Supporting Information

Evaluating the Bioaccessibility of Flame Retardants in House Dust Using an In Vitro Tenax Bead-Assisted Sorptive Physiologically Based Method

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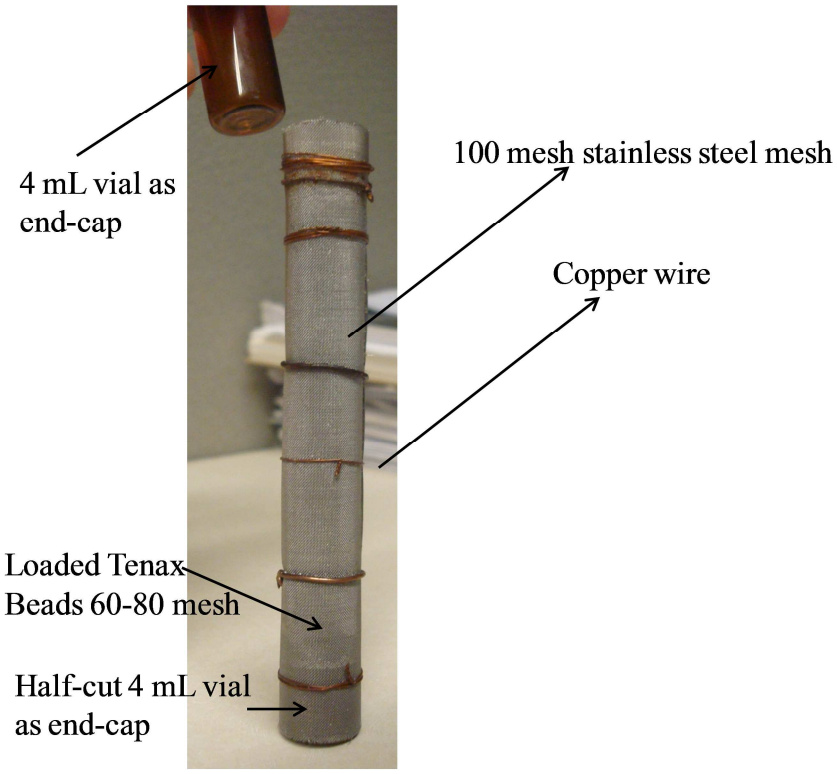

Figure S1. Schematic showing the TA trap that was designed using 100 mesh stainless steel mesh

