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Serum elimination half-lives adjusted for ongoing exposure of tri-to hexabrominated diphenyl ethers: Determined in persons moving from North America to Australia

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Abstract

The objective of the study was to determine the human serum elimination half-life of polybrominated diphenyl ethers (PBDEs) adjusted for ongoing exposure in subjects moving from a higher exposure region (North America) to a lower exposure region (Australia).

The study population was comprised of exchange students and long-term visitors from North America moving to Brisbane, Australia (N = 27) and local residents (N = 23) who were followed by repeated serum sampling every other month. The local residents were sampled to adjust for ongoing exposure in Australia. Only one visitor remained in Australia for a period of time similar to the elimination half-life and had a sufficiently high initial concentration of PBDEs to derive a half-life. This visitor arrived in Australia in March of 2011 and remained in the country for 1.5 years. Since the magnitude of PBDE exposure is lower in Australia than in North America we observed an apparent 1st order elimination curve over time from which we have estimated the serum elimination half-lives for BDE28, BDE47, BDE99, BDE100, and BDE153 to be 0.942,

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CRediT authorship contribution statement

Andreas Sjödin: Conceptualization, Formal analysis, Investigation, Methodology, Project administration, Supervision, Validation, Visualization, Writing-original draft, Writing-review & editing. **Jochen F. Mueller:** Conceptualization, Supervision. **Richard Jones:** Formal analysis. **Andre Schütze:** Methodology. **Lee-Yang Wong:** Methodology. **Samuel P. Caudill:** Methodology. **Fiona A. Harden:** Supervision. **Thomas F. Webster:** Methodology. **Leisa-Maree Toms:** Project administration, Investigation, Visualization, Writing-original draft, Writing-review & editing.Appendix A. Supplementary data

Disclamer

The findings and conclusions in this report are those of the authors and do not necessarily represent the official position of the Centers for Disease Control and Prevention (CDC). Use of trade names is for identification only and does not imply endorsement by the CDC, the Public Health Service, or the US Department of Health and Human Services.

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1.19, 1.03, 2.16, and 4.12 years, respectively. Uncertainty in the estimates were estimated using a Monte Carlo simulation. The human serum elimination half-life adjusted for ongoing exposure can allow us to assess the effectiveness and reduction in exposure in the general population following phase out of commercial penta-and octaBDE in 2004 in the United States.

Keywords

PBDE; Polybrominated diphenyl ether; Half-Life; United States; Australia

1. Introduction

Polybrominated diphenyl ethers (PBDEs) were used as flame-retardants in consumer articles in North America until they were phased out from the U.S. market in 2004 (commercial pentaBDE and octaBDE) and 2013 (decaBDE) (U.S. Environmental Protection Agency: Washington and D., 2017). These chemicals were phased out due to observed health concerns that included among others learning disabilities in mice (Viberg et al., 2002, 2003a, 2003b; Eriksson et al., 2001) and other thyroid-mediated effects (McDonald, 2002). At the time of the phase out from the U.S. market, there were also indications of human health effects in epidemiological investigations and additional concerns were observed after the withdrawal from the U.S. market; including but not limited to lower birth weight (Harley et al., 2011), learning disabilities (Eskenazi et al., 2013; Adgent et al., 2014; Braun et al., 2014), changes in thyroid hormone concentrations (Herbstman et al., 2008; Chevrier et al., 2010, 2011; Vuong et al., 2015) and reduced fecundability (Harley et al., 2010).

However, after the withdrawal from the market, human exposure did not end since this widely used chemical is present in materials manufactured before the ban and in recycled materials which are released into the indoor environment (Stapleton et al., 2005; Sjodin et al., 2008) as well as in the food that we eat (Schecter et al., 2010). In the indoor environment, PBDEs are sorbed to dust particles. This is because PBDEs were used as an additive flame retardant not covalently bound to the polymer backbone allowing migration out of the material over time (Alaee et al., 2003). Commercial pentaBDE containing tri-to hexabrominated congeners were used in numerous applications such as polyurethane foam, in padded furniture such as couches, and in the pad used under wall-to-wall carpets (Alaee et al., 2003). The more highly brominated octaBDE (6–9 bromines per molecule) and decaBDE technical products were used among others in hard plastics and rubber applications for usage in, for example, electronics (Alaee et al., 2003).

