Assessment of human body burdens of PBDEs at e-waste recycling sites in Taizhou, China utilizing a one-compartment pharmacokinetic model

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Introduction

The release of polybrominated diphenyl ethers (PBDEs) during poorly controlled e-waste recycling causes significant contamination of the surrounding environment, including animals that form a significant part of the diet of local people(1). High daily PBDE intakes are of concern because these contaminants may cause adverse health effects in humans, particularly in children, including adverse growth and development effects, immunotoxcity, genotoxicity, and endocrine system disruption. The current study utilized estimated dietary PBDE intakes (based on measured PBDE concentrations in locally-sourced foods) and applied a one-compartment pharmacokinetic (PK) model to predict the body burden of PBDEs in adults and children from e-waste recycling areas in Taizhou, China, and compared these estimates with previously reported human body burdens in these locations.

Materials and methods

PBDEs body burdens were estimated as previously reported(2) using the following equation:

$$C_{BDE} = \frac{D_{BDE} \times ABS_{BDE}}{k_{BDE} \times BL}$$

Where: C_{BDE} is the lipid-based concentration of the specific PBDE congener (ng/g lw); D_{BDE} is the daily intake of the specific congener (ng/day) via diet or dust; ABS_{BDE} is the absorption fraction; BL is the body lipid mass (g); and k_{BDE} is the congener-specific first order dissipation rate in the body (day^{-1}). Daily adult BDE-209 intake via ingestion of dust was estimated based on concentration data from Ma et al.(3) and dust consumption rates of 0.03 g/day(4). Daily consumption rates for various food products considered in our study are summarised in Table 1. Data on body weight and lipid content in Chinese adults(5) and Chinese children(6) were taken from studies reporting body composition of corresponding cohorts. Human half-life values of most BDEs were taken from previously published estimates(7, 8) while medians of available values for lower- and higher brominated congeners were assigned for those corresponding BDEs with unknown half-lives (i.e. BDE-66, 138, and 197).

Results and discussion

Predicted adult PBDE body burdens were, in general, lower than those calculated for children (Table 2). The comparison among gender groups revealed that adult females (20 – 26 years old) had noticeably lower predicted concentrations for most congeners than males of the same age, while for 5- to 11-year old children, no such gender-related differences were apparent. This is consistent with previous studies that have also reported little or no significant gender-related differences in PBDE body burdens(9–11). For all age-groups, BDE-153 and BDE-47 were the dominant congeners in predicted PBDE body burdens (accounting, on average, for 56% and 20%, respectively, of the ∑PBDE), followed by BDE-99 and BDE-154.

Table 1 Published data and data used in this study on daily consumption rates for food products.

Food type	Daily consumption rate, g/day			
	Published data		Daily rate used in this study	
	Adult	Child	Adult	Child
Fish & shrimp	-	40 – 50[12]		
freshwater fish	25[13]	-	25	20
shrimp	22[13]	-	22	20
Poultry & meat	-	30 - 40[12]		
pork	30[13]	-	30	10
chicken	16[13]	-	16	10
duck	12[13]	-	12	10
Liver	11[13]	-		
chicken liver	-	-	5.5	5
duck liver	-	-	5.5	5
Egg	60[12]			
chicken egg	-	-	30	30
duck egg	-	-	30	30
Culinary oils	25 - 30[12]		30	30

Table 2 Predicted PBDE body burdens (ng/g lw) using PK model

PBDEs	Predicted PBDEs body burden, ng/g, lw							
	Adult				Child			
	median		Range		median		Range	
	male	female	male	Female	boy	girl	boy	Girl
BDE-47	39.6	33.4	4.16 - 204	3.51 – 171	59.6	61.6	5.75 – 301	5.94 – 311
BDE-66	0.50	0.43	0.03 - 8.55	0.02 - 7.21	0.78	0.81	0.05 - 12.9	0.05 - 13.3
BDE-100	4.12	3.47	0.36 - 21.2	0.30 - 17.8	6.12	6.33	0.5 - 30.6	0.52 - 31.7
BDE-99	19.4	16.4	4.07 - 131	3.43 – 110	31.8	32.7	6.90 – 212	7.13 – 219
BDE-85	0.16	0.13	0.04 - 3.75	0.04 - 3.16	0.25	0.26	0.07 – 6.40	0.07 - 6.61
BDE-154	12.5	10.5	1.84 - 42.0	1.55 - 35.4	19.5	20.1	2.83 - 61.3	2.92 - 63.4
BDE-153	105	88.3	7.59 - 707	6.40 – 596	176	182	12.6 – 1000	13.0 – 1030
BDE-138	3.72	3.14	0.05 - 11.3	0.04 - 9.52	6.35	6.56	0.08 - 17.4	0.08 - 18.0
BDE-183	3.82	3.22	0.19 - 66.1	0.16 - 55.7	6.55	6.76	0.31 - 97.9	0.32 – 101
BDE-197	0.51	0.43	0.02 - 10.2	0.02 - 8.57	0.86	0.88	0.03 - 15.4	0.03 - 15.9
BDE-207	0.73	0.61	0.03 - 6.12	0.03 - 5.16	1.22	1.26	0.05 -9.05	0.05 - 9.35
BDE-209	0.70	0.59	0.04 - 5.65	0.03 - 4.76	1.14	1.17	0.06 -8.62	0.06 - 8.90