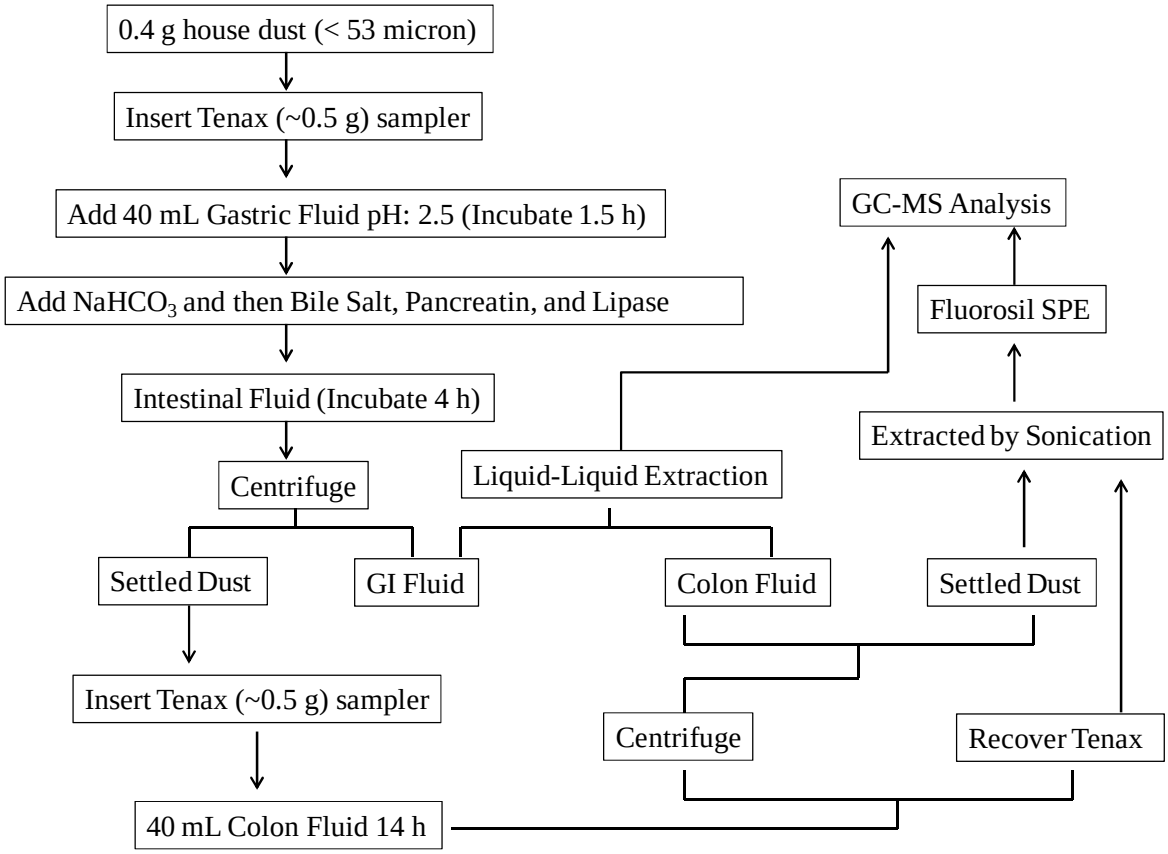

Figure S2. Flow chart displaying the steps involved in the dust sample incubation, cleanup and analysis.

Text S1. Chemical Analysis

The extraction, cleanup and analysis of FRs in the recovered dust samples and TA were modified from our previously published methods[1, 2]. Dust and TA were first extracted with acetone to remove all the water residues, then extracted two times with hexane:acetone (1:1). All extracts were combined. F-BDE 69 was used as an internal standard for tri-nonaBDEs, EH-TBB, and BEH-TEBP, and ^{13}C BDE-209 was used as an internal standard for BDE-209. D-TDCIPP and d-TPHP were used as internal standards for TCEP/TCIPP/TDCIPP and TPHP; respectively. An ENVI-Florisil SPE column (500 mg, 3 mL) was used to clean and purify the dust extracts. The SPE column was first conditioned with 5 mL methanol and rinsed with 3 mL hexane. Then the dust extract (in hexane) was loaded on the SPE using 0.5 mL hexane and 4 mL hexane was used to elute hydrophobic FRs (e.g., PBDEs, EH-TBB, and BEH-TEBP) in fraction one (F1). Subsequently, most OPFRs were eluted in fraction two (F2) using 10 mL ethyl acetate. After evaporation, 13C-CDE-141 and d-TCEP were spiked into each sample to serve as a recovery standard (measure recoveries of internal standards). PBDEs, EH-TBB, and BEH-TEBP were analyzed using gas-chromatography coupled to a mass spectrometry detector (GC-MS, Agilent GC 6890N, MS 5975, Newark, DE) operating in the negative chemical ionization (NCI) mode. OPFRs were analyzed by GC/MS operated in electron ionization (EI) mode. The extraction and analysis of the foam and recovered TA were similar to the dust samples. Due to the high levels of FRs in the foam, foam/TA bead and digestive extracts were diluted 100 times and 20 times; respectively. Surrogate standards were spiked and no further cleanup was performed. To analyze the FRs in the digestive fluid, 20 mL of the digestive fluid was first treated with 6 M HCl to denature the protein and then liquid-liquid extracted with hexane:ethyl acetate (1:1) three times. The extracts were combined and concentrated to 1.0 mL for chemical analysis. The surrogate standard and recovery standard were identical to the standards described above. The analysis of TBBA was performed by liquid-chromatography mass spectrometry (LC-MS/MS) operating in negative electron-spray ionization (ESI-) as described in our previous study[3].

