



Foam Replacement Environmental Exposure Study (FREES): Biomonitoring Results

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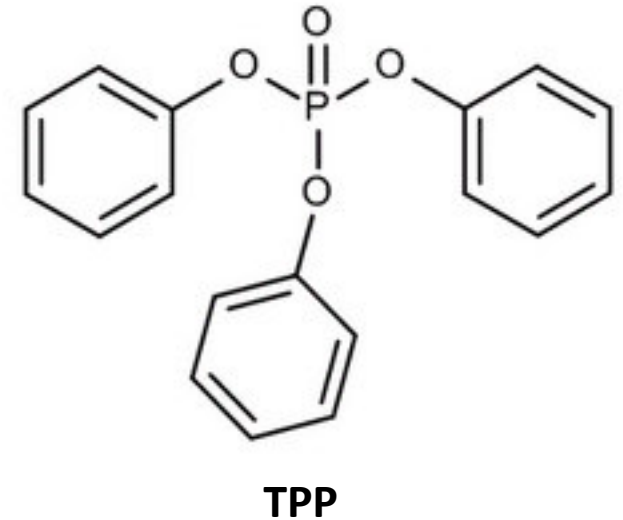
Complementing the existing study with biomonitoring

- Dust and foam:
 - UC Davis partnering with Green Science Policy Institute, Silent Spring Institute, and Environmental Working Group
 - DTSC foam analysis, UC Davis dust analysis
- Urine and serum:
 - Biomonitoring California
 - DTSC urine and serum analyses
 - Titled: Foam Replacement Environmental Exposure Study (FREES)



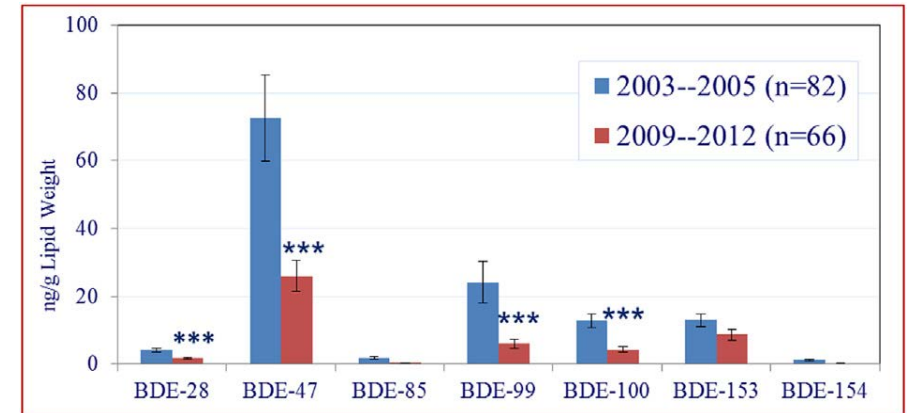
Biomonitoring analytes

- Polybrominated diphenyl ethers (PBDEs)
 - Prominent in foam furnishings: BDE-47, 99, 100, 153
 - Additional BDEs: 17, 28, 66, 85, 154, 183, 196, 197, 201, 202, 203, 206, 207, 208, and 209
- Organophosphate flame retardants (OPFRs)
 - Triphenyl phosphate (TPP)
 - Metabolite: diphenyl phosphate (DPP)
 - Tris (1,3-dichloro-2-propyl) phosphate (TDCPP)
 - Metabolite: bis (1,3-dichloro-2-propyl) phosphate (BDCPP)
 - Tris (2-chloroethyl) phosphate (TCEP)
 - Metabolite: bis-2-chloroethyl phosphate (BCEP)



Current time trends in PBDE biomarkers

- Environmental levels decreasing since ban in 2005 of 2 formulations
- Biomarkers were observed decreasing
- Some studies showing a recent plateau
- Biological half-lives still only partially understood
 - Estimates of 0.4-5.4 years for BDEs 47, 99, 100
 - Estimates of 3.5-11.7 years for BDE-153



Levels of PBDEs in breastmilk in CA women
Guo et al. 2015

Current time trends in OPFR biomarkers

- Environmental exposures increasing since the PBDE partial phase-out
- Studies showing large increases in biomarkers since 2002
- First time reported in NHANES (2013-2014)
 - 4 OPFRs detected in >81%
- Much shorter biological half-lives – hours
- Levels reflect much shorter times of recent exposure

Objective and approach

- Test if changes in biological levels of flame retardants is different between couch/foam replacers and a comparison group
 - Accounts for population time trends
 - Reduces impact of between-person differences (sex, race, age)

Comparison group: Intraprogram Pilot Study (IPP)

- Periodic sampling of volunteers
 - Mostly staff from OEHHA, DTSC, and CDPH
 - For testing or demonstration of laboratory methods
- IPPs in 2016 and 2017 focused on flame retardants
 - Participants removed from statistical analyses if had replaced furniture or moved homes
 - Similar demographics and environmental awareness to FREES participants

Participant numbers

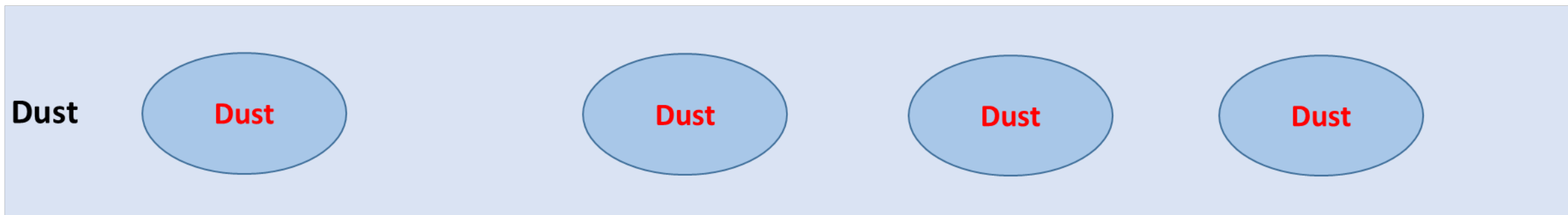
	First biological samples and 12 month samples
FREES - biomarkers	25
FREES and UC Davis dust study	23
IPP	28