Having a reliable and robustly determined elimination half-life is essential for estimating ongoing exposures and internal concentration of a chemical in the general population after production ceased. Unfortunately, available elimination half-lives of lower brominated congeners (tri-to hexaBDEs) are imprecisely determined and potentially biased (Geyer et al., 2002; Wong et al., 2013). On the other hand, the elimination half-lives of hepta-and decaBDE have been determined in electronics recycling workers by monitoring their decline in serum concentration during their vacations from work to range from weeks (decaBDE) to months (heptaBDE) (Thuresson et al., 2006a). The half-life of, for example, 2,2′,4,4′-

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tetraBDE (BDE47), which is the congener commonly measured at the highest concentrations, has been reported to be in the range of 0.4–3 years (Geyer et al., 2002; Wong et al., 2013).

We aim to estimate the serum elimination half-life in persons moving from North America (Canada and the United States) to Australia, which is known to have a lower background exposure level and an average PBDE serum concentration of about one tenth of the United States (Sjödin et al., 2014a; Toms et al., 2018). PBDEs were never manufactured in Australia and import of technical PBDE mixtures were prohibited in 2007 with an exception for manufactured goods (Comonwealth of Australia, 2007). The serum concentration in Australian permanent residents were also determined to enable adjustment of the half-life calculation for ongoing exposure in Australia. The determined elimination half-life can allow us to evaluate the magnitude in exposure reduction in the general population following the phase out of commercial penta-and octaBDE in 2004 in the United States. The PBDE serum concentration has over the past decade been monitored and shown to decrease in the bi-annual National Health and Nutrition Examination Survey (Toms et al., 2009, 2012; Sjödin et al., 2008). However, without knowing the human elimination rate it is not possible to estimate the magnitude in reduction of exposure over time.

2. Materials and methods

We measured the decline in repeated serum samples collected following a move from North America to Brisbane, Australia for deriving a serum elimination half-life. Local residents were also sampled for six months to estimate the general population concentration in Australia to enable adjustment for ongoing exposure of the North American subjects while residing in Australia. Most subjects were exchange students or affiliated with the University hence the sampling period was limited to the University semester corresponding to approximately six months in most cases (Table 1). Non fasting serum was collected.

2.1. Measurement of PBDEs in serum

Serum PBDE concentration was measured using established methods at the Centers for Disease Control and Prevention (Sjödin et al., 2004; Jones et al., 2012). Briefly, serum samples are extracted using liquid-liquid extraction, employing an automated Liquid Handling instrument (Gilson 215 Liquid Handler®, Gilson, Inc.). Required sample pretreatment prior to extraction is performed on the liquid handler, including automated addition of (i) internal standards, (ii) methanol with a manual vortex step in-between each addition. Hydrochloric acid is added manually to denature proteins in the serum enabling efficient extraction of target compounds. During the extraction step the target analytes are transferred from a water medium to an organic solvent.

Removal of co-extracted lipids, is performed by elution (5% DCM in hexane; 10 mL) of the extract through a column containing from the top 0.25 g of silica and 1 g of silica/sulfuric acid (33% by weight). This step is automated using the Rapid Trace® (Caliper Life Sciences).

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Serum concentrations are determined using gas chromatography isotope dilution high resolution mass spectrometry (GC/ IDHRMS), which minimizes or eliminates many interferences associated with low-resolution measurement of organohalogen compounds. Spitless injection is used on a short GC column (DB-5HT; 15 m length, 0.1 mm film thickness, 0.25 mm ID) enabling the determination of high molecular weight compounds such as decabromodiphenyl ether (BDE-209) having a molecular weight close to 1000 amu. Electron impact ionization (EI) is used. The two most abundant ions in the isotopic cluster (fragment or molecular ion) are monitored for the target analyte as well as for the 13C-labeled internal-surrogate standard. Quantification is made against a calibration curve covering the full concentration range of the target analytes.

