fish consumption (Rantakokko et al., 2006, 2008; Toyoda et al., 2000). There are no epidemiologic data on the health effects on OTCs, but according to numerous animal studies, the critical effect of OTCs is immunotoxicity. A scientific panel of the European Food Safety Authority (EFSA) has assessed the health risks to consumers associated with exposure to OTCs in foodstuffs and established a group Tolerable Daily Intake (TDI) of 250 ng/kg body weight (bw) for the sum of DBT, TBT, TPhT and DOT, due to the similarity of their immunotoxic properties (EFSA, 2004).

The aim of this study was to describe the OTC exposure through fish consumption and to assess the associated potential health risks in a Finnish population.

2. Material and Methods

2.1. Sample collection and chemical analysis

The project was coordinated by the Finnish Food Safety Authority Evira. The fish samples from Finnish waters were collected by the Finnish Game and Fisheries Research Institute between May 2005 and April 2007. A detailed description of the study areas, sample preparation, analysis and OTC concentrations has been given previously (Rantakokko et al., 2010). In the present study, we used OTC concentration data from (a) 61 samples of various species from five open sea areas of the Finnish coast of the Baltic Sea and 23 samples of various species from three large freshwater lakes ($n=84$), (b) 7 samples of domestic farmed fish, (c) 2 samples of imported fish and (d) 113 samples of perch (*Perca fluviatilis*) from 10 coastal and 12 freshwater harbors, dockyards and busy shipping routes. In addition, 50 samples of imported seafood were obtained by the Finnish Food Safety Authority's inspectors from Finnish first destination and veterinary border controls between June and September 2008. The preparation and chemical analysis of the samples of imported seafood followed the same methods that were used for the analysis of the domestic fish samples. The chemical analysis of monobutyltin, dibutyltin (DBT), tributyltin (TBT), monophenyltin, diphenyltin, triphenyltin (TPhT) and dioctyltin (DOT) by high-resolution GC/MS was performed in an accredited testing laboratory (Code T077, EN ISO/IEC 17025) of the National Institute for Health and Welfare, Chemical Exposure Unit.

2.2. Calculation of intake through fish consumption

In the intake calculations, a sum of DBT, TBT, TPhT and DOT was used, in order to enable the comparison between the calculated average daily intake and the EFSA's TDI. The sum of these four compounds is hereafter referred to as ΣOTC concentration. The average daily intake of ΣOTCs (ng/kg bw day^{-1}) through fish consumption was calculated by multiplying the consumption of fish (g day^{-1}) with the median ΣOTC concentration of the corresponding species (ng/g fw) and dividing the product by a body weight of 60 kg. Concentrations that were below the limit of quantification (LOQ) were calculated as one half of the respective LOQ.

First, we calculated the ΣOTC intake of the average Finnish consumer, specified by the most commonly consumed species of fish in Finland. The calculation was performed by multiplying the consumption of individual species with its corresponding median ΣOTC concentration and dividing the product by a body weight of 60 kg. ΣOTC concentration data of Baltic herring (*Clupea harengus membras*), pike (*Esox lucius*), perch, vendace (*Coregonus albula*), whitefish (*C. lavaretus*), pike-perch (*Sander lucioperca*), salmon (*Salmo salar*), burbot (*Lota lota*), sprat (*Sprattus sprattus*), flounder (*Platichthys flesus*) and bream (*Abramis brama*) ($n=84$) as well as farmed rainbow trout, whitefish and char (*Salvelinus alpinus*) ($n=7$) and various species of imported fish ($n=52$, of which 2 were from the 2005–2007 sampling and 50 from the 2008 sampling) were used (Table 1). For some species, no concentration data was available and data from a similar species was used instead. For sea trout (*S. trutta m. trutta*), rainbow trout (*Oncorhynchus mykiss*), grayling (*Thymallus thymallus*) and cod (*Gadus morhua*), ΣOTC concentration was assumed to be that of salmon, since the species are close relatives and/or are pelagic. For roach (*Rutilus rutilus*) and ide (*Leuciscus idus*), ΣOTC concentration was assumed to be that of bream, since they belong to the same family. The average consumption of different domestic and imported fish species in 2007 was obtained from the Finnish Game and Fisheries Research Institute.

Secondly, we used a four-field approach in order to estimate the exposure of the non-average Finnish man and woman by applying four different scenarios: (a) average consumption of uncontaminated seafood, (b) average consumption of contaminated seafood, (c) high consumption of uncontaminated seafood and (d) high consumption of contaminated seafood (Table 2). Here, we have used the term "uncontaminated seafood" to describe the fish collected from open sea areas and freshwater lakes ($n=84$), farmed fish ($n=7$) and imported fish and seafood

Table 1
Average daily intake[a] of ΣOTCs from different species of domestic and imported seafood as well as total average daily intake of ΣOTCs from fish (ng/kg bw day^{-1}) and proportion from total average daily intake for each fish species.

Fish species	n	ΣOTC[b] (ng/g fw)	Consumption (g day^{-1})	Average daily intake[a] (ng/kg bw day^{-1})	Proportion from total average daily intake (%)
Domestic wild fish					61
Herring	20	13 (6.2–33)	1.1	0.24	7.3
Pike	10	12 (1.1–51)	2.0	0.42	13
Perch	13	16 (1.2–44)	1.9	0.52	16
Vendace	4	1.9 (0.94–8.2)	1.6	0.05	1.6
Whitefish	7	4.5 (0.97–18)	0.82	0.06	1.9
Pike-perch	8	31 (1.2–75)	0.79	0.41	13
Salmon, sea trout, rainbow trout, grayling, cod[c]	6	11 (7.2–13)	0.96	0.18	5.4
Burbot	5	5.9 (0.85–55)	0.16	0.02	0.5
Sprat	3	8.9 (7.8–11)	<0.01	<0.01	
Flounder	2	8.1 (4.7–11)	<0.01	<0.01	
Bream, roach, ide[d]	6	8.0 (0.95–205)	0.60	0.08	2.5
Domestic farmed fish					4.0
Rainbow trout	4	2.3 (2.0–3.9)	2.9	0.11	3.5
Other	3	2.4 (2.3–5.0)	0.47	0.02	0.6
Imported fish					35
Salmon and rainbow trout	7	3.2 (0.98–54)	10	0.54	17
Tuna	10	2.2 (0.85–225)	4.7	0.17	5.3
Saithe	3	1.3 (0.94–1.5)	1.1	0.02	0.7
Shrimp	5	0.85 (0.85–0.85)	1.6	0.02	0.7
Herring and sardine preserves	6	3.0 (1.1–33)	1.4	0.07	2.1
Other	21	1.4 (0.85–8.5)	13	0.29	9.1
Total	143	6.8 (0.85–225)	45	3.2	100

[a] Calculated for a person weighing 60 kg.
[b] Median and range for the sum of dibutyltin (DBT), tributyltin (TBT), triphenyltin (TPhT) and dioctyltin (DOT).
[c] ΣOTC concentration of salmon was used.
[d] ΣOTC concentration of bream was used.