of Ministers, 2008). As a result, it is considered that only stable α-HBCD possibly finally remains in the environment; therefore, wild and farmed fish inhabiting far from the pollution sources in coastal would naturally have profiles dominated by α-HBCD due to less uptake of prey much polluted with γ-HBCD. In fact, certain fish in deep sea have also been reported to be α-isomer dominant (Takahashi et al., 2010). Hence, correlation between stable α-HBCD but not ΣHBCDs and fat content was re-examined for the wild fish of the order Perciformes on the both cases of each region and all the regions; however, also here, no Spearman's rank correlation was found except for Japanese sea bass of the Tohoku region. The reason would be the same as that for the less correlation between ΣHBCDs and fat content as described in Section 3.1. From these results, it is speculated that the HBCD pollution in Japanese coastal fish at present may be ubiquitous, but uneven in concentration and isomer profiles among regions and fish species.

3.3. Estimation of daily intake of HBCDs

The daily intake of ΣHBCDs from fish was provisionally estimated. According to the Japan Nutrition Survey (The National Nutrition Survey in Japan, 2002), an average Japanese adult consumes 87.8 g of fish and fish products per day. Therefore, the intake of ΣHBCDs from fish can be simply calculated to be from 0 ng person^{-1} d^{-1} for both Pleuronectiformes and Crustacea to 184, 113 and 65.9 ng person^{-1} d^{-1} for Anguilliformes, farmed Salmoniformes and Perciformes, respectively, by multiplying median values of each category in Table 1 with the amount of consumed fish and fish products. However, it can range from 0 ng person^{-1} d^{-1} for Crustacea to 1700, 846 and 407 ng person^{-1} d^{-1} for Clupeiformes, Anguilliformes and Perciformes, respectively, when using the mean values instead of the median. To avoid overestimation and underestimation, it is considered proper to use median values of ΣHBCDs to calculate the representative daily intake of ΣHBCDs. On the assumption that the body weight of an average Japanese adult is 50 kg, the intakes from fish will become about 3.7, 2.3 and 1.3 ng (kg body weight)$^{-1}$ d^{-1} for Anguilliformes, farmed Salmoniformes and Perciformes, respectively, as the fish that result in the top three EDI values. These EDIs correspond to 0.000036%∼0.000013% of the no-observed-adverse-effect level (NOAEL, 10.2 mg (kg body weight)$^{-1}$ derived from the two-generation reproductive toxicity study (Ema et al., 2008), and thus we can conclude that the EDI for the Japanese populace is not a serious amount.

In comparison with other studies carried out around the world, as shown in Table 2, the top three EDIs of our study (3.7, 2.3 and 1.3 ng (kg body weight)$^{-1}$ d^{-1}) were within the range of another Japanese EDI derived from oysters and mussels (Ueno et al., 2010). Furthermore, the EDIs of this study were below the EDI of the United Kingdom (Driffield et al., 2008; Fernandes et al., 2008) (5.9–7.9 ng (kg body weight)$^{-1}$ d^{-1}) and comparable to that of Sweden and the Netherlands (1.9/2.15 and 1.5–2.9 ng (kg body weight)$^{-1}$ d^{-1}) (Lind et al., 2002; de Winter-Sorkina et al., 2003), however, the EDI of Japan was above that of the United States (Schecter et al., 2010), Norway (Knutsen et al., 2008), Belgium (Roosens et al., 2009) and China (Shi et al., 2009) (0.50, 0.2/0.3, 0.09/0.12 and 0.432 ng (kg body weight)$^{-1}$ d^{-1}, respectively). Here, these intakes should be carefully interpreted, because there are differences in the samples from which intakes were estimated and calculation methods as shown in Table 2. In particular, daily intakes were calculated using concentrations in fish in the four studies of Japan, Netherlands and Belgium and the rest were calculated using total diet samples or wide range of food items. Ideally speaking, dietary intakes should be estimated using total diet samples; accordingly, this work is now underway in our laboratory. Because HBCDs are hydrophobic like PCBs, fish with high biomagnifications factors are considered very important among all foodstuffs as the source of human exposure to HBCDs, particularly for Japan, it will be allowed to consider intake from fish as provisional daily intake until total diet study is completed. However, when EDI is calculated using concentrations of highly contaminated fish species, sometimes it may exceed EDI derived using total diet samples. Therefore, it will be better that provisional intakes from fish seen in Table 2, which is Japanese intake from Anguilliformes (eel) in this study and Belgian intake from eel (Roosens et al., 2010) should be considered as the possible maximum estimates. In the studies of Sweden and Norway, the intake from fish was reported to be predominant. However, it was reported that as a HBCD contributor to EDI, meat and dairy food were important for the populations of USA and the other European countries. For China, meat and meat products were also important, except for Shanghai City, where is supposed to consume much fish as same as Japan. The daily intake from fish was recently reported to be 0.12 ng (kg body weight)$^{-1}$ d^{-1} in a Dutch study (van Leeuwen and de Boer, 2008). Taken together with the results of another Dutch study (de Winter-Sorkina et al., 2003), the intake from fish would account for at most 4% of the total EDI for the Dutch people. In the studies of UK and USA, the intake from fish accounted for at most ca.10% (FSA, 2006; Schecter et al., 2010). Thus, excluding several countries, the contribution of fish to total EDI may be now small for ΣHBCDs, compared with that for ΣPCBs (51%) (Schecter et al., 2001). However, if the consumption of HBCD products increases or continues hereafter, fish may become a bigger contributor in food for the intake of ΣHBCDs as same as the case for ΣPCBs.

In summary, from this study, it was suggested that the EDI of ΣHBCDs by the Japanese people is higher compared with foreign peoples, because fish is a favorite foodstuff for the Japanese people, however, it is not yet especially serious level right now, judged from the value of LOAEL for HBCD.

4. Conclusion

The HBCD isomer-specific monitoring of food is so far insufficient and therefore is needed now in order to assess the status of HBCD pollution accurately (Covaci et al., 2006; Kakimoto et al., 2008a). Through this study, for the first time it was discovered that ubiquitous HBCD (mainly α-HBCD) pollution exists in various types of fish collected at markets near the coast of four regions of Japan, and it was also suggested that the concentrations and HBCD isomer profiles of fish would differ depending on how much prey associated with HBCDs in each region is available and what species fish is. In some fish samples, extremely high ΣHBCDs concentrations with the γ-HBCD isomer dominant were observed, suggesting strongly influence by HBCD discharged from industrial plants. Moreover, the daily intakes in Japan from fish of the three categories were calculated to be higher in this study, compared with the reported dietary daily intake in several other countries. It has been recently reported that dust can be a greater source of exposure to HBCDs like polybrominated diphenyl ether (Roosens et al., 2009), we also should pay attention to such other sources. However, when considering the frequency and probability of exposure, the importance of food as an exposure source remains. From the viewpoint of prevention of adverse health effects due to unintended body burden of HBCDs detected in fisherman's serum (Weiss et al., 2006) and human milk (Johnson-Restrepo et al., 2008; Kakimoto et al., 2008b), it will be necessary, as long as HBCD products are used, to monitor the time course variation (trend) of the contents of HBCD pollutants in food, especially in fish, which is well-known to accumulate and magnify chemical pollutants via the food chain.

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