DIOXINS IN FISH AND SHELLFISH: CONCENTRATIONS AND INTAKE IN JAPAN

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Introduction

Food is generally recognized as the main route of human intake of polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (dl-PCBs), which are known collectively as dioxins. Total diet studies have revealed that over 90% of the dietary intake of dioxins in Japan comes from ingestion of fish and shellfish [1,2]. For risk assessments, it is therefore important to estimate dioxin intakes from fish and shellfish. We have been carrying out a nationwide survey of dioxin concentrations in various retail foods, including fish and shellfish, in Japan. Here, we report the dioxin concentrations in fish and shellfish on the Japanese market and our estimates of the dioxin intakes resulting from consumption of these products.

Materials and Methods

Samples: Fish and shellfish were purchased from different locations throughout Japan during 1998–2009. General edible parts (muscle tissues of fish, crab, and shrimp; muscle tissues of molluscs; and whole soft body tissues for clams and oysters) were homogenized and then analyzed for dioxins.

Dioxin analyses: Extraction, cleanup, and analysis of dioxins were performed in accordance with a previously reported protocol using high-resolution gas chromatography/high-resolution mass spectrometry[3]. The limits of detection (LODs) for PCDD/Fs were 0.01–0.05 pg/g. The LODs for dl-PCBs were 0.1 pg/g for non-*ortho* PCBs and 1 pg/g for mono-*ortho* PCBs. The toxic equivalent (TEQ) concentrations were calculated by using WHO toxic equivalency factors (2005). In calculating the total TEQ concentration in a sample, it was assumed that all isomer concentrations lower than the LODs were equal to zero.

Estimation of dioxin intakes in fish and shellfish by a Monte Carlo simulation: A probabilistic approach with a Monte Carlo simulation was used to estimate dioxin intakes in fish and shellfish. Food consumption data were calculated based on the values obtained in Japanese Nutritional Surveys performed from 2004 to 2006. Dioxin concentrations in fish and shellfish were obtained from a nationwide survey of dioxins in foodstuffs; this survey was performed under a Health Sciences Research Grant from the Ministry of Health, Labor and Welfare of Japan during the period 1998 to 2007. Dioxin intakes were estimated as follows:
Step 1. Assumption of the distribution of consumption of fish and shellfish groups. Fish and shellfish were divided into 13 groups according to the Japanese Nutritional Survey. The probability of consumption of each group was estimated from the group's percentile consumption data in the Japanese Nutritional Survey. A lognormal distribution was assumed for consumption data.
Step 2. Assumption of the distribution of dioxin concentrations in the fish and shellfish groups. Data on dioxin concentrations in fish and shellfish (approximately 650 samples) were divided into the same 13 groups. A lognormal distribution was assumed for the dioxin concentrations in 11 of the 13 groups. A gamma distribution was assumed for the dioxin concentrations in the remaining two groups, because a lognormal distribution was unsuitable for their fitting. The distributions of dioxins as well as PCDD/Fs and dl-PCBs in each group were obtained.
Step 3. Monte Carlo simulation. A random number "a" was generated from a binominal distribution based on the probability of consumption of each of the 13 groups defined in step 1. A random number "b" was generated from the lognormal distribution of the consumption data defined in step 1. "a × b" was defined as the consumption of each of

the 13 groups. A random number “c” was generated on the basis of the distribution determined in step 2 and was defined as the dioxin concentration in each of the 13 groups. “a × b × c” was defined as the dioxin intake from each fish and shellfish group. However, because of the limited availability of data on dioxin concentrations in three of the groups consumed at low rates, dioxin intakes from fish and shellfish in these groups were finally calculated by multiplying the average dioxin concentrations in the groups by their consumption rates. Total dioxin intakes from fish and shellfish were calculated as the sum of dioxin intakes in each fish and shellfish group. The simulation was run with 20,000 iterations.

Results and Discussion

Figure 1 shows the dioxin concentrations (PCDD/Fs + dl-PCBs) in popular retail fish in Japan. The concentrations varied significantly depending on the fish species, although the individual samples had a wide range of dioxin concentrations. In particular, high average concentrations (>1.0 pg TEQ/g fresh weight) were found in yellowtail, mackerel, tuna, horse mackerel, and sardine, which are very popular in the Japanese market and are relatively fatty fish. In tuna, it should be noted that the average concentration was highly elevated by a small number of extremely highly contaminated samples. Among the fish species, saury and eel showed relatively low levels of contamination. Figure 2 shows the dioxin concentrations in popular retail shellfish and molluscs. They were found to have lower dioxin concentrations than the retail fish. This is probably because shellfish and molluscs generally have a low fat content. Figure 3 shows the TEQ contribution of PCDD/Fs and dl-PCBs in relatively highly contaminated samples of various species. There were some differences among the samples. In the fish, dl-PCBs were dominant, contributing about 80% of the total TEQ. On the other hand, in the shellfish and molluscs, PCDD/Fs contributed more than 50% of the total TEQ.