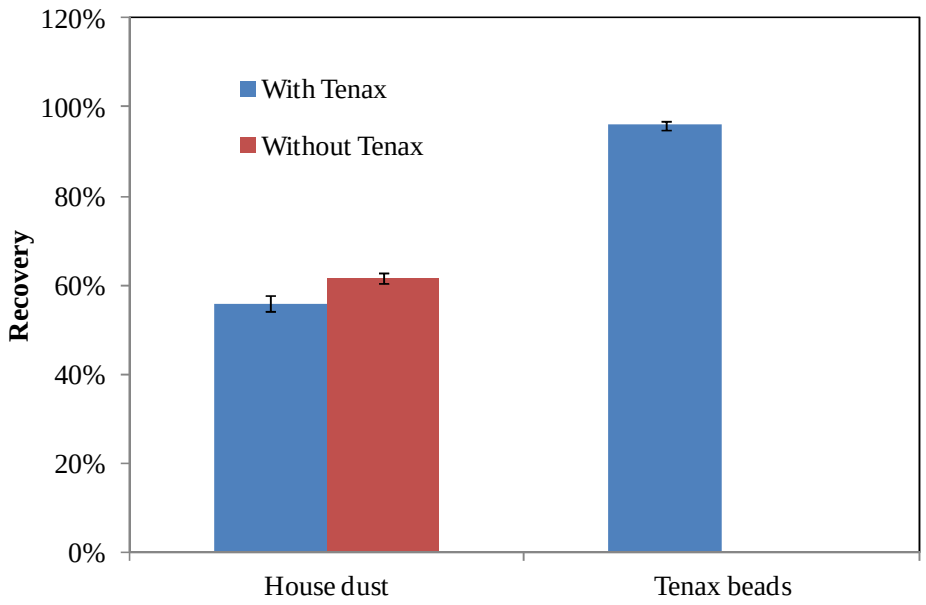

Figure S3. Recovery of TA and SRM2585 (n = 3) relative to the amount added before incubation. The mass of dust recovered without adding TA was run for comparison purposes. Error bar represents the standard deviation of triplicate analyses.