The typical measurement CV of included quality control samples was approximately 5% and three blind duplicate samples created during the serum collection in Australia did not vary more than −4.4% (BDE47) to 2.7% (BDE153) and were not decoded as blind duplicates until after completion of measurement and reporting of the data back to Australia. A field blank comprised of bovine serum (Sigma Aldrich B8655) previously shown not to contain PBDEs at a level exceeding the limit of the detection of the measurement method (Sjödin et al., 2004; Jones et al., 2012) was included among the unknown samples and blinded to CDC personnel until after the final data had been reported. This field blank was exposed to serum collection supplies (transfer pipette and serum storage vial) and shipped to the CDC labeled as an unknown study specimen. Limit of detection (LOD), ^{13}C-internal standard recovery and CV of quality control samples by PBDE congener is given in Table S1.

2.2. Subjects

We recruited participants among exchange students moving from North America (Canada and the United States) to Brisbane, Australia and persons living in the general area of the city of Brisbane from March 2011 to June 2013. Recruitment was through advertisement at local Universities in Queensland (Queensland University of Technology and The University of Queensland) as well as word of mouth. Inclusion criteria were as follows: the subjects must have lived in the same general location (city and/or state) in their country of origin for at least 5 years with plans to remain in Brisbane, Australia for a minimum 6 months. All participants were required to have blood drawn (25 ml at Sullivan Nicolaides Pathology) every two months for the duration of their stay in Australia or every 6 months for Australian permanent residents. We recruited a total of 27 subjects (Table 1) from North America and 23 subjects from Brisbane Australia. For most participants from North America, the length of stay was 6 months (Table 1), coinciding with the University semester. The longest period of collection for a non-permanent Australian resident was for a Canadian, male for which the first and last sample spanned 547 days. The participants completed a questionnaire capturing basic demographic information: sex, birth country, and years lived in Brisbane (Australians) and years in previous state/province (North Americans) as well as frequency of consuming fish (Table 1). Retention of participants was encouraged by obtaining comprehensive contact information, maintaining monthly contact, provision of a snack pack (chocolate, fruit and juice) and a gift card of $15 Australian dollar equivalent to $10 USD after the phlebotomy.

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We obtained ethics approval from Queensland University of Technology Ethics Committee and The University of Queensland Ethics committee. The Centers for Disease Control and Prevention (CDC) was determined to not be engaged in human subjects' research and were exempt from Internal Review Board review and no personally identifiable information was made available to CDC researchers.

The inclusion criteria for deriving an elimination half-life of PBDEs for a subject were: (*i*) four or more serum samples collected, (*ii*) having an initial serum concentration at least three-fold the average serum concentration in Australia (Fig. 1 and Table S2) with data below the LOD set to zero and (iii) a sample collection period of more than one year corresponding to the approximate half-life of most PBDE congeners investigated except BDE153. Unfortunately, only one North American subject met the inclusion criteria. That subject had a total of 6 serum draws during a period of 1.5 years and an initial BDE47 concentration of 34.1 ng/g serum lipid (Figs. 1 and 2).

2.3. Estimation of half-life

The decline in PBDE serum concentration was assumed to follow Equation (1) as presented in Russell et al. (2015) where: t is the time in days since the collection of the initial serum sample after arriving in Australia; $C_{Australia}$ is the steady state concentration for a subject living in Australia; C_0 is the subject's initial serum concentration (initial serum sample collected at $t = 0$), $C(t)$ is the subject's concentration at time t and k_e is the elimination rate constant adjusted for ongoing exposure that is related to the elimination half-life by Equation (2). The underlying assumptions are first order kinetics, a one-compartment model and that the sampled Australian residents represents the Australian steady state serum concentration, e.g. $C_{Australia}$.