We estimated the dioxin intakes in the general Japanese population by using a Monte Carlo simulation. The distribution of dioxin concentrations in each fish and shellfish group had a longer tail on the higher value side than on the lower side. Therefore, extremely high concentrations of dioxins were generated in the simulations. To avoid this phenomenon, in each group a maximum dioxin concentration of up to three times the observed maximum concentrations was set in the simulations. The distribution of dioxin intake in the general population from fish and shellfish is shown in Figure 4. The estimated average dioxin intake was 1.03 pg TEQ/kg bw/day (0.36 pg TEQ/kg bw/day for PCDD/Fs and 0.73 pg TEQ/kg bw/day for dl-PCBs). The averaged dioxin intake was well below the Japanese tolerable daily intake (TDI) (4 pg TEQ/kg bw/day), and it was almost identical to the dioxin intake of fish and shellfish in our 2006 total diet study [1]. The estimated median, 90th percentile, and 95th percentile of the intake distribution were 0.43, 2.59, and 3.93 pg TEQ/kg bw/day, respectively. The estimated 95th percentile intake was close to the Japanese TDI. However, the 95th percentile intake would have yielded an overestimation of the true value, because fitting of the distribution to the dioxin concentrations in some fish and shellfish groups was not completely achieved. Fish groups, including horse mackerel, sardine, tuna, yellowtail, and fish products (dried and salted fish), are the main sources of dioxin intake in fish and shellfish by the Japanese.

Acknowledgments

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References

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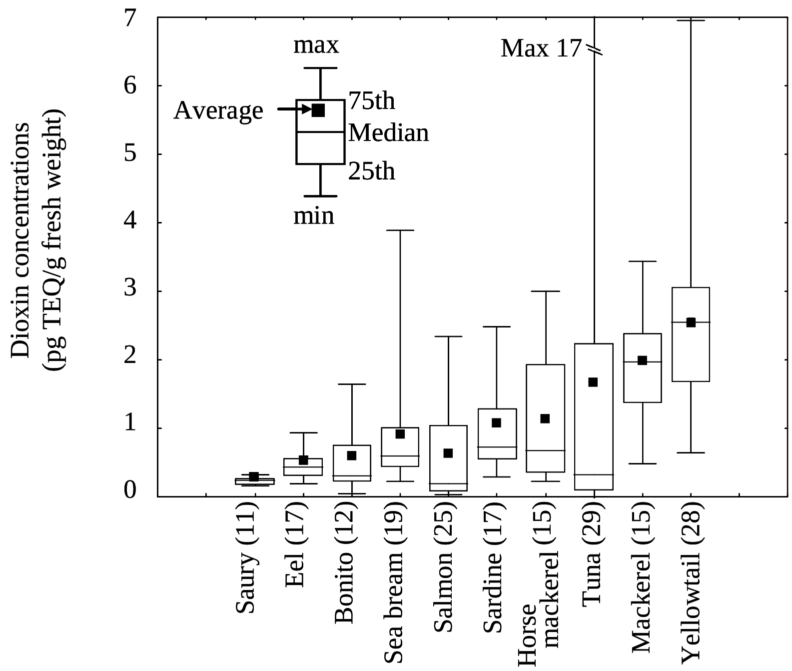

Figure 1. Dioxin concentrations in popular fish samples in Japan. The values in parentheses indicate the number of samples.

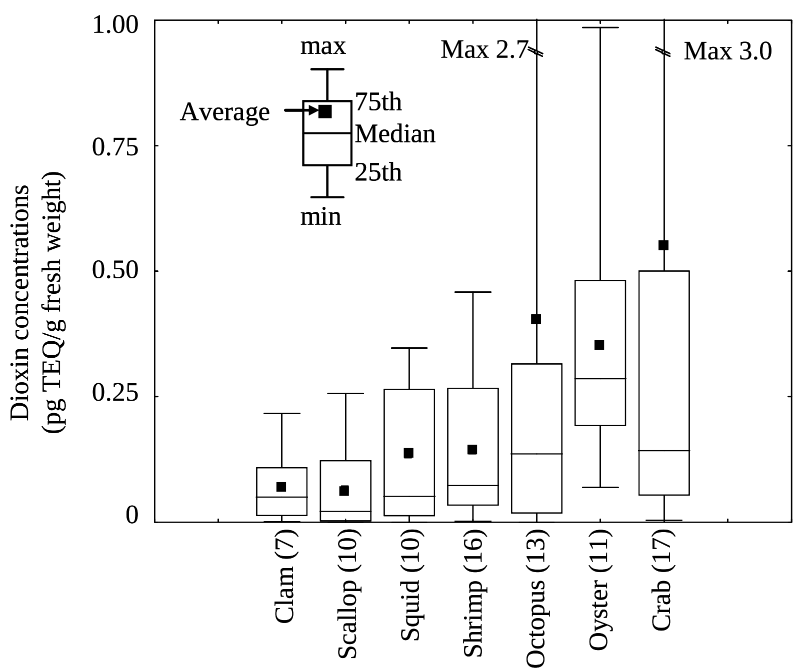

Figure 2. Dioxin concentrations in popular shellfish and mollusc samples in Japan. The values in parentheses indicate the number of samples.