Participant characteristics

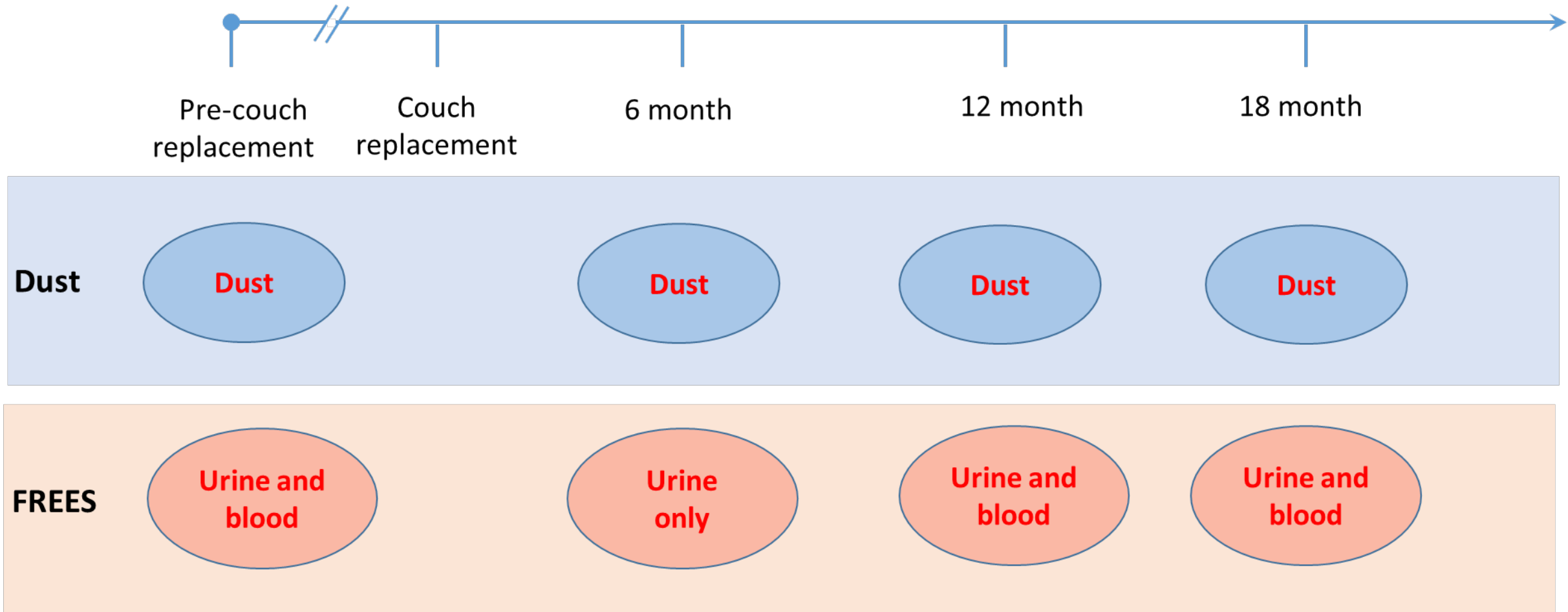
Sex	FREES	IPP
Female	17 (68%)	19 (68%)
Male	8 (32%)	9 (32%)

Race/ethnicity	FREES	IPP
White	17 (68%)	22 (79%)
Asian	4 (16%)	6 (21%)
Hispanic	2 (8%)	
Black	1 (4%)	
Other	1 (4%)	