$$C(t) = C_{Australia} + (C_0 - C_{Australia})e^{-k_e t} \quad (1)$$

$$t_{1/2} = \ln(2)/k_e \quad (2)$$

2.4. Statistical Approach

Due to the fact that only one subject was determined to fulfill all inclusion criteria for calculation of elimination half-life, we decided to use a Monte Carlo (Harrison, 2010) simulation approach to estimate the coverage interval for the uncertainty in the derived elimination half-lives. The simulation was conducted in SAS Enterprise Guide 7.1 (Cary, NC). In the simulation, the variable $C_{Australia}$ was defined as a random generated number from the natural logarithm distribution derived from the Australian subjects (weighted by the number of samples collected per participant and data below the LOD set to the LOD divided with the square root of two). In each iteration of the Monte Carlo simulation, each data point corresponding to the Australian permanent residents and the North American visitor was multiplied by a random number from a Gaussian distribution with a mean of 1.0 and a standard deviation of 0.05 corresponding to the typical analytical uncertainty of the measurement estimated from included QC samples, i.e., CV of 5%.

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The SAS program iteratively determined the optimum fit for k_e by analyte for 10,000 random generated values of $C_{Australia}$ including random generated analytical measurement uncertainty for each measured concentration. The optimization was performed by first calculating the sum of squared differences between calculated and measured concentrations at each time point for a k_e of 0.0001. There after, k_e was successively incremented by 0.0001 until the sum of squared differences increased relative to the previous iteration. The program then restarts with a high value of k_e of 0.004 and similarly calculates the sum of squared differences for each subtraction of 0.0001 from the last k_e until the sum of squared differences increased from the previous iteration. The values of k_e where the sum of squares increase relative to the previous iteration when adding or subtracting 0.0001 brackets the optimum value of k_e. The procedure above was then repeated by adding and subtracting 0.00001 corresponding to a 10-fold higher resolution between the values of k_e that brackets the optimum fit-value to more precisely determine the optimum value of k_e. There after every value of k_e between the refined upper and lower boundary of k_e was calculated with a resolution of 0.000001. The lowest value of sum of squares obtained was set to be the optimum fit value of k_e. Due to the successively smaller step distance of the iterative process the optimum fit value of k_e could be identified with a minimum of three significant figures in only 82 iterations regardless of the true fit value of k_e located between 0.0001 and 0.004. The SAS program completed in less than 30 h, 10,000 iterations for each of 5 analytes.

The remaining North American subjects that were not included in the half-life calculation with more than one sample collected (n = 20) were used to validate the optimum fit of k_e. This was done by calculating the concentration at each time point $C(t)$ using Equation (1) with the median of all optimum values of k_e, each subject's initial concentration (C_{0i} and with $C_{Australia}$ set to the median Australian concentration by congener. The measured serum concentrations for all subjects with t > 50 days were plotted against the predicted serum concentration (Figs. S5–S9). The least squares coefficient of determination (r^2) for measured concentration vs. calculated from initial concentration were considered the predictability of the determined value of k_e.

Change over time in Australian background concentration ($CAustralia$ in equation (1)) was investigated by a simple linear random effect mixed model where subject is a random effect, and time by year is a fixed effect independent variable.

3. Results

The average concentrations of PBDEs were 2.1 [BDE153] — 7.3 [BDE47] fold higher in North American than in Australian subjects (Fig. 1 and Table S2). North American subjects followed during their stay in Brisbane, Australia had a decreasing concentration while Australian subjects varied in a non-consistent direction during the study period (data not shown). The one subject meeting the inclusion criteria for deriving an elimination half-life had a serum concentration that was 3.5 [BDE153] to 16 fold higher than the average Australian concentration. Concentration over time for this subject who was followed for 1.5 years is given in Fig. 2 and S1–S4. The median Monte Carlo-derived half-lives with 2.5th and 97.5th percentile ranges (Fig. 3) and model parameters are given in Table 2.

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The r^2 for the linear regression of measured vs. predicted serum concentration, including samples from all subjects (except the subject used for deriving the half-life estimate) that were collected more than 50 days after entering Australia was in general high (range: 0.964 [BDE153]-0.989 [BDE28]). Similarly, samples collected 90–180 and more than 180 days after entering Australia fell on the same regression line of measured vs. predicted concentration (Figs. S5–S9).