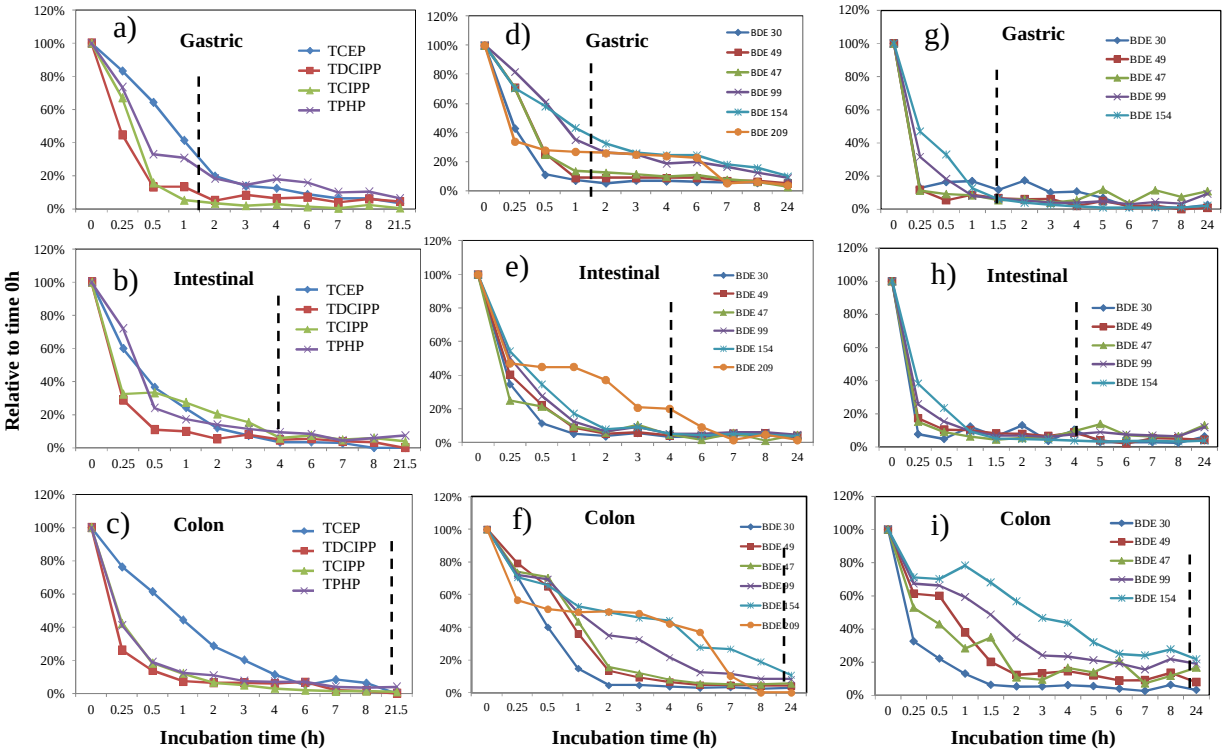

Figure S4. Relative mass of OPFRs (TCEP, TDCIPP, TCIPP, and TPHP) and PBDEs in spiked gastric, small intestinal, and colon fluid relative to Time 0. Figure a-c): OPFR sorption kinetics in the high spike level (2 µg/mL); Figure d-f): PBDE sorption kinetics in the high spike level (2 µg/mL); and Figure g-i): PBDE sorption kinetics in the low spike level (10 ng/mL). BDE209 and OPFR sorption kinetics in low spike level were not shown (<MDL). Dashed lines indicate the incubation times in stomach (t = 1.5 h), small intestine (t = 4 h), and colon (t ~ 16 h) fluid.

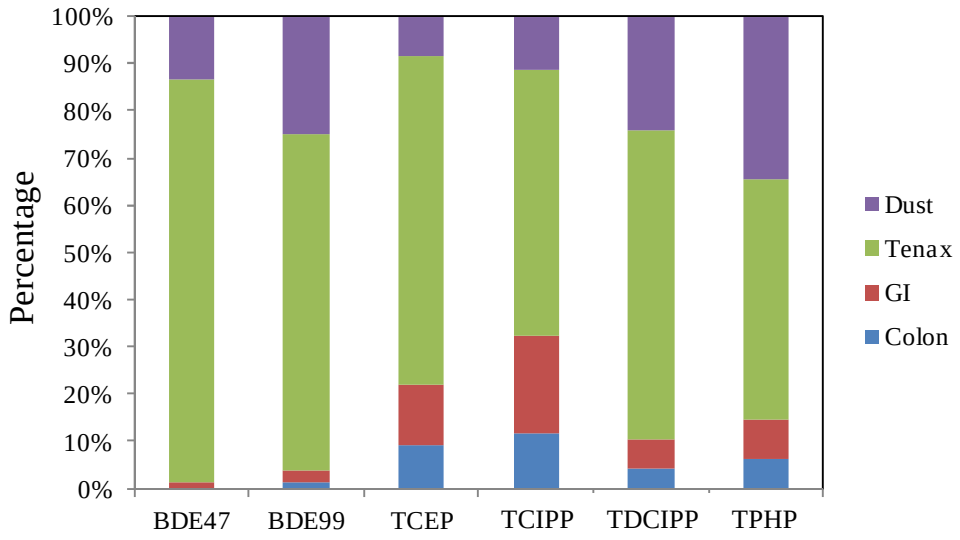

Figure S5. The distribution of BDE-47, BDE-99, and several OPFRs in four compartments including dust, TA, gastric-intestinal fluid and colon fluid after incubation.