UC Davis dust sampling timeline



FREES biomonitoring timeline



Timing comparison with Intraprogram Pilot Study

IPP



FREES

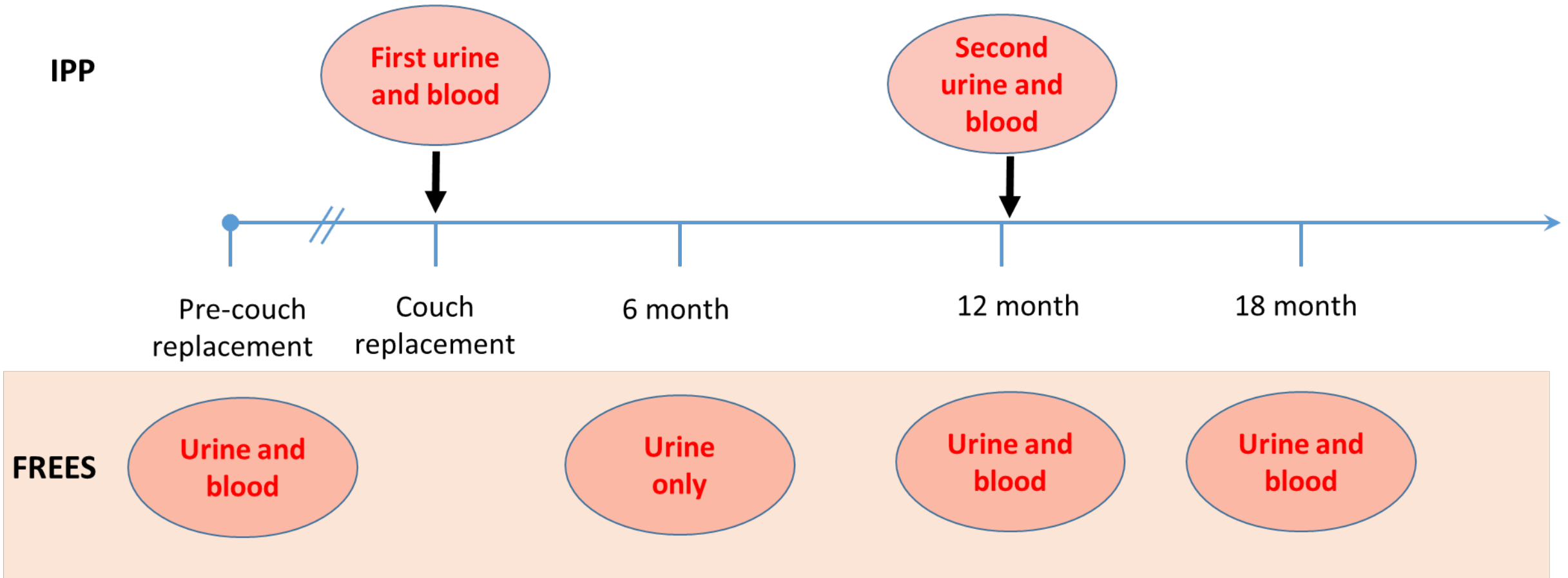
Urine and
blood

Urine
only

Urine and
blood

Urine and
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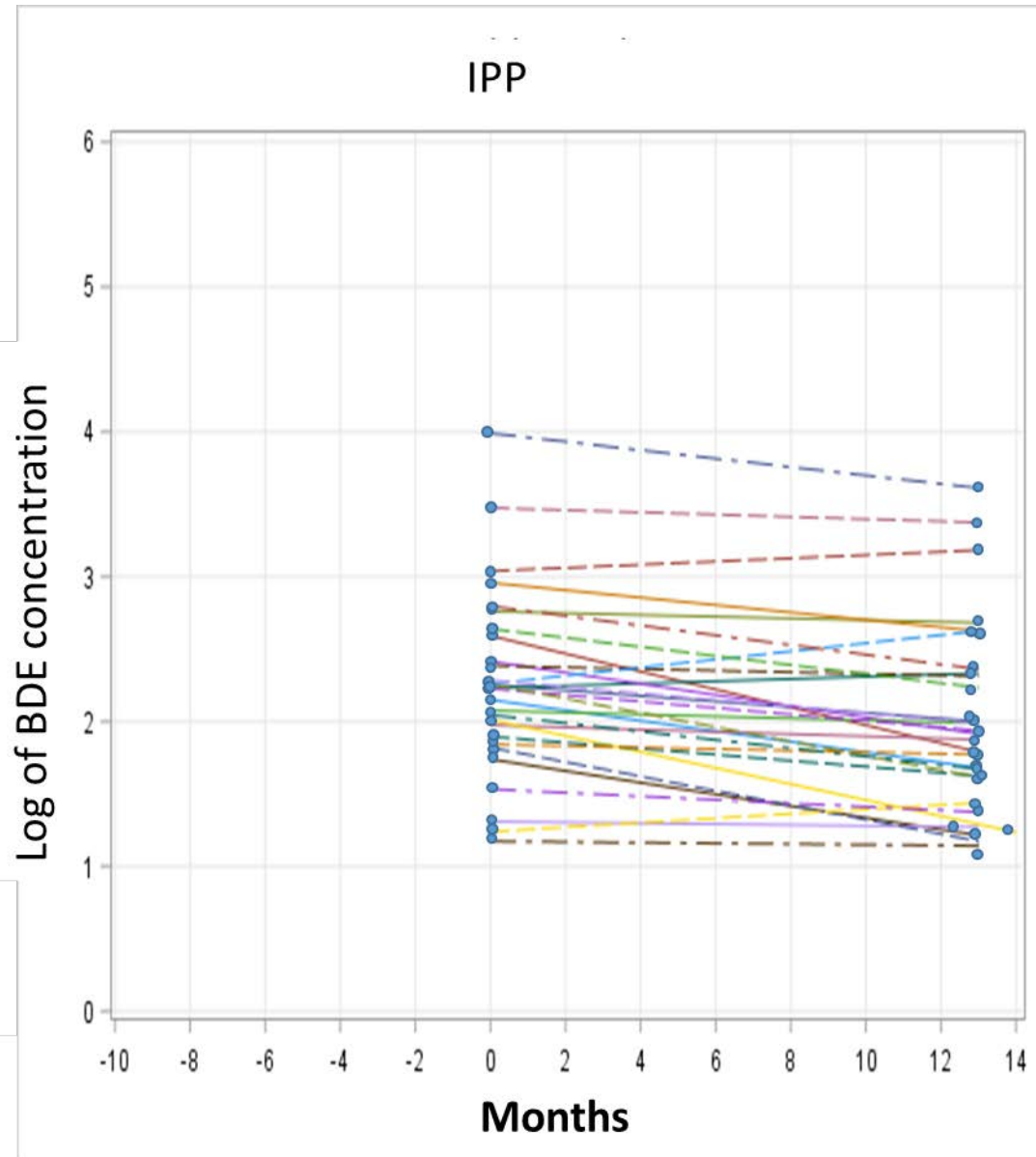
Timing comparison with Intraprogram Pilot Study