The concentration of BDE47 in Australian residents was found to decrease with sample collection time with a significant negative slope of −0.800 ng/year (95%CI −0.233 to −1.37) and with an intra class correlation coefficient (ICC) of 0.831. Other investigated congeners did not have a significant association with sample collection time (Table S3). Due to this observation the Monte Carlo simulation was repeated for BDE47 with a variable $C_{\_Australia}$ that decreased with time from middle point of collected samples (Day 273) to the last sample collected (Day 547) and increased from the middle time point back to the first sample collected (Day 0) at a rate of 0.800[ng/ year]/365 = 0.00227 [ng/day]. The determined half-life estimate from this refined calculation did not change more than 0.6% (data not shown); hence, a variable $C_{\_Australia}$ was not used in the presented Monte Carlo simulation for any PBDE congener.

4. Discussion

We observed a significantly higher concentration of PBDEs in North American subjects than in Australians (Fig. 1) consistent with previous reports (Sjödin et al., 2008, 2014a, 2014b; Toms et al., 2009, 2012, 2018). This difference in population concentration enables us to study the decline in serum concentration with length of time in Australia. Directly measured serum elimination half-lives from subjects transitioning from a high to a low exposure situation has been missing in the literature except in the case of the relatively short lived congeners from a study of e-waste recyclers being exposed occupationally to technical Octa- and DecaBDE (Thuresson et al., 2006a). The decline in these workers' serum concentration was measured before and after their summer vacation spanning approximately four weeks and the half-lives of BDE183 and BDE209 were reported to be 3 months and 2 weeks (Thuresson et al., 2006b), respectively. Other studies include population estimates of half-lives from dietary intake with the assumption of steady state concentration or extrapolation from determined half-lives in the rat (Geyer et al., 2002) and population level pharmacokinetic models (Wong et al., 2013). The estimated half-lives using these approaches (Geyer et al., 2002; Wong et al., 2013) ranged between 0.37 and 3.0 years for BDE47; 2.9–8.2 years for BDE99; 1.6–2.9 years for BDE100; and 3.5–12 years for BDE153. Earlier estimates of PBDE half-lives tended to be longer than the estimates we have derived here from a North American visitor to Australia being 1.5–3.5, 2.8–7.0, 0.74–1.6, and 1.6–3.9-fold the half-life of BDE47, BDE99, BDE100, and BDE153, respectively. The initial serum concentration of the North American visitor to Australia was within the upper 75th to 90th percentile of the American population estimated in the National Health and Nutrition Examination Survey (NHANES) and could hence be considered not an unusual concentration in North America but still in the upper range of the distribution (Sjödin et al., 2008).

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In our case, we have used persons moving from a higher exposure location (North America-Canada and United States) to a lower exposure location (Brisbane, Australia) and followed them with repeated sampling during their stay in Australia. Recruitment was done at local Universities in Australia, which limited the recruitment to persons affiliated with the University, i.e., exchange students or employees who mostly stayed in Brisbane for less than 6 months and Australian resident students and employees of the University. Due to the limited sampling period length and the requirement of the North American subjects to have an initial serum concentration higher than the Australian background concentration, we were limited to one subject who had a high initial concentration (Fig. 1) corresponding to between the 75th and 90th percentile concentration of the NHANES 2003/04 survey. This male Canadian subject was available to provide blood samples over a 1.5 year stay in Brisbane Australia corresponding to between 1.6 (BDE28) and 0.4 (BDE153) serum half-lives. The uncertainty in this subject's half-life estimate was derived from a Monte Carlo simulation where the effect on uncertainty in the parameters used to derive the estimated half-life were examined. The results in this study are unique because it is the first estimated half-life adjusted for background exposure in a person from North America. However, we cannot estimate the variability in half-life between different persons other than to conclude that the derived half-life explained the elimination of PBDEs in other subjects studied for a shorter period in Australia (50 to <90 days, 90 to <180 days and 180 to <270 days) as shown in Figs. S5–S9. Unfortunately, it was not possible to increase the minimum number of days of residence in Australia used for this validation of the derived half-life estimate due to the limited number of samples available with longer residence time (Table 1). However, due to the agreement between the measured and extrapolated concentration calculated from an initial concentration upon arriving in Australia it is reasonable to assume that the derived half-life estimate is applicable to other persons and/or populations similar to this study.