Predicted body burdens of BDE-197, BDE-207 and BDE-209 were, in general, lower than those estimated for BDEs with lower bromination level. Comparison of predicted PBDE body burdens with reported measured PBDE concentrations in adult's blood (Table 3) strongly suggests that, other than for BDE-209, dietary intake is one of the most important sources of human exposure to PBDEs at informal e-waste recycling areas. Predicted median body burdens of 7 out of 8 BDE congeners included in the comparison (Table 3) fell well within the range of those reported for the Wenling cohort. For BDE-209, predicted concentration was far below those previously measured in adult blood. The comparison of predicted *vs* measured PBDEs concentration in adult blood for the Luqiao cohort showed a similar pattern, though in this case,

our predicted BDE-154 and BDE-153 median concentrations exceeded maximum values measured in adults.
In the case of children, our median predicted body burdens of all BDE congeners exceeded those reported in children's blood in Luqiao (pooled samples) and were also higher than the corresponding maximum values (Table 3).

Table 3 Comparison of predicted vs measured PBDE body burdens (ng/g lw) in blood of adults from Luqiao and Wenling e-waste sites, and of children from Luqiao site, Taizhou, China. n/a – not analysed

PBDEs	Adult					Child		
	This study* (predicted)	**Luqiao** [14]		**Wenling** [14]		**This study* (predicted)**	**Luqiao** [15]	
	median	range	median	range	median	median	range	median
BDE-47	36.5	1.22 – 51.2	3.64	2.88 – 222	25.3	60.6	0.66 – 5.51	4.60
BDE-100	3.80	0.08 – 4.98	0.66	0.64 – 21.2	3.12	6.23	0.16 – 4.42	2.86
BDE-99	17.9	0.44 – 17.8	1.06	1.14 – 23.0	5.74	32.3	0.54 – 17.1	13.8
BDE-154	11.5	0.14 – 6.12	0.78	0.54 – 12.6	1.70	19.8	0.23 – 5.37	1.39
BDE-153	96.7	2.38 – 68.5	10.5	8.40 – 119	21.9	179	2.10 – 8.04	3.43
BDE-138	3.43	n/a	n/a	n/a	n/a	6.45	0.12 – 12.1	0.60
BDE-183	3.52	1.00 – 11.6	3.44	2.60 – 22.8	6.72	6.66	0.83 – 2.40	1.45
BDE-209	0.65	17.2 – 378	64.5	84.5 – 556	172	1.16	n/a	n/a

* - average value for males and females

In summary, there are two main discrepancies between predicted & measured PBDE body burdens:

- Substantially lower predicted BDE-209 concentrations than measured in adult blood;
- Substantially higher predicted concentrations of BDE-47, 99, 100, 138, 153, 154 and 183 than those measured in children's blood.

To assess the possible contribution of BDE-209 from sources additional to diet that may explain the high concentrations of this congener in human blood, we calculated the contribution of dust ingestion to predicted BDE-209 body burdens as there are a number of studies reporting the importance of this pathway of human exposure to BDE-209[16–19]. However, even when additional exposure via dust ingestion was incorporated into our PK model, our median predicted BDE-209 body burdens (1.01, 0.85, 2.23, and 2.30 ng/g lw for male, female, boy and girl, respectively) still fell short of measured values reported in adults.
We proffer the following possible reasons for these discrepancies between predicted and observed body burdens:

- Scatter in published estimates of PBDE half-lives that can result in a wide range of predicted values of body burdens, and absence of half-life values for some congeners;
- Limitations of steady state modelling applied to dynamic processes taking place at e-waste recycling areas, which may result in misrepresentation of actual patterns of exposure to some congeners (e.g., BDE-209). We hypothesise that lower brominated congeners that have longer half-lives are influenced less by the application of a steady state PK model than those with shorter half-lives (e.g., BDE-209); more complex PK modelling would be beneficial for less persistent congeners;
- Absence of data on differences in preferential metabolic pathways and accumulation of PBDEs between different human populations;

- With respect to comparison of predicted *vs* measured PBDE concentrations in children's blood, the fact that Shen et al. (2010) reported results in pooled (7 pools of 21 individual samples) rather than in individual samples, would reduce the range of values and may also affect their median values. Furthermore, another study conducted in children's blood from Luqiao[20] reported the ∑PBDE concentrations to be 664.28 ± 262.38 ng/g lw, which is over 20 times higher than that reported by Shen et al. (2010) at 32 ± 18 ng/g lw for the same range of BDE congeners. Unfortunately, Han et al. (2011) did not provide congener-specific BDE concentrations, but we note that our median estimated ∑PBDE body burden of 312 ng/g lw matches much more closely to the value reported by Han et al. (2011).
- We cannot be certain that PBDE absorption, accumulation, metabolic and elimination processes in children, and therefore biological half-lives, are the same as those in adults. It is known for some pharmaceuticals that anatomical, physiological and biochemical changes through childhood can affect pharmacokinetics/pharmacodynamics and hence the bioavailability of drugs[21], but we do not know if such differences are significant also for contaminants such as PBDEs.
- Limited availability of monitoring data for PBDEs in food and in blood of representative e-waste-impacted people, and uncertainty about dietary composition and daily intakes. For example, we do not know what dietary intakes were for children whose blood was analysed for PBDEs by Shen et al. (2010). Hence it is difficult to make a definite conclusion on differences between predicted and measured values in this case. Nevertheless, the fact that our diet-based predictions for most PBDE body burdens agree well with those reported by others in blood of e-waste-impacted adult population gives some confidence in the validity of our assumed dietary composition as outlined in Table 1.

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