Dates of samples

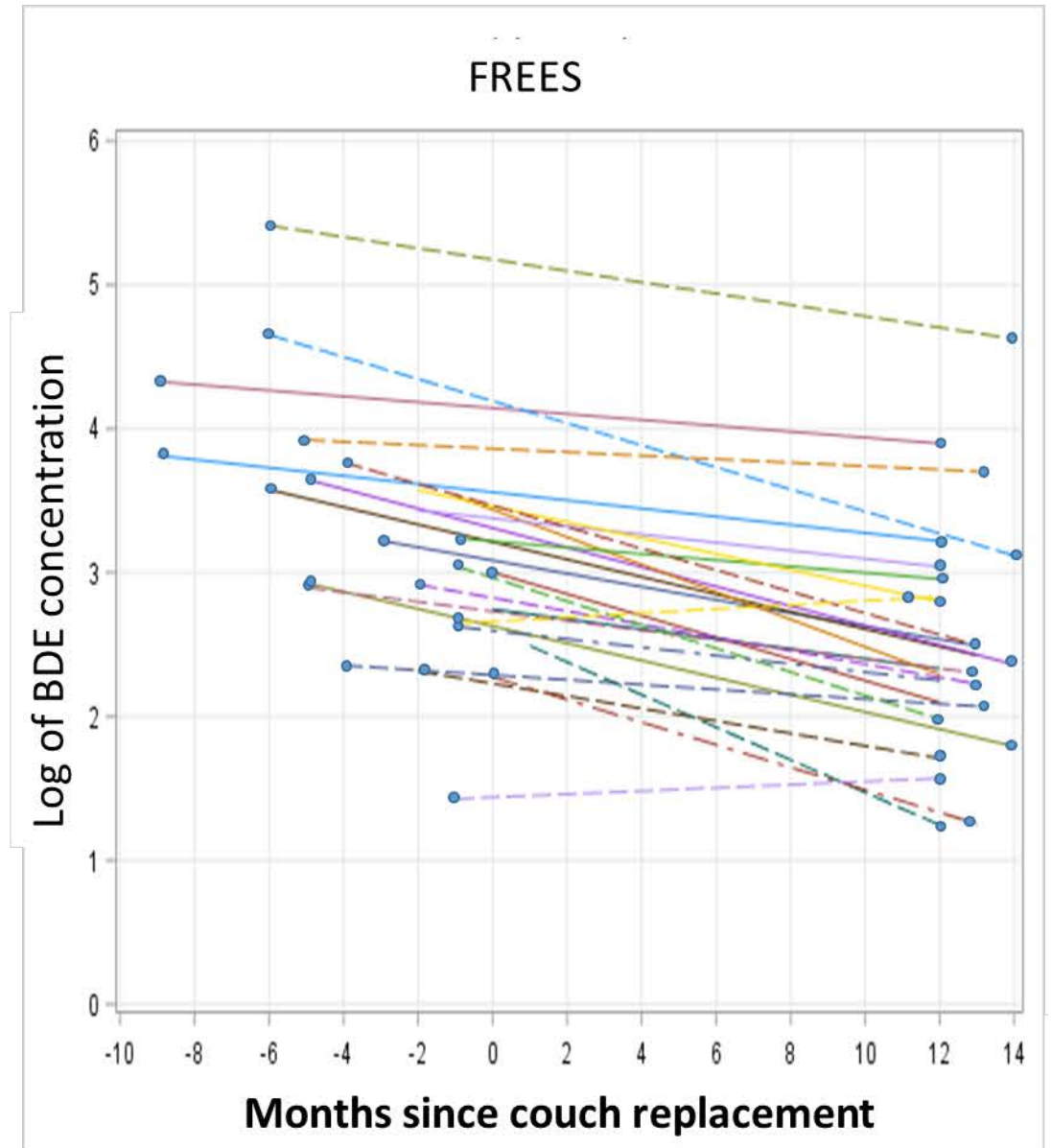
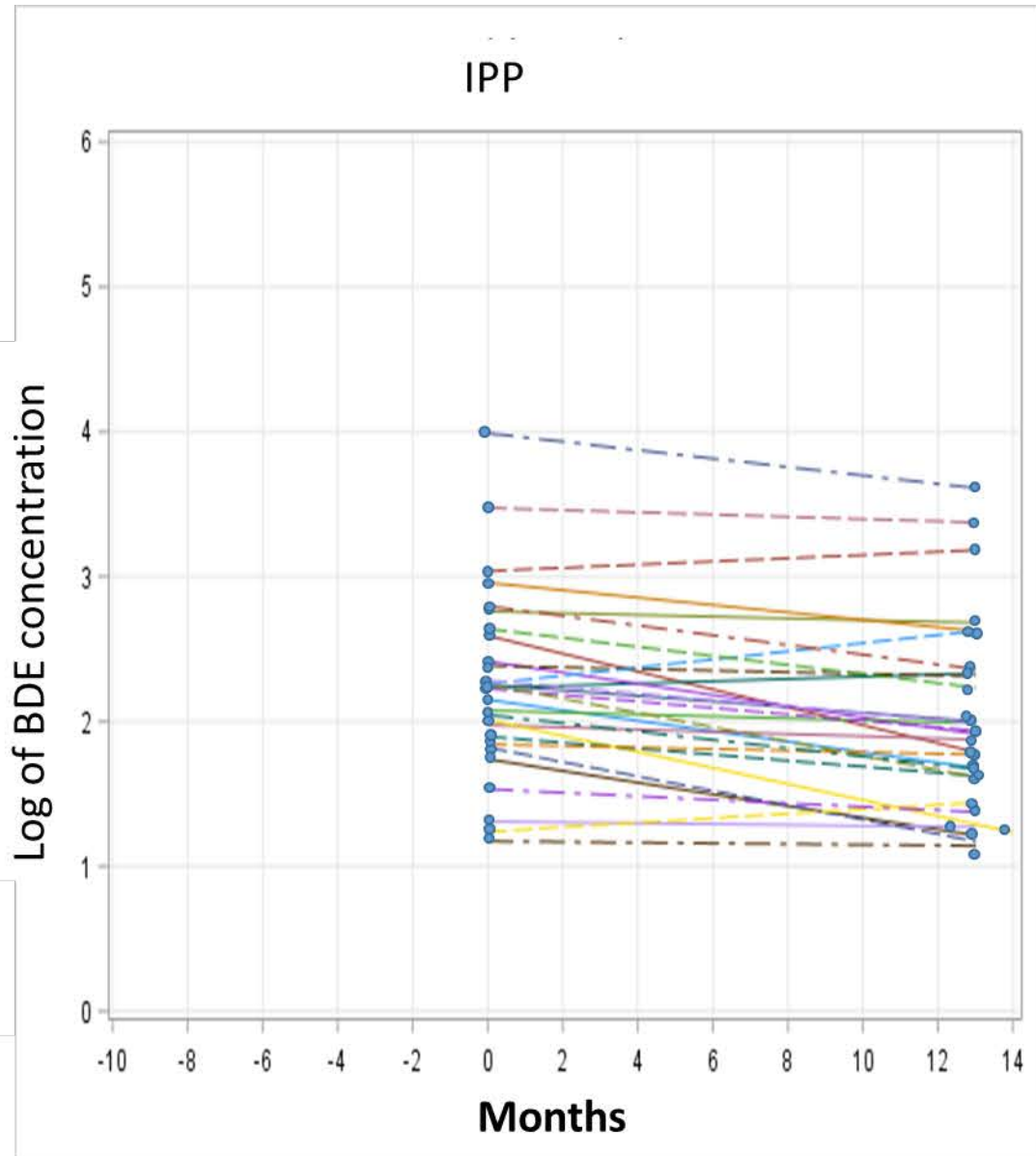
		Median	Range
Comparison Interval (yrs)	FREES	1.23	0.8 - 1.8
	IPP	1.08	1.0 - 1.1
First sample	FREES	10/2015	9/2015 - 9/2016
	IPP	8/2016	8/2016 - 9/2016
12 month sample	FREES	4/2017	10/2016 - 10/2017
	IPP	9/2017	9/2017 - 10/2017