Elimination half-life estimates unaffected or adjusted as in our case for ongoing exposures through food, dust ingestion and/or other sources of exposure are an important parameter to study. For example, half-lives are used to assess the effect of regulations intended to decrease and/or eliminate a population's exposure to a pollutant such as PBDEs. In the Unites States the NHANES, which is administered by the National Center for Health Statistics (NCHS), uses measurements performed by the National Center for Environmental Health (NCEH) and others to investigate pollutant trends over years and even decades as in the case of PBDEs, lead and other pollutants of concern.

Technical PBDE mixtures known as Penta-and OctaBDE based on their average bromination degree were voluntarily withdrawn from the United States market in 2004 because of concerns of adverse health outcomes that included, among others, learning disabilities in mice (Viberg et al., 2002, 2003a, 2003b; Eriksson et al., 2001) and other thyroid-mediated effects (McDonald, 2002). The CDC has, since the withdrawal from the market, monitored the serum concentrations in pooled measurements in five biannual surveys covering the time period 2005/06 through 2013/ 14 with later measurements expected. In these data sets BDE47 has decreased by 49%, 53%, 19% and 45% in 12–19, 20–39, 40–59 and subjects over the age of 60 years, respectively. Considering that this period spans approximately ten half-lives, we can conclude that elimination from the Unites States market has decreased

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exposures to these pollutants but not at all eliminated exposures that are clearly ongoing, although at about half the rate compared to the years 2005/06.

Exposure sources of PBDEs to the general population include furniture manufactured prior to the phase out. It is known that furniture typically is used for long periods of time and that it is not uncommon for furniture to have a secondary usage period after being discarded by the first owner. Exposure to PBDEs in the United States population could also result from recycling of polyurethane for use in carpet pads, which may have an even longer usage period than furniture before being discarded. BDE153 has the longest measured half-life of approximately 4 years. Exposure to this congener has shown variability in exposure by age in the NHANES including a decrease by 17% among those 12–19 years old and a 9.9% among those 20–39 years old but an increase by 55% and 38% among those 40–59 years old and over 60 years old respectively (Sjödin et al., 2019). These findings indicate that exposure to PBDEs has not ceased in the United States population; however, routes of exposure and/or sources of exposure have changed.

4.1. Limitations

The limitations of this investigation of elimination half-lives adjusted for ongoing exposure include a lack of subjects with a sufficiently long study period due to subjects being exchange students mostly limited to one semester in Australia. As a result, we were limited to using one subject with a long sampling period of 1.5 years who also had an unusually high initial PBDE serum concentration corresponding to between the 75th and 90th percentile of NHANES 2003/04. However, the derived half-life was consistent with the decreasing concentration in other subjects with a shorter sample collection period (Figs. S5–S9). Other limitations include limited length of time studied especially for BDE153 where subjects were sampled over a period representing only 40% of one half-life. This short sampling period may potentially have introduced bias in the estimate of the half-life of BDE153 or decreased the precision of the estimate but it is clear that BDE153 has a half-life that is 2–4 fold longer than the other tri-to pentaBDE congeners investigated here. Further, all tri-to hexaBDEs investigated here have much longer biological half-lives than BDE183 and BDE209 which are measured to be only weeks or months.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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HIGHLIGHTS

- Calculated human serum elimination half-lives for tri-to hexabrominated diphenyl ethers.
- Sampling period of 1.5 years was more than one elimination half-life for most PBDEs.
- Serum elimination half-life was adjusted for ongoing exposures.
- Estimated serum elimination half-lives were 1–4 years, depending on the congener.