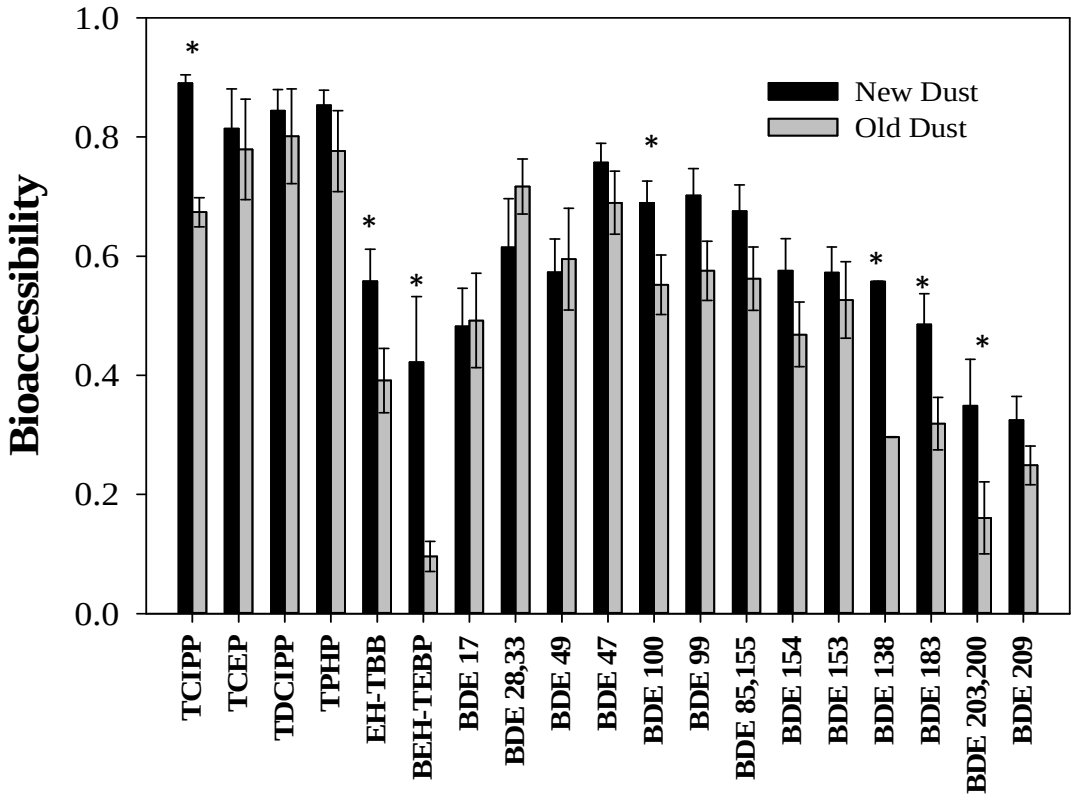

Figure S6. Flame retardant bioaccessibility measures in old dust samples (n=7, collected in 2006) and new dust samples (n=9, collected in 2010). Error bar represents the standard error.

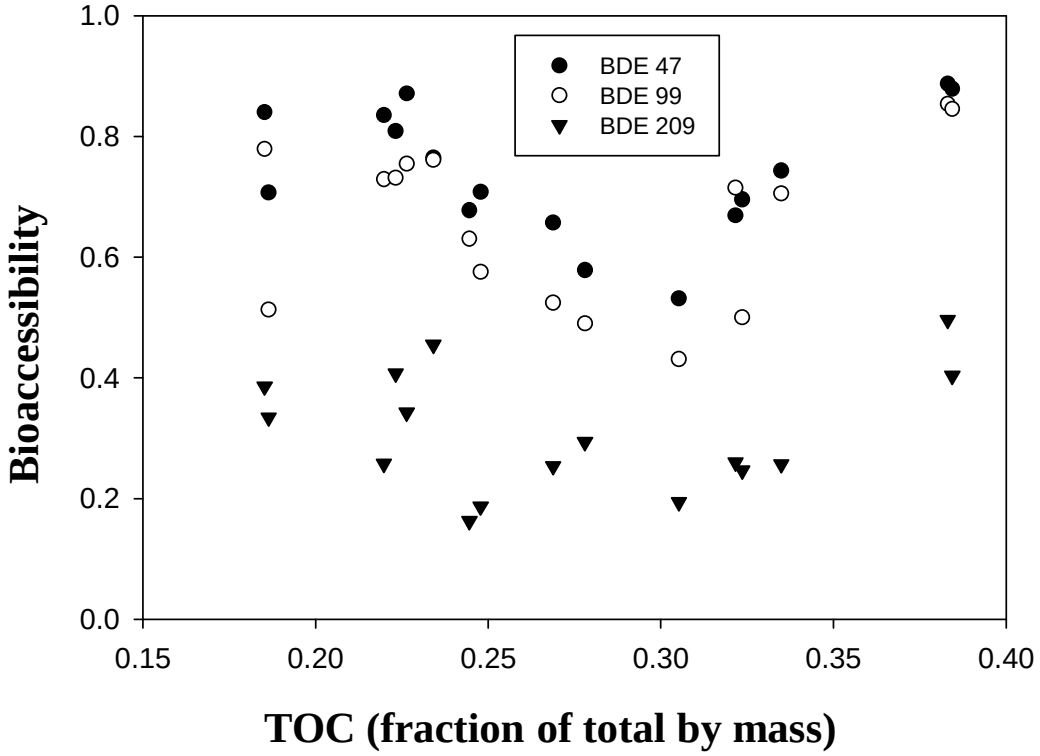

Figure S7. Association between measured bioaccessibility of BDEs and TOC in the various dust samples analyzed in this study (n=17).