Schematic – example change in a PBDE over time

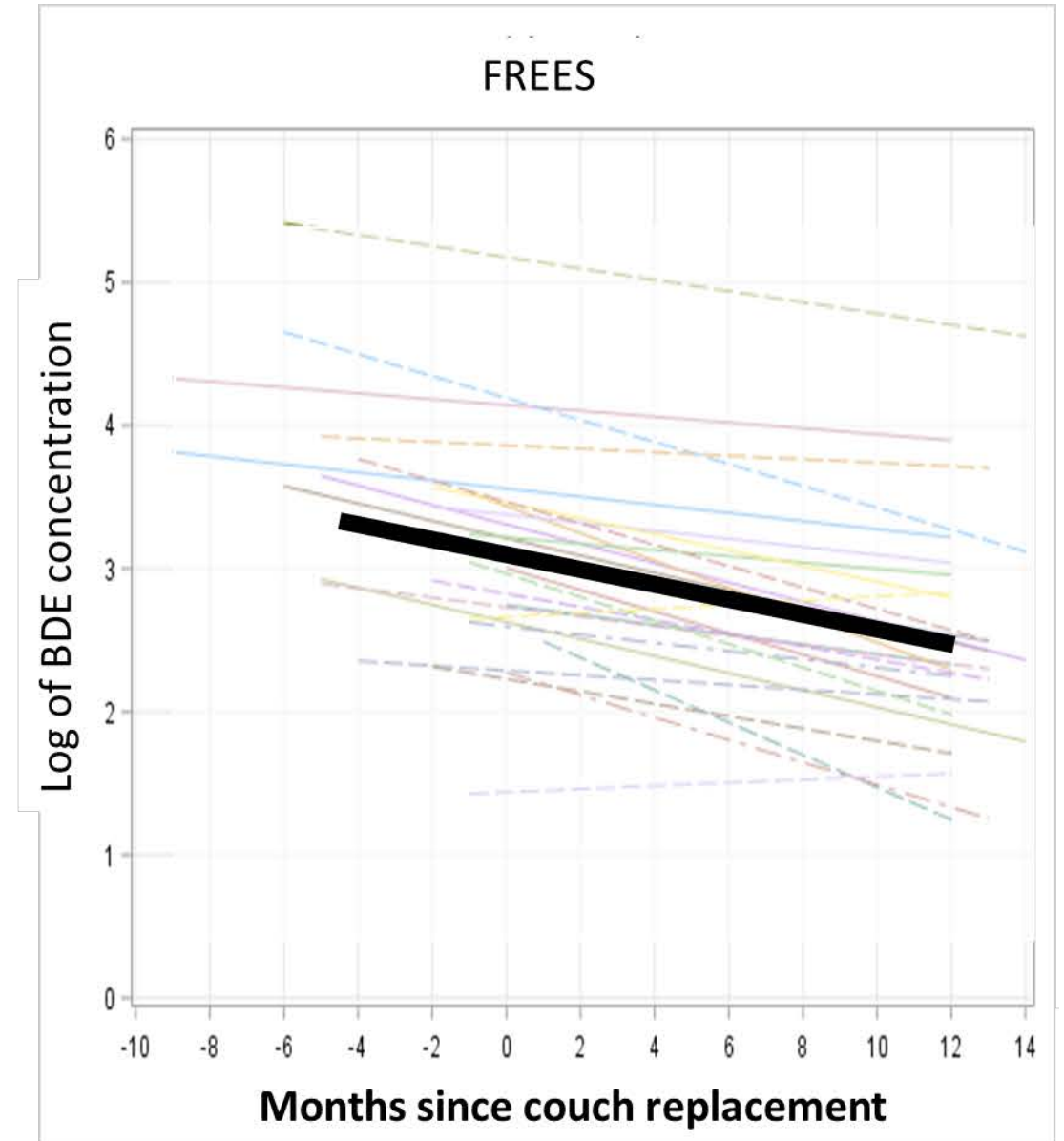
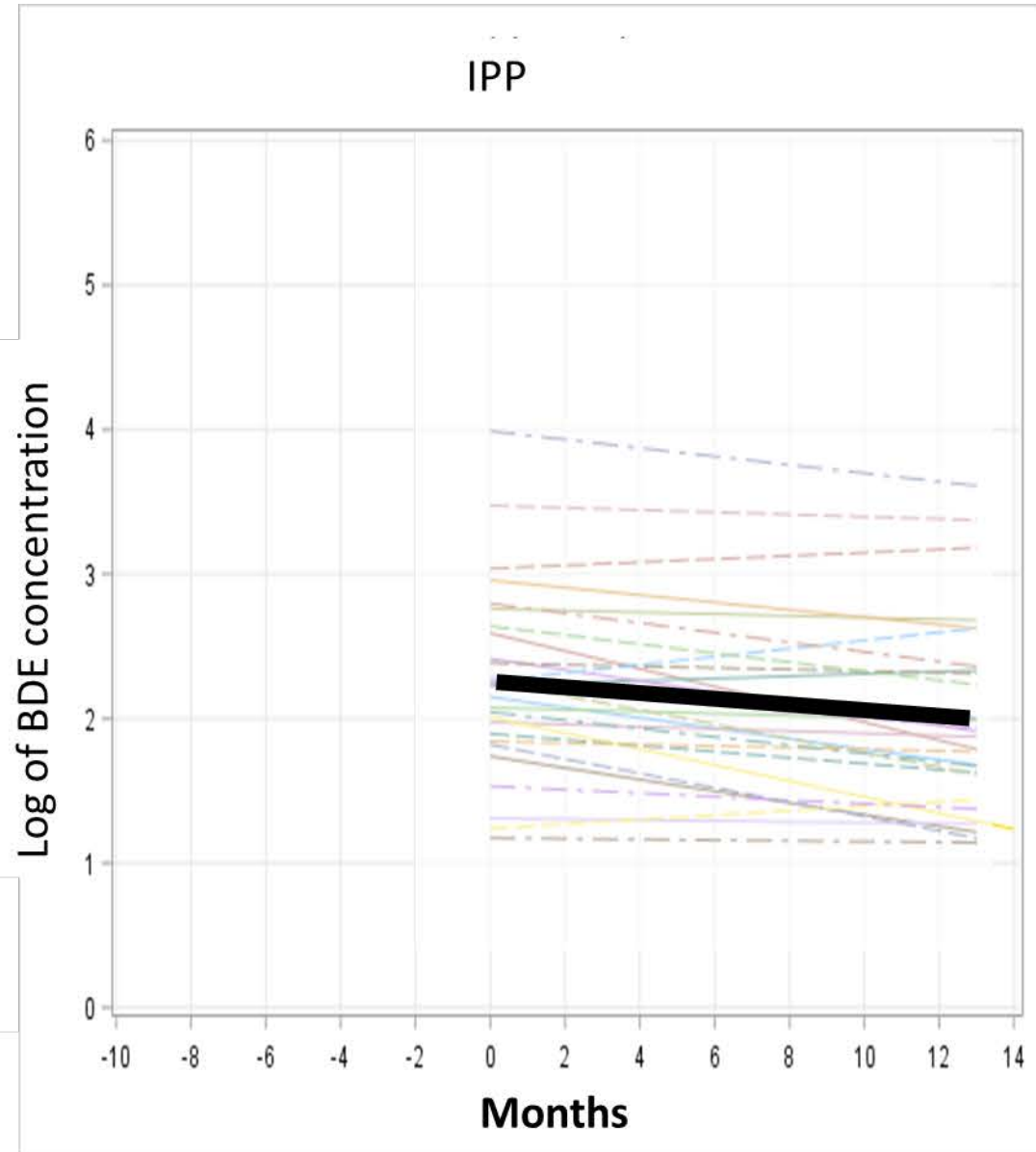