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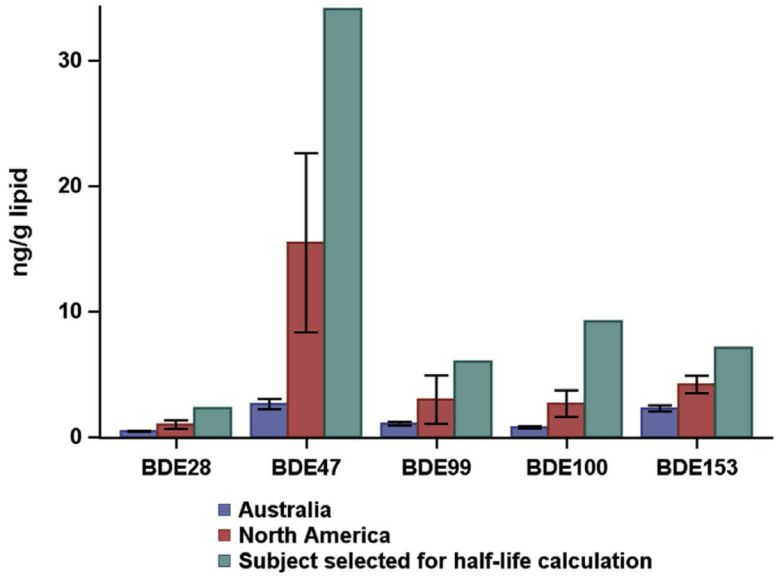

Fig. 1.

Mean and 95% confidence interval (CI) for the Australian subjects (N = 23) weighted by number of samples collected per subject, mean and 95% CI for the first samples collected from North American subjects (N = 27) upon entering Australia and concentration for the single sample selected for determination of elimination half-life adjusted for ongoing exposure.

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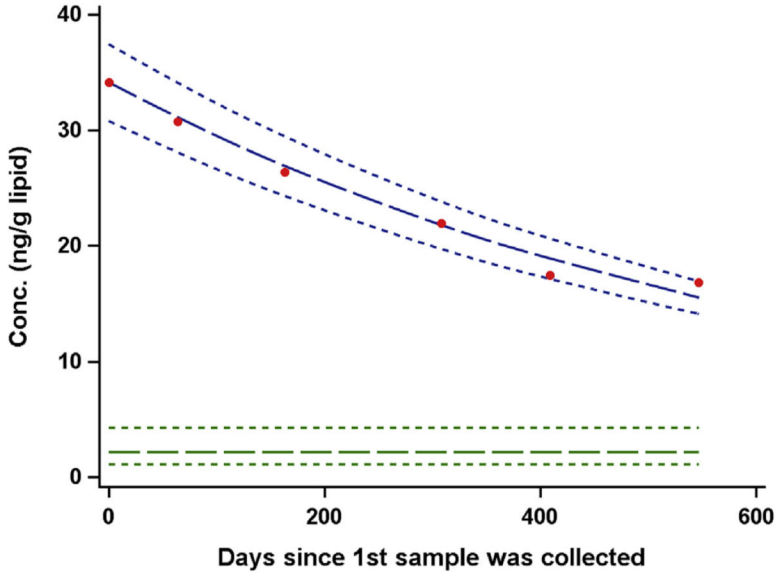

Fig. 2.

Elimination of BDE47 with residence time (1.5 years) in Australia for the single subject selected for half-life determination. Red dots show actual measured concentration and blue lines show median concentration and 2.5th and 97.5th percentiles of concentration estimates from 10,000 iterations (see Materials and methods for underlying assumptions). Green lines show median and 2.5th and 97.5th percentiles of the Australian subjects. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

