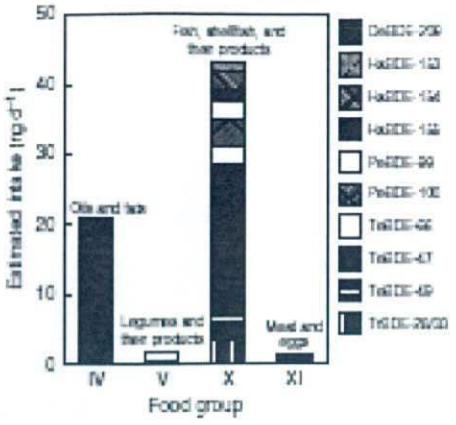

Figure 2. Estimated daily intakes of PBDEs from foods. Results were calculated by assuming the non-detected values as zero.

(88 ng day^{-1}) (Schecter et al. 2006), Spain (82 ng day^{-1}) (Bocio et al. 2003), Japan (68 ng day^{-1}) (Ashizuka et al. 2004), Finland (43 ng day^{-1}) (Kiviranta et al. 2004), Belgium (23 ng day^{-1}) (Voorspoels et al. 2007), and the Netherlands (13 ng day^{-1}) (De Winter-Sorkina et al. 2003).

Figure 3 reveals the estimated dietary intakes of pentaBDE and decaBDE by different age groups in terms of ng kg b.w.$^{-1}$ day^{-1} (ND = 0). The average body weights of the nine different age groups, namely, 1–6, 7–14, 15–19, 20–29, 30–39, 40–49, 50–59, 60–69, and ≥70 years, were 16, 36, 56, 58, 61, 62, 59, 58, and 53 kg, respectively. The intakes per kg body weight of pentaBDE were relatively high in the senior groups of 60–69 years (1.1 ng kg b.w.$^{-1}$ day^{-1}) and ≥70 years (0.98 ng kg b.w.$^{-1}$ day^{-1}) as well as in the infant group of 1–6 years (1.4 ng kg b.w.$^{-1}$ day^{-1}). In contrast, the intakes per kg body weight of decaBDE were clearly age dependent, with the highest and lowest values detected in the infant (1–6 years) (0.90 ng kg b.w.$^{-1}$ day^{-1}) and senior groups (≥70 years) (0.24 ng kg b.w.$^{-1}$ day^{-1}), respectively. The relatively high intakes per kg body weight of pentaBDE and decaBDE in the infant (1–6 years) group were attributed to the low body weights of infants, and these results are consistent with those reported by Bocio et al. (2003) and Schecter et al. (2006). The difference between the age-related intake trends of pentaBDE and decaBDE was mainly attributed to the difference in the dietary habits between each age group. The senior adults (60–69 and ≥70 years) tended to have higher intakes of 'fish, shellfish, and their products' but lower intakes of 'oils and fats' than young (20–29 and 30–39 years) and middle-aged adults (40–49 and 50–59 years). For example, the average daily intakes of 'fish, shellfish, and their products' in the six different age groups of 20–29, 29–39, 40–49, 50–59, 60–69, and ≥70 years were 75.2, 77.6, 94.8, 78.2, 116, and 97.9 g day^{-1}, respectively. For 'oils and fats', the average daily intakes in these six different age groups were 13.7, 13.0, 12.0, 11.0, 9.1, and 7.0 g day^{-1}, respectively (Ministry of Health, Labour and Welfare, Japan 2003, 2004, and 2006). 'Fish, shellfish, and their products' and 'oils and fats' were the main sources of pentaBDE and decaBDE, respectively (Figure 2). Hence, higher intake of the former and lower intake of the latter led to the higher intake of pentaBDE and lower intake of decaBDE in the senior adults than in the young and middle-aged adults.

Regarding the group showing the highest PBDE intake, i.e. the 1–6-year-old infants, the lower, middle, and upper bounds of the intakes per kg body weight were calculated as 0.0014, 0.013, and 0.024 μg kg b.w.$^{-1}$ day^{-1} for pentaBDE, and 0.0009, 0.012, and 0.024 μg kg b.w.$^{-1}$ day^{-1} for decaBDE, respectively. In addition, for an average person (54 kg b.w.), the lower, middle, and upper bounds of the intakes per kg body weight were calculated as 0.0009, 0.006, and 0.011 μg kg b.w.$^{-1}$ day^{-1} for pentaBDE, and 0.0004, 0.006, and 0.011 μg kg b.w.$^{-1}$ day^{-1} for decaBDE, respectively. These values were two to five orders of magnitude lower than the reference doses of penta- and decaBDE (2 and 10 μg kg b.w.$^{-1}$ day^{-1}, respectively), both of which were proposed by the US Environmental Protection Agency (n.d. a, c). These results suggested that the current status of dietary exposure to PBDEs was not likely to cause adverse health effects, and the risk was judged acceptable in Japan as well as in the other reported countries.