Figure S8. Microscopic imaging (60 time magnification) of fragmented foam with particle size a) < 100 µm, b) <250 µm, and c) <500 µm in series.

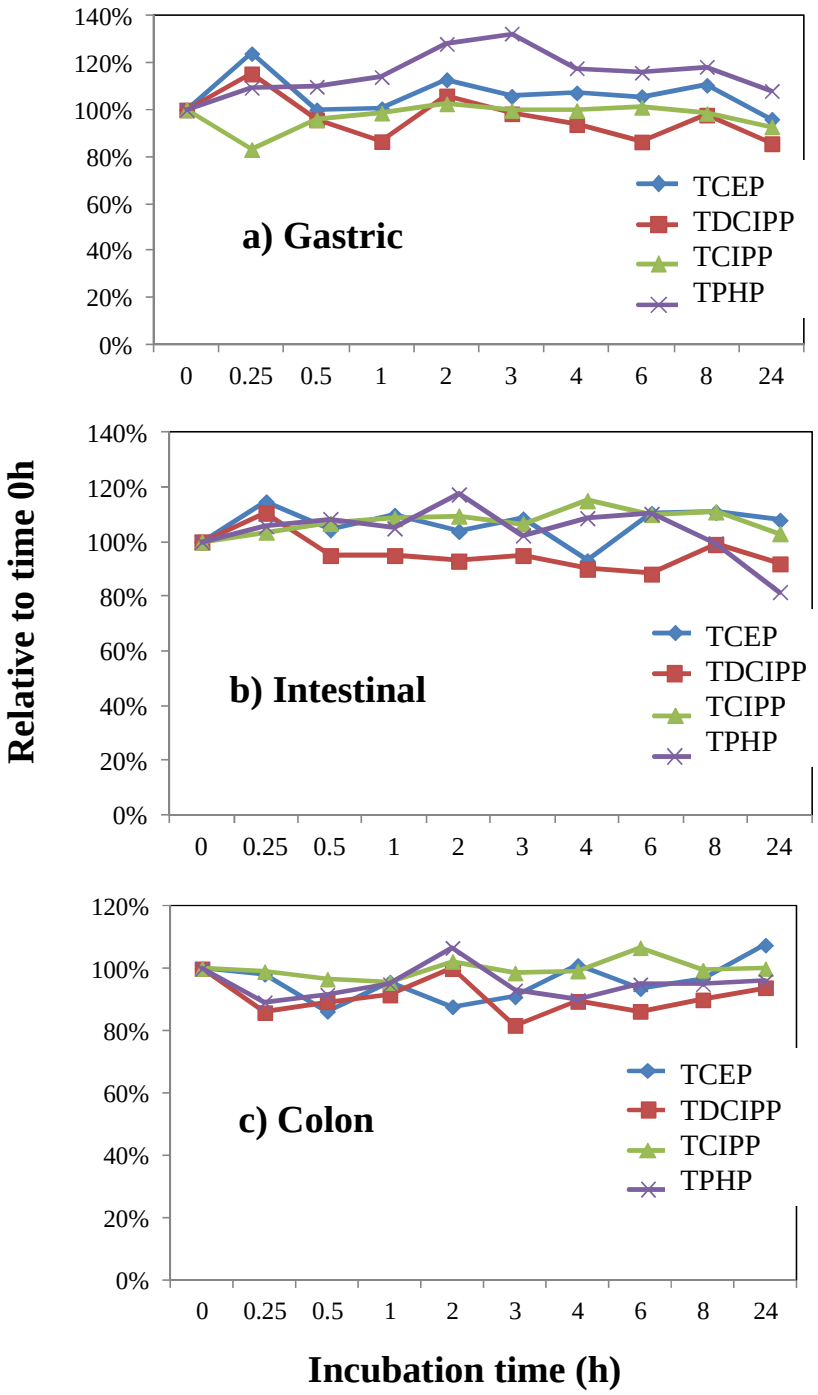

Figure S9. Relative amounts of TCEP, TDCIPP, TCIPP, and TPHP in the gastric, intestinal, and colon fluid during incubation at 37℃.

