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# Organotin levels in seafood and its implications for health risk in high-seafood consumers

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**Abstract**

Fish and fishery products are considered as the main source of organotin compounds (OTC). Unfortunately, little national contamination data is available to assess food exposure of organotins from French consumers. To provide a more accurate estimate of risks to human health, the butyltin, phenyltin and octyltin compounds sampling in four French coastal areas were measured in 159 composite samples (96 fresh and frozen fish, 28 mollusks, 14 crustaceans, 1 echinoderm, 11 canned foods, 4 smoked fish, 5 prepared seafood-based dishes) by capillary gas chromatography coupled with a microwave induced plasma atomic-emission spectrometer (CGC–MIP-AES). In these samples, butyltins were usually predominant and the range of the contamination levels was generally below those of earlier studies (fish: mean 5.6; min–max 1.1–23 µg/kg; fishery products: mean 6; min–max 0.8–14 µg/kg). Fish, especially tuna, salmon, mackerel, saithe/coalfish and cod were largely the main contributors (38%) to the total organotin exposure. With the supplementary contribution of great scallop, surimi, squid and oysters, the exposure exceeded 50% in all. However, the utmost OTC exposure was lesser than 47% of the provisional tolerable weekly intake [EFSA (European Food Safety Agency). Opinion of the Scientific Panel on Contaminants in the food chain on a request from the Commission to assess the health risks to consumers associated with exposure to organotins in foodstuffs. (Question N°EFSA-Q-2003-110). The EFSA Journal, 102, 1–119, 2004. http://www.efsa.eu.int]. Nobody would exceed this limit. Finally, as this study has some limitations and since some other sources and health effects have not been clearly evaluated, it appears rational from public health and environmental viewpoints to continue to reduce the OTC levels in the environment.


*Keywords:* Organotins; Speciation; Seafood safety; Dietary exposure; Survey; Ocean and human health

## 1. Introduction

Occupational exposure represents the greatest exposure to organotin compounds (OTC). Food, especially fish and fishery products is considered to be the main sources. The main species are likely to be tri-substituted compounds (tributyltin TBT and triphenyltin TPT), which have been used extensively as biocides in wood preservatives, in antifouling paints for boats and as pesticides (ATSDR, 2003; EFSA, 2004; Duft et al., 2005; AFSSA, 2006). Mono- and di-substituted are generally used in mixtures in various amounts as polyvinyl-chlorides stabilizers (PVC). TBT compounds are on the list of priority substances in the field of water policy (EC, 2001). Antifouling paints have been restricted in many

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countries because of the recognized adverse effects of these compounds on both humans and aquatic ecosystems. Regulation 782/2003/EC bans the application of OTC on ships as from 7/1/2003 and to eliminate their presence on ships from 1/1/2008 (EC, 2003). Nevertheless, because these compounds are persistent in the environment and tend to accumulate through the food chain (Duft et al., 2005), reservoir sources may continue to contribute substantially to human exposure of these compounds for a long time.

Recently, many toxicological evaluations were published (WHO-IPCS, 1999a,b, 2001, 2006; ATSDR, 2003; EC, 2004; EFSA, 2004; AFSSA, 2006). A no observed adverse effect level (NOAEL) for immunotoxicity of 0.025 mg/kg bw/day was identified for TBT oxide from chronic feeding studies in rats (Vos et al., 1990; Cooke et al., 2004; Tryphonas et al., 2004). An oral reference dose (RfD), a guidance value for oral exposure, and an intermediate-duration oral minima risk level (MRL) of 0.0003 mg/kg/day for tributyltin oxide have been successively derived (US EPA, 1997; WHO-IPCS, 1999b; ATSDR, 2003). An uncertainty factor of 100 was applied to the NOAEL (10 for animal to human extrapolation and 10 for human variability). Because tributyltin (TBT), dibutyltin (DBT), triphenyltin (TPT) and dioctyltin (DOT) exert their immunotoxic effects by similar mode of action and potency, it seemed reasonable to establish a group tolerable daily intake (TDI) for these OTC (EFSA, 2004; EC, 2004). In the absence of specific studies on combined effects it seemed justified to consider the immunotoxic effects of these compounds as additive. By applying a safety factor of 100, a group TDI of 0.25 μg/kg bw for TBT, DBT, TPT and DOT compounds was established (based on TBT oxide molecular mass, this group TDI is 0.1 μg/kg bw when expressed as Sn content or 0.27 μg/kg bw when expressed as TBT chloride). This group TDI seemed relevant by the AFSSA experts (AFSSA, 2006).

Even though some studies have recently estimated OTC exposure from ingestion of especially fish and shellfish (Cardwell et al., 1999; Kannan et al., 1995; Keithly et al., 1999; WHO-IPCS, 1999b; Belfroid et al., 2000; SCOOP, 2003; EFSA, 2004; OT-SAFE, 2004; Tesfalidet, 2004; Lee et al., 2005; FSA, 2005; AFSSA, 2006; COMPRENDO, 2006), the amount and percent of exposure from food in the French population has not been characterized well.

The aim of the present study is to compensate for this current situation. The food sample size is largely higher than in earlier studies and above all more representative of French high-consumers, as well as the consumption survey used to cross these occurrence data to estimate the exposure is much more accurate than those previously used. For the first time, this paper characterizes specifically the French population's OTC exposure of male and female adults (18–64 years), seniors (+65 years) and women of child-bearing age (18–44 years) from seafood.

## 2. Materials and methods

### 2.1. Food consumption data

Nine hundred and ninety-six fish and fishery products adult high-consumers (at least two meals a week) aged 18 and over were recruited and selected (about 250 people per study zone, see Table 6) by using a validated food frequency questionnaire (FFQ) in four French coastal areas (Fig. 1) within a radius of 20–25 km around (Le Havre (English Channel), Lorient and La Rochelle (Atlantic Ocean), Toulon (Mediterranean Sea)). The distribution of individuals questioned within each region was calculated on the basis of the number of inhabitants published earlier (INSEE, 1999). The recruitment was ensured by a "random-route" protocol i.e. choosing the first address at random then by door-to-door canvassing every five doors. The person who was interviewed was the member of the family whose first name was the first in alphabetical order and who was used to consuming seafood. If the person was not at home or did not fulfill the selection criteria, it was the second one and so on. The inclusion criteria were the age (>18), permanent residency in the zone for several years and the consumption of seafood at least twice a week. For each consumer, frequencies of consumption were collected for 83 seafood species (once a day, three or four times a week, etc.). The portion sizes usually consumed were estimated using a validated book of sample photographs, and weekly consumptions were assessed by crossing the frequencies and the portion sizes. Details of the consumption survey have been described elsewhere (Leblanc, 2006; Bemrah-Aouachria et al. submitted for publication).

### 2.2. Food sampling

All sampling details have been described elsewhere (Sirot et al., in press). Food samples covering 88–100% of fish and seafood mainly consumed were purchased in these regions between January and April 2005 by using an adapted sampling method (Leblanc et al., 2005) based on standardized international methodology (WHO, 1999, 2002, 2004). Sampling was took into account in the form of purchase (fresh, frozen, canned...) and