- Biological elimination
 - First order kinetics
 - Expect a log-linear decrease

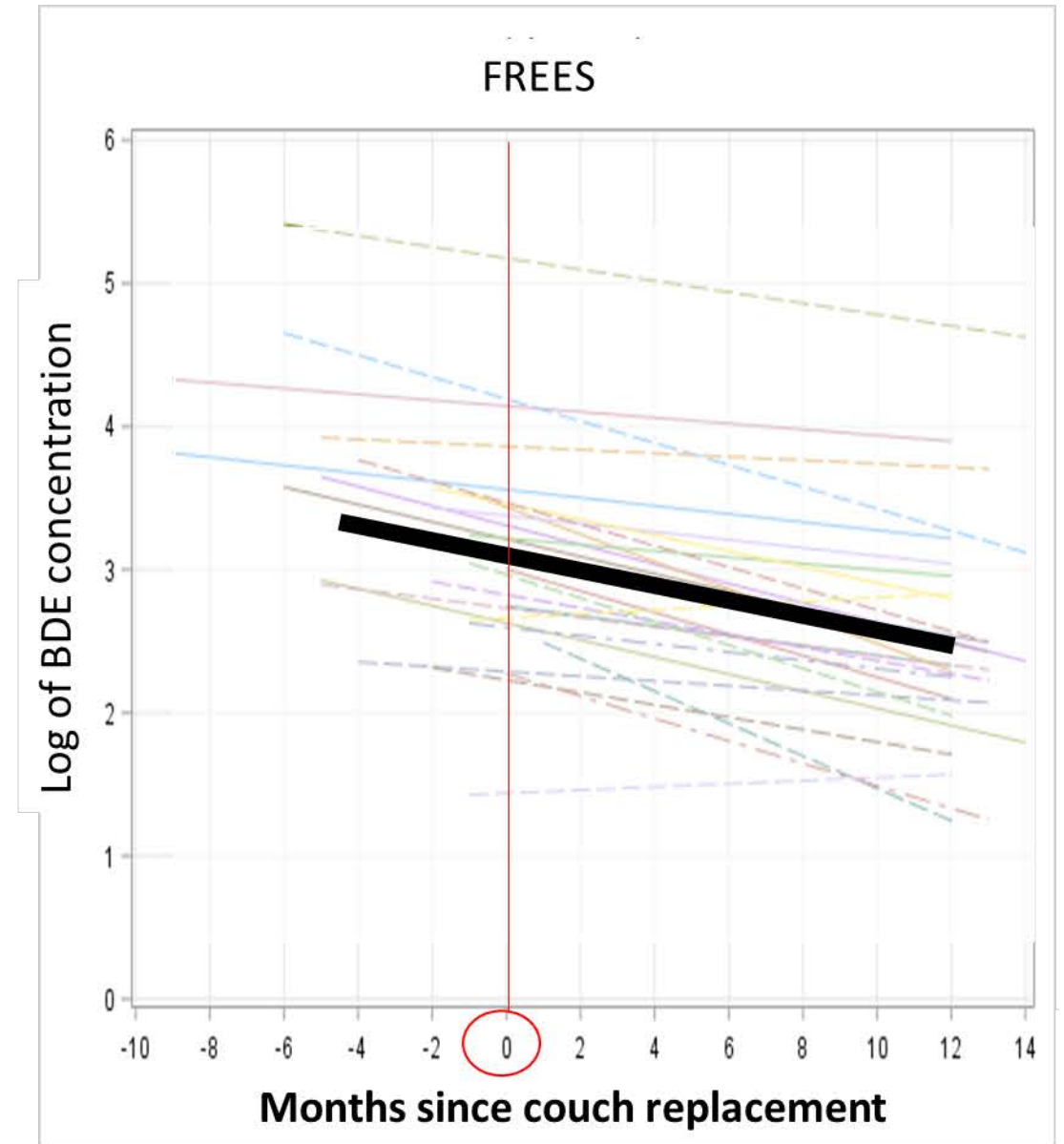
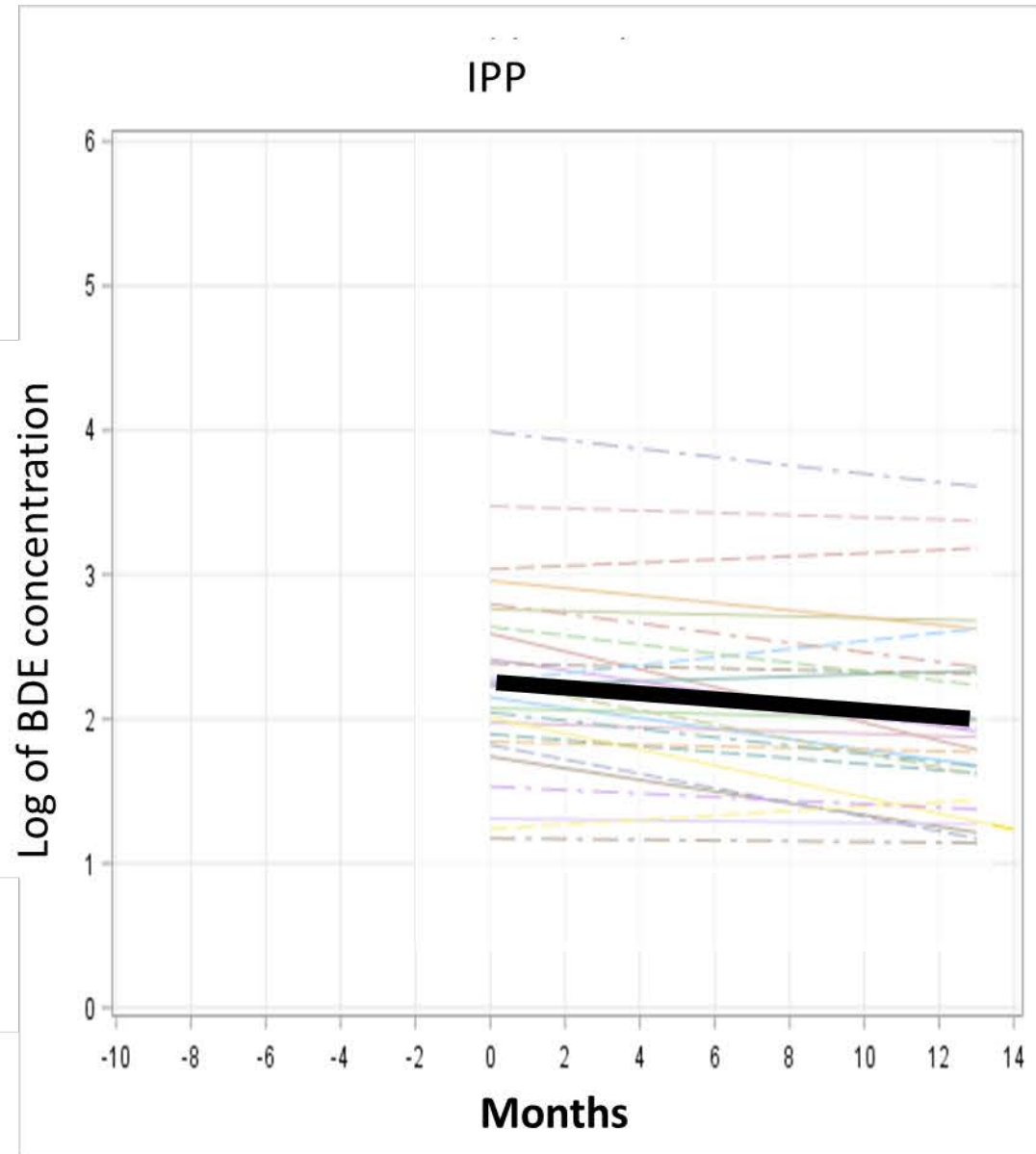
Schematic – example change in a PBDE over time



Schematic – example change in a PBDE



Schematic – example change in a PBDE



Results

- Preliminary findings

Initial PBDE concentrations - combined

Geometric means, lipid adjusted (ng/g lipid)

	FREES + IPP (2015-2016, n=53)	CA Teachers' Study (2011-2015, n = 1253) ^a
BDE 47	15.04	14.6
BDE 99	4.73	--
BDE 100	2.74	2.62
BDE 153	6.23	5.72

Other BDE detection frequencies:

- BDE 28 40%
- All others < 12% (17, 66, 85, 154, 183, 196, 197, 201, 202, 203, 206, 207, 208, and 209)