PentaBDE constituents such as TeBDE-47 and PeBDE-99 were dominant in food groups V, X, and XI. This is substantially in accordance with a previous TDS study by Ashizuka et al. (2004). Ashizuka et al. did not study the levels of OcBDEs, NoBDEs, and DeBDE in their TDS samples. In the present study, a high proportion of DeBDE-209 was observed in the group IV food samples, which mainly consisted of vegetable oils (Figure 2). Although a variety of vegetable oils are sold in the market, the status of PBDE contamination in them remains unclear. To the best of our knowledge there has only been one report regarding the PBDE levels in individual vegetable oil samples (Jacobs et al. 2004). Jacobs et al. studied the PBDE levels (#28, #47, #99, #100, #153, #154, and #183) in one blended vegetable oil and three linseed oil supplements and found no PBDE residues in these samples. There have been several reports about the PBDE levels in some 'oils and fats' composite food samples (Bocio et al. 2003, Ashizuka et al. 2004, Darnerud et al. 2006, UK Food Standards Agency 2006). The UK Food Standards Agency (2006) has reported the concentration of DeBDE-209, which was the most dominant congener, in the composite sample of oils and fats collected in 2003 to be 0.29 ng g^{-1}. However, there have been no reports regarding the

Table 4 Comparison of estimated dietary intake of PBDEs in different countries.

Country	Daily intake per capita (ng d^{-1})			Sampling year	Target congeners	Sampling method and food categories	Subject population	Reference
	Lower bound	Middle bound	Upper bound					
UK	91	NA	117	1999–2000	47, 99, 100, 153, and 154	Duplicate diet: omnivorous diet (n = 10)	Average of 10 individuals	Harrad et al. 2004
USA	88	NA	NA	2003–2004	17, 28, 47, 66, 77, 85, 99, 100, 138, 153, 154, 183, and 209	Market basket: dairy, meat, fish, eggs, and fat products	Typical adult male: 20–39 years, 70 kg-bw	Schecter et al. 2006
Spain	82	97	NA	2010	Te-OctBDEs	Total diet: 54 composite samples (11 food groups)	Typical adult male: 70 kg-bw	Bocio et al. 2003
Japan	NA	94	NA	1995	47, 99, 100, and 153	Duplicate diet (n = 39)	Adult female: average 50.5 years	Wada et al. 2005
Japan	68	NA	NA	NA	47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, and 183	Duplicate diet (n = 6)	6 individuals: age, gender, and body weight were not specified	Ashizuka et al. 2004
Finland	43	NA	45	1997–1999	47, 99, 100, 153, and 154	Total diet: 10 composite samples	Average of 2862 individuals: 24–64 years, 76 kg-bw, gender was not specified	Kiviranta et al. 2004
Sweden	NA	51	NA	1999	47, 99, 100, 153, and 154	Market basket: 6 composite samples of fish, meat, dairy products, eggs, fats/oils, pastry	Average of male and female: 17–79 years, 73.7 kg-bw	Darnerud et al. 2001
Belgium	23	35	48	2005	28, 47, 99, 100, 153, 154, and 183	Market basket: fish, seafood, meat products, eggs, dairy products, fast food	Average person	Voorspoels et al. 2007
The Netherlands	13	NA	213	NA	28, 47, 99, 100, 153, and 154	Single food (n = 91): dairy, dairy products, eggs, meat, poultry, animal fats, fish, and vegetable oil	Average of 6250 individuals: 65.8 kg-bw	de Winter-Sorkina et al. 2003
Japan	46	330	610	2006	PentaBDE (26 Tr-HpBDE congeners)	Total diet: 14 composite samples	Average of 4011 males and females: 54 kg-bw, include all age groups except 0-year-old	This study
Japan	21	310	600	2006	DecaBDE(206, 207, 208, and 209)	Total diet: 14 composite samples	Average of 4011 males and females: 54 kg-bw, include all age groups except 0-year-old	This study

Notes: Lower-, middle-, and upper-bound intakes were estimated by assuming the nondetected values as zero, one half of the limit of detection, and the limit of detection, respectively. Abbreviations: NA, not available.

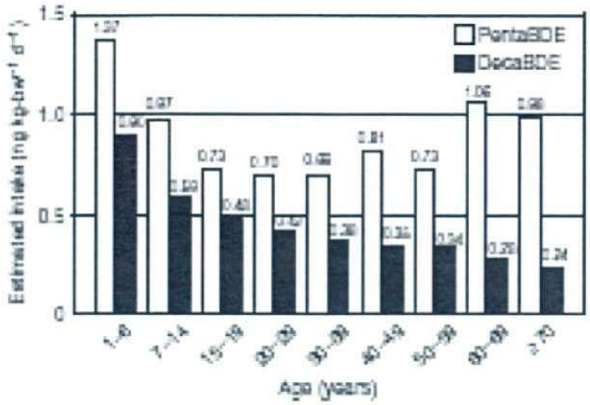

Figure 3. Estimated daily intakes of PBDEs from foods by different age groups. Results were calculated by assuming the non-detected values as zero. Intakes from breast milk and formula were not included.

concentration of DeBDE-209 in individual vegetable oils.