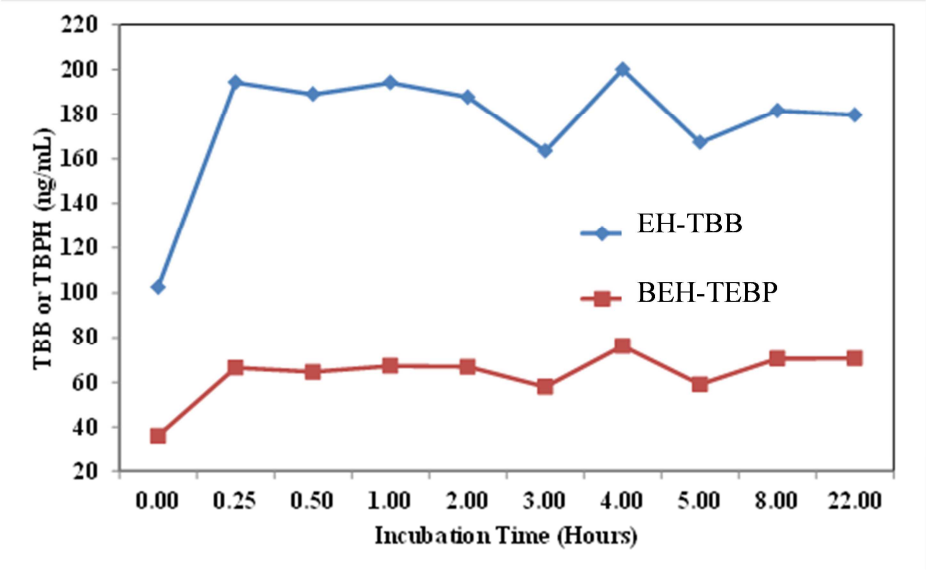

Figure S10. Concentrations (ng/mL) of EH-TBB and BEH-TEBP in intestinal fluid without addition of lipases at 37 ℃.

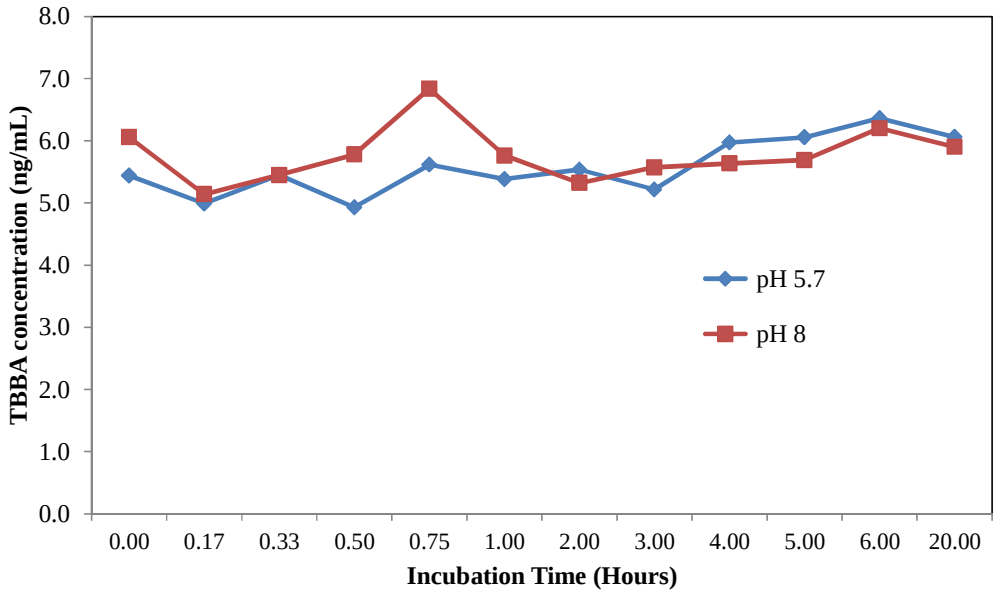

Figure S11. Concentrations of TBBA measured in intestinal fluid with 0.5 g TA added at two different pH values (5.7 and 8) at 37℃.