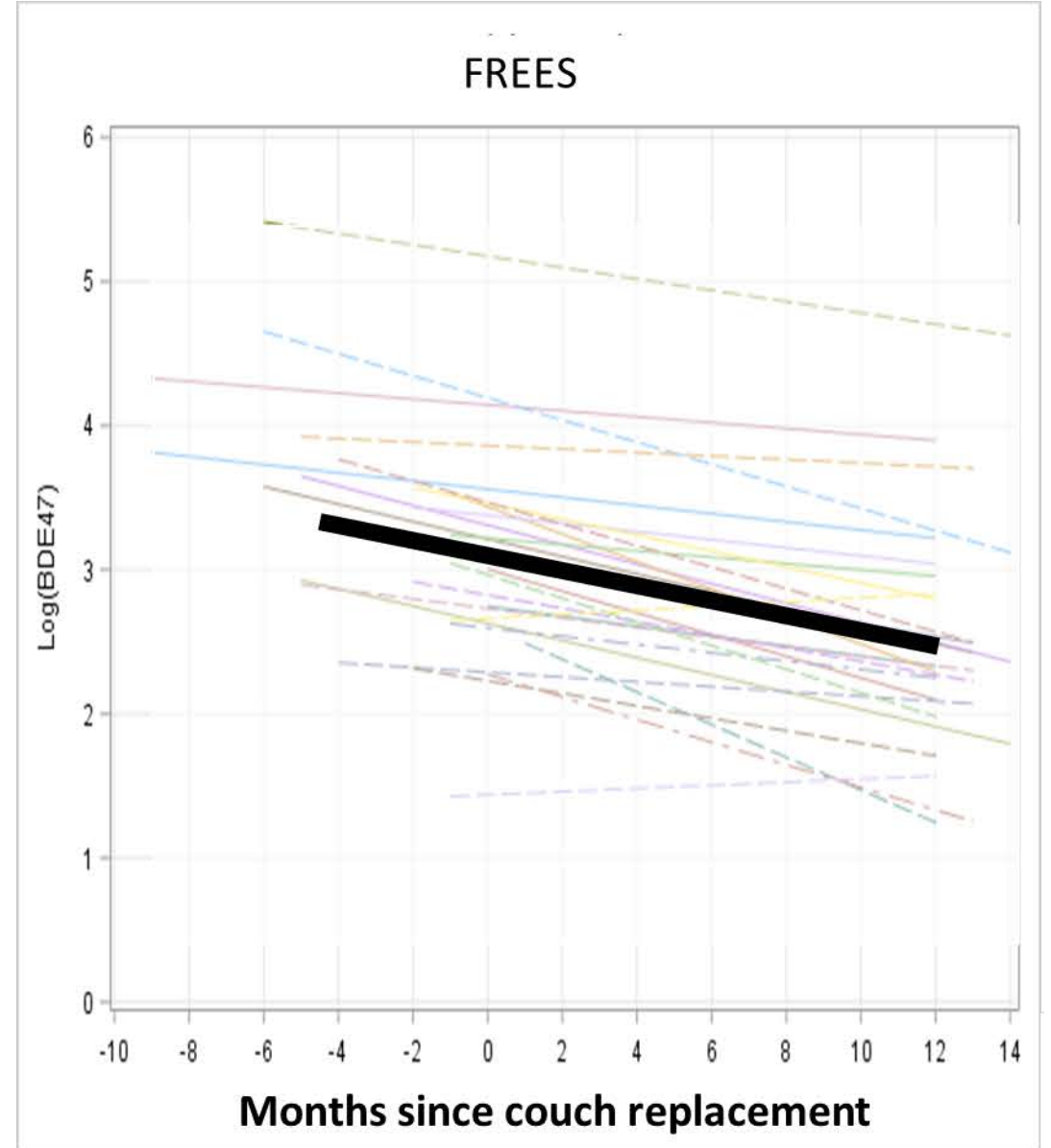
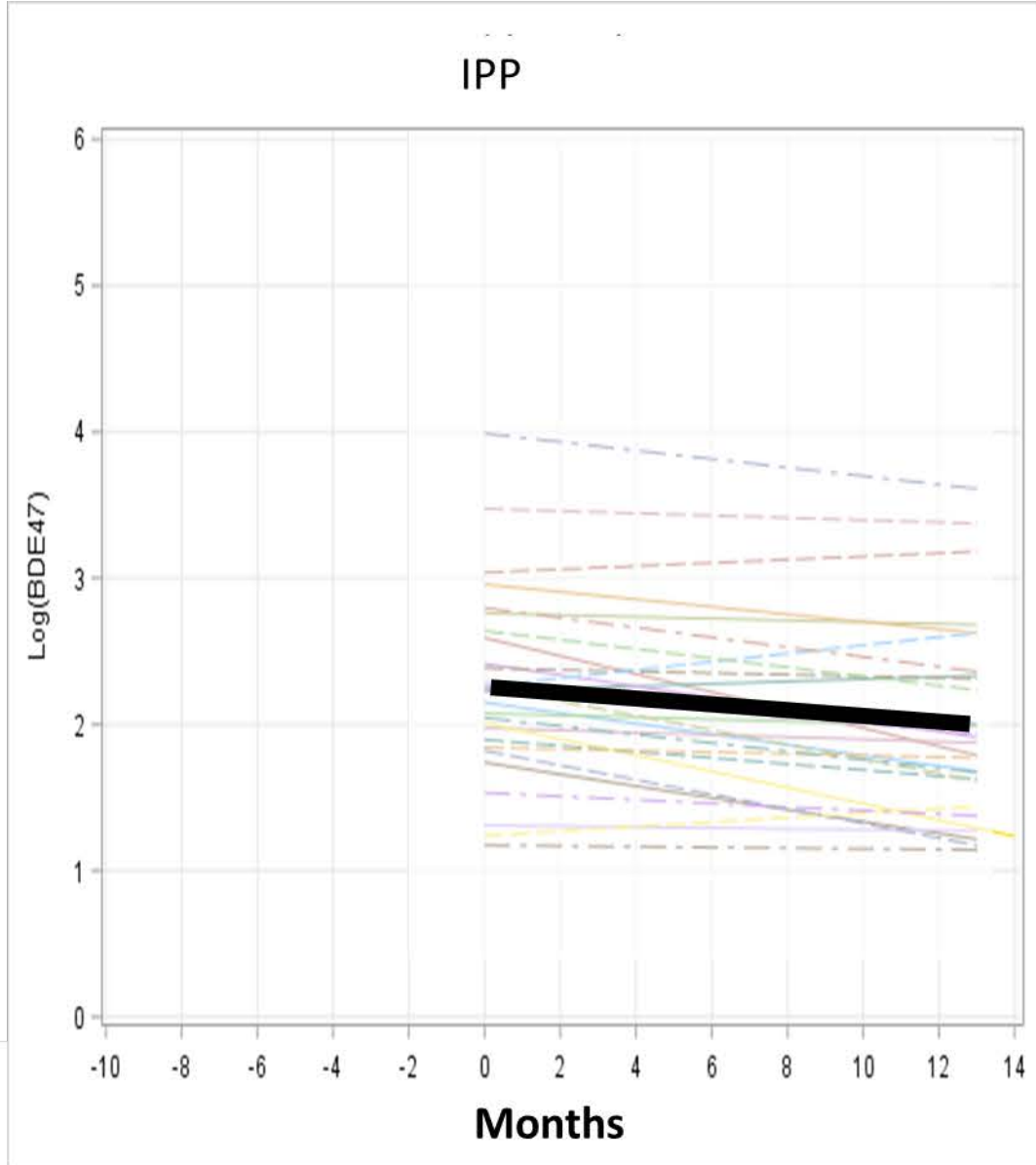
^aHurley et al. 2018