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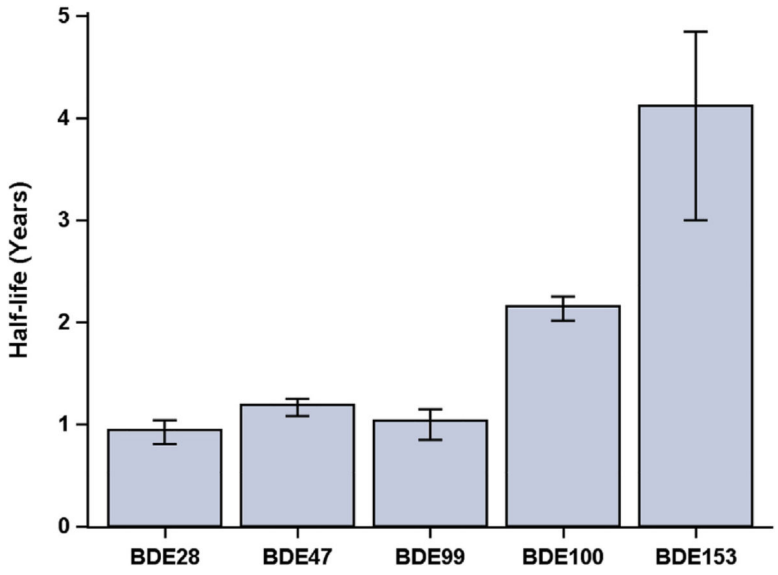

Fig. 3.

Median estimate of elimination half-life adjusted for ongoing exposure for tri-to hexaBDE congeners. Limit bars indicate 2.5th and 97.5th percentile of Monte Carlo estimates.

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Table 1

Summarized questionnaire information, number of samples collected and length of sample collection for Australian and North American subjects.

Measure	Continent of residents	
	North America	Australia
	N = 27	N = 23
	Median (range)	Median (range)
Age (Years):	21 (19–34)	36 (22–57)
Body mass index (kg/m^2):	23.7(17.7–64.8)	25.6 (18.0–32.7)
Length of sampling period (Days):[a]	84(15–547)	312(131–413)
Number of subjects with X samples collected:	**Count**	**Count**
X = 1	3	4
X = 2	7	1
X = 3	14	3
X = 4	2	9
X = 5	0	6
X > 6	1	2
Sex:	**N(%)**	**N(%)**
Female	22(81%)	11 (48%)
Male	5 (19%)	12(52%)
Duration of residence:		
>5 to <10 years	3 (11%)	5 (22%)
>10 years	24 (89%)	18 (78%)
Birth Country:		
Canada	10 (37%)	0
United States	15 (56%)	0
Australia	0	9 (39%)
Other	2(7%)	14(61%)
Frequency of fish and sea food consumption:		
Never Eat	5 (19%)	2(9%)
Less than once per week	14 (52%)	11 (48%)
Once per week	7 (26%)	6 (26%)

Measure	Continent of residents	
	North America	Australia
	N = 27	N = 23
	Median (range)	Median (range)
Twice a week or more	1(4%)	4(17%)

[a]Excluding subjects with only one sample collected.

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Table 2

Serum half-life of polybrominated diphenyl ethers based on one subject with 6 repeated serum measurements spanning over 547 days from a total of 10,000 iterations with median and range of model parameters indicated.

Congener	Half-life (Years)		Model Parameters [Median (range)][a]		
	Median	(P2.5 -P97.5) [b]	C_0 [c]	$C_{australia}$ [d]	Ke [e]
BDE28	0.942	(0.803–1.04)	2.30 (1.88–277)	0.407 (0.224–0.848)	0.00202 (0.00174–0.00331)
BDE47	1.19	(1.08–1.25)	34.1(26.1–42.1)	2.136 (0.615–7.02)	0.00160 (0.00148–0.0021)
BDE99	1.03	(0.845–115)	6.01 (0.629–0.968)	0.916 (0.345–2.64)	0.00184 (0.00157–0.00394)
BDE100	2.16	(201–2.25)	9.18(4.75–7.2)	0.679 (0.274–174)	0.000881 (0.000811–0.00105)
BDE153	4.12	(3–4.84)	7.10(5.6–8.72)	2.016 (0.741–5.16)	0.000461 (0.000362–0.00157)

[a] Detailed definition of model parameters is given under heading “Statistical Approach”.

[b] Coverage interval 2.5th percentile to 97.5th percentile.

[c] Initial concentration of North American subject (ng/g lipid).

[d] Background concentration (ng/g lipid).

[e] Elimination rate constant.

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