To confirm the presence of DeBDE-209 in vegetable oils, we performed an additional analysis using individual oil samples obtained from rapeseed, corn, safflower, sesame, olive, and soybean (Table 2). It was observed that seven out of the 18 oil samples contained DeBDE-209 as a major or secondary dominant congener at approximately ppb levels (0.7–2.6 ng g^{-1}; Table 5). These results partially explained the reason for the high proportion of DeBDE-209 found in the group IV food samples. Sample #2 was the most contaminated rapeseed oil, and it contained TeBDE-47, PeBDE-99, and DeBDE-209 at concentrations of 0.59, 1.8, and 1.5 ng g^{-1}, respectively. Other minor constituents of PeBDE (#49, #66, #85, #100, #138, #153, and #154) were also detected in sample #2. The results indicated that this vegetable oil sample was contaminated with both pentaBDE and decaBDE.

Samples #2 (rapeseed oil), #9 (corn oil), and #12 (safflower oil) were manufactured by the same company under the Japanese Agricultural Standard; of these three oils, only the #2 rapeseed oil was contaminated with PBDEs. The bottles used for these three oils were identical in shape and were all made from polyethylene terephthalate. Therefore, it is difficult to speculate that the contamination of the rapeseed oil occurred due to the migration of PBDEs from the plastic bottle to the oil. A possible pathway of contamination was the migration of PBDEs from flame-retarded components that could have been used in the oil-manufacturing facilities. If several PBDE-treated components were used in the oil-manufacturing facilities, a part of the PBDEs could have migrated from these components to oil products during oil extraction and refining. The difference between the contamination levels among these oil products could be explained by the vast variety of oil-manufacturing procedures/apparatuses used for these products. Another possible pathway of oil contamination involved the absorption and adsorption of PBDEs by the original farm plants during their growth processes. Mueller et al. reported that both radish (*Raphanus sativus* L.) and summer squash (*Cucurbita pepo* L.) absorbed pentaBDE from contaminated soil in a model experiment (Mueller et al. 2006). Thus, farm plants probably absorb a part of the PBDEs from contaminated soil. Hale et al. (2001) reported that eleven biosolid fertilizer (recycled sewage sludge) samples that were collected from different regions in the USA all contained high concentrations of penta- and decaBDEs (1800–2290 and 84.8–4890 ng g^{-1} dry weight, respectively) (Hale et al. 2001). The land application of biosolids may increase PBDE levels in farm plants and their products. However, the relationship between PBDE levels in plants and those in the soil has remained unclear. Further research is needed to reveal the pathways of oil contamination and to examine the absorption and adsorption of PBDEs by farm plants.

The results raise another concern regarding the behaviour of PBDEs in vegetable oil at high temperatures. It is known that considerable amounts of highly toxic polybrominated dibenzo-*p*-dioxins/dibenzofurans (PBDDs/PBDFs) can be formed from PBDEs under thermal stress conditions (Ebert and Bahadir 2003). In addition, it is expected that certain food-related stores/companies (e.g. fast food restaurants, food processing factories, etc.) continuously re-use heated vegetable oils for long periods. However, information regarding the behaviour of PBDEs in vegetable oils at high temperatures is lacking. Further studies are required to examine the formation of toxic PBDDs/PBDFs from PBDEs in heated vegetable oils under specific cooking conditions.

e 5. Concentrations of PBDEs in vegetable oil samples (ng g^{-1}).

ple	TeBDE-49	TeBDE-47	TeBDE-66	PeBDE-100	PeBDE-99	PeBDE-85	HxBDE-154	HxBDE-153	HxBDE-138	NoBDE-208	NoBDE-207	NoBDE-206	DeBDE-209	Total PBDEs (sum of 36 congeners)
	ND (0.01)	0.07	ND (0.01)	0.02	0.14	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.05)	0.14	0.08	2.4	2.9
	0.01	0.59	0.01	0.19	1.8	0.06	0.10	0.13	0.02	ND (0.05)	0.12	0.08	1.5	4.6
	ND (0.01)	0.15	ND (0.01)	0.04	0.45	0.02	ND (0.01)	ND (0.01)	ND (0.01)	0.06	0.16	0.09	2.6	3.6
	ND (0.01)	0.06	ND (0.01)	0.01	0.13	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.05)	0.11	0.05	1.8	2.2
	ND (0.01)	0.03	ND (0.01)	0.01	0.06	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.05)	0.12	0.05	1.9	2.2
	ND (0.01)	0.03	ND (0.01)	ND (0.01)	0.04	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.05)	0.09	0.05	1.3	1.5
	ND (0.01)	0.01	ND (0.01)	ND (0.01)	0.03	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.01)	ND (0.05)	0.06	ND (0.05)	0.7	0.8

s: Only congeners detected in at least one sample are shown in the table.
Bs were not detected in samples nos. #5, #6, #8, #9, #10, #11, #12, #13, #14, #15, and #17.
concentrations of total PBDEs were calculated by assuming nondetected values as zero.
reviations: ND, not detected. Limits of detection are in parentheses.

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