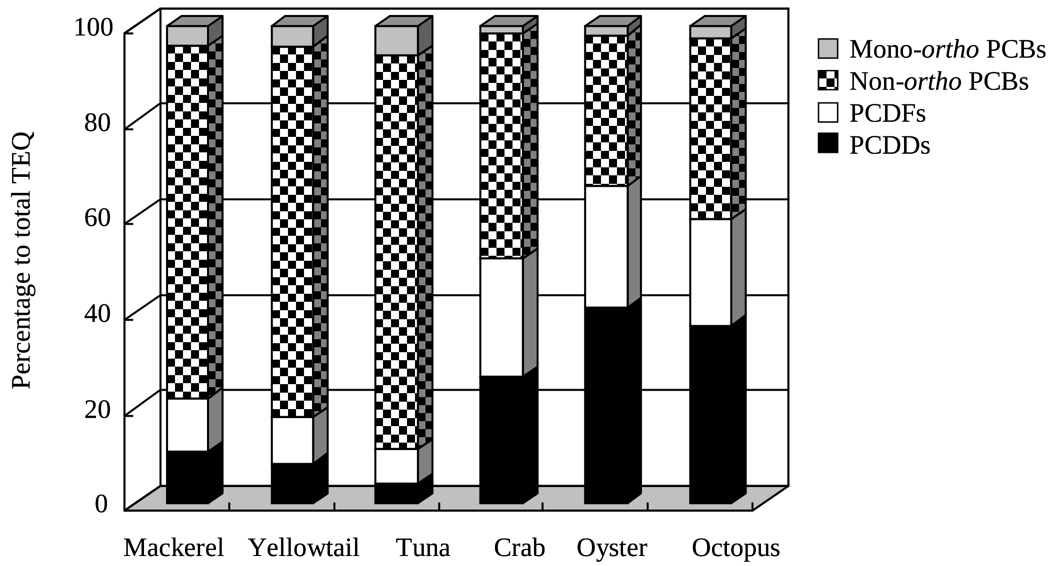

Figure 3. Percentage contributions to the total TEQ among highly contaminated samples.

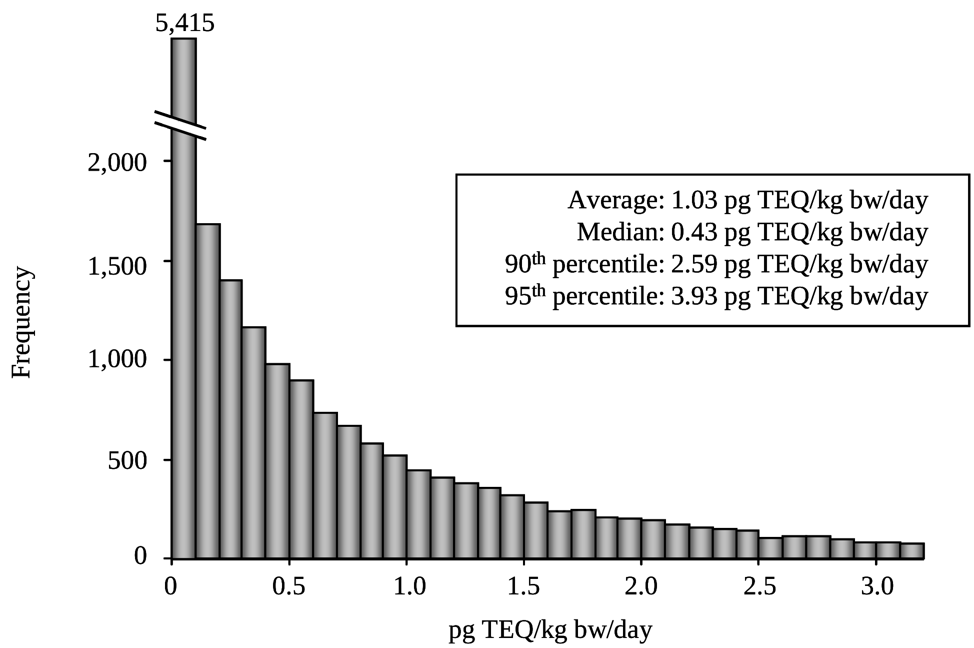

Figure 4. Distribution of dioxin intake in general population in Japan estimated by Monte Carlo simulation.