Table S1. Bioaccessibility measurements for OPFRs, FM550, and PBDEs in house dust samples (< 53 µm, n = 17)

	TCIPP	TCEP	TDCIPP	TPHP	EH-TBB	BEH-TEBP	BDE 17	BDE 28,33	BDE 49	BDE 47	BDE 100	BDE 99	BDE 85,155	BDE 154	BDE 153	BDE 138	BDE 183	BDE 203,200	BDE 209
DS1	91%	84%	83%		43%					68%	62%	63%	62%	58%	46%		44%	26%	16%
DS2	83%	76%	85%	78%	62%	79%		54%	53%	67%	68%	72%	67%	60%	46%		48%	23%	26%
DS3	93%	98%	93%	93%	67%	2%		91%	88%	89%	80%	85%	82%	74%	73%		55%	31%	50%
DS13	87%		64%	84%	67%	56%	37%	52%	59%	76%	72%	76%	72%	70%	65%	56%	59%	46%	45%
DS14	86%	48%	79%	80%	51%	36%	41%	33%	63%	74%	68%	71%	69%	60%	56%		43%	30%	26%
DS15	92%	65%	98%	76%	64%	49%	57%	82%	56%	84%	76%	78%	78%	34%	71%		66%	42%	39%
DS16	94%	88%	88%	91%	77%	60%	59%	36%	56%	88%	84%	85%	80%	78%	68%		66%	76%	40%
DS5	90%	96%	79%	92%	36%	14%		65%	44%	66%	55%	52%	51%	44%	42%		26%		25%
DS6	86%	96%	92%	88%	35%	0%		79%	39%	70%	54%	50%	46%	40%	49%		30%	6%	25%
DS7					34%	3%	43%	61%	56%	71%	51%	58%	55%	45%	44%		28%	8%	19%
DS8	73%	79%			61%		39%	85%	82%	87%	74%	75%	79%	70%	68%		52%	47%	34%
DS9			93%	95%	38%	6%	69%	81%	69%	71%	53%	51%	51%	41%	40%	30%	31%	12%	33%
DS10	69%	89%	85%	87%	32%	11%		63%	40%	58%	43%	49%	46%	35%	38%		23%	3%	29%
DS11	60%	72%	80%	77%	27%	10%		65%	25%	53%	41%	43%	43%	32%	79%		19%	17%	19%
DS12	70%	96%	90%	68%	42%	18%	46%	85%	81%	84%	68%	73%	69%	59%	55%		37%	20%	26%
DS17	65%	53%	53%	61%				62%	64%	60%	55%	54%	51%	46%	45%			15%	13%
DS4	92%	99%	89%	94%	35%	4%		91%	85%	87%	71%	73%	74%	59%	57%		46%	37%	28%

DS1, DS2, DS3, DS13, DS14, DS15, DS16, DS5, and DS6 were collected in 2010. DS7, DS8, DS9, DS10, DS11, DS12, and DS17 were collected in 2006. DS4 was collected in 2008.

References

1. Fang, M.; Webster, T. F.; Gooden, D.; Cooper, E. M.; McClean, M. D.; Carignan, C.; Makey, C.; Stapleton, H. M., Investigating a novel flame retardant known as V6: measurements in baby products, house dust, and car dust. *Environ. Sci. Technol.* **2013,** *47*, (9), 4449-54.
2. Stapleton, H. M.; Misenheimer, J.; Hoffman, K.; Webster, T. F., Flame retardant associations between children's handwipes and house dust. *Chemosphere* **2014,** *30*, (14), 00039-3.

3. Roberts, S. C.; Macaulay, L. J.; Stapleton, H. M., In vitro metabolism of the brominated flame retardants 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (TBB) and bis(2-ethylhexyl) 2,3,4,5-tetrabromophthalate (TBPH) in human and rat tissues. *Chem. Res. Toxicol.* **2012,** *25*, (7), 1435-41.