Initial PBDE concentrations

Geometric means, lipid adjusted (ng/g lipid)

	IPP (n=28)	FREES (n=25)	Comparison p-value
BDE 47	9.52	25.09	<0.01
BDE 99	3.26	7.18	<0.01
BDE 100	1.77	4.48	<0.01
BDE 153	4.13	9.89	<0.01

BDE 47 change over time



PBDE changes over time

Chemical	Study	% change over 1 year	Difference in slopes p-value*
BDE 47	IPP	-21%	
	FREES	-43%	<0.01
BDE 99	IPP	-23%	
	FREES	-41%	0.01
BDE 100	IPP	-16%	
	FREES	-36%	<0.01
BDE 53	IPP	-17%	

Preliminary findings

*p-value for t-tests of slopes

Initial OPFR concentrations

Geometric mean, unadjusted ($\mu\text{g/L}$)

	FREES + IPP (2015-2016, n=53)	NHANES, 20+ years (2013-2014)
BCEP	1.01	0.38
BDCPP	1.31	0.72
DPP	1.22	0.73

Initial OPFR concentrations

Geometric means, specific gravity adjusted ($\mu\text{g/L}$)^a

	IPP (n=28)	FREES (n=25)	Comparison p-value
BCEP	1.22	2.00	0.03
BDCPP	1.96	1.95	0.99
DPP	1.41	2.44	<0.01

^aUsing reference SG of 1.017

OPFR analytical approach

- Analytical approach is different because of short half-lives
- Expect initial drop from removal of couch followed by more stable measurements
- Compare before and after (“12 month” time point)
 - Use log-transformed specific gravity adjusted measurements
 - Linear regressions with repeated measurements
- Examine FREES correlations in 6, 12, and 18 month values

BCEP

	Timepoint	Modeled geometric mean (µg/L)	% Change	
IPP	0 month	1.22		
IPP	12 month	2.23	+84%	
FREES	Pre-couch replacement	1.97		
FREES	12 month	1.74	-12%	

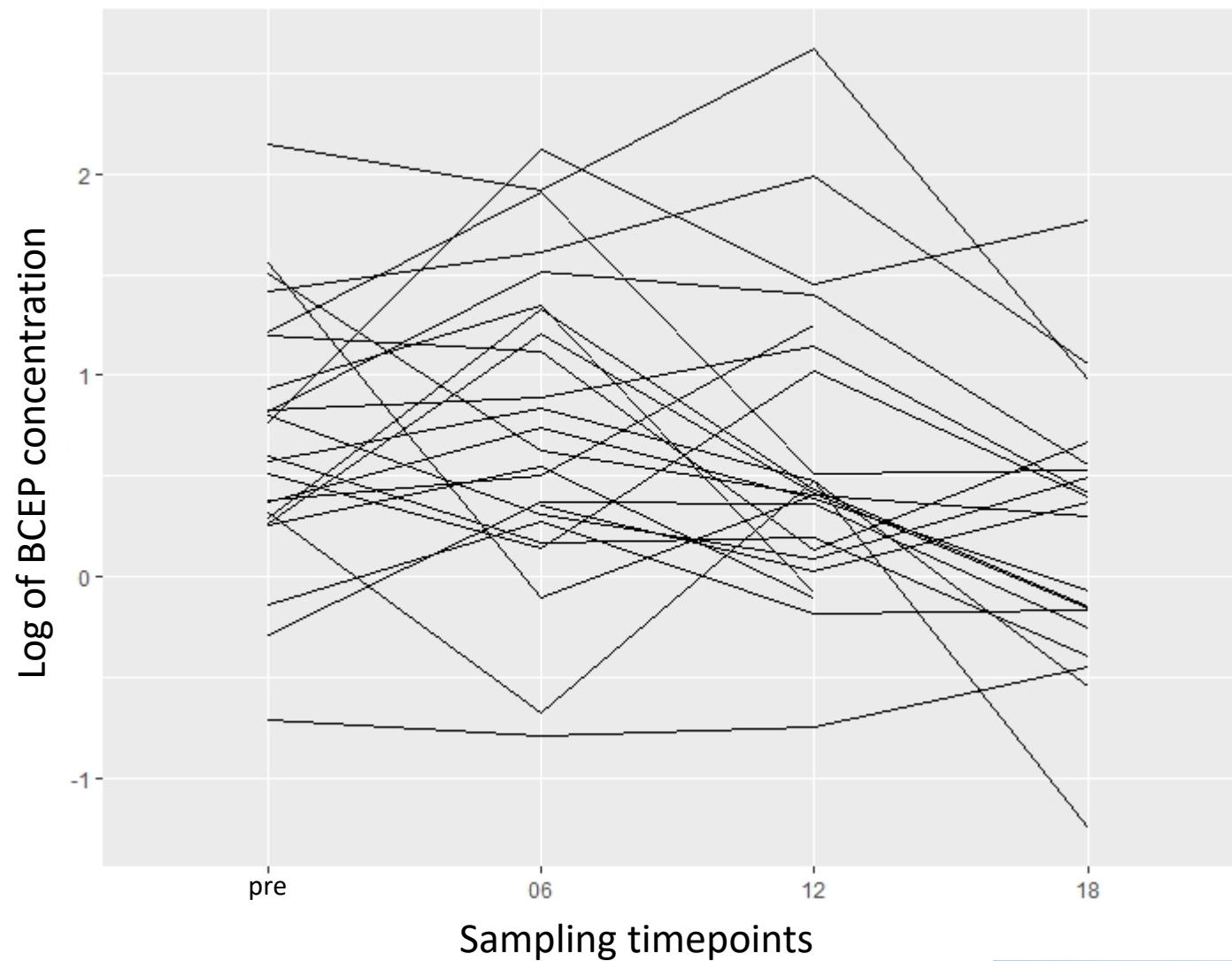
FREES levels go down, but not significantly

BCEP - FREES participants

Moderate correlations
Over 6, 12, 18 months

ρ : 0.59-0.68

Intraclass
Correlation
Coefficient (ICC): 0.57



BDCPP

	Timepoint	Modeled geometric mean (µg/L)	% Change	P-value for change
IPP	0 month	1.96		
IPP	12 month	1.60	-18%	0.24
FREES	Pre-couch replacement	1.95		
FREES	12 month	0.92	-53%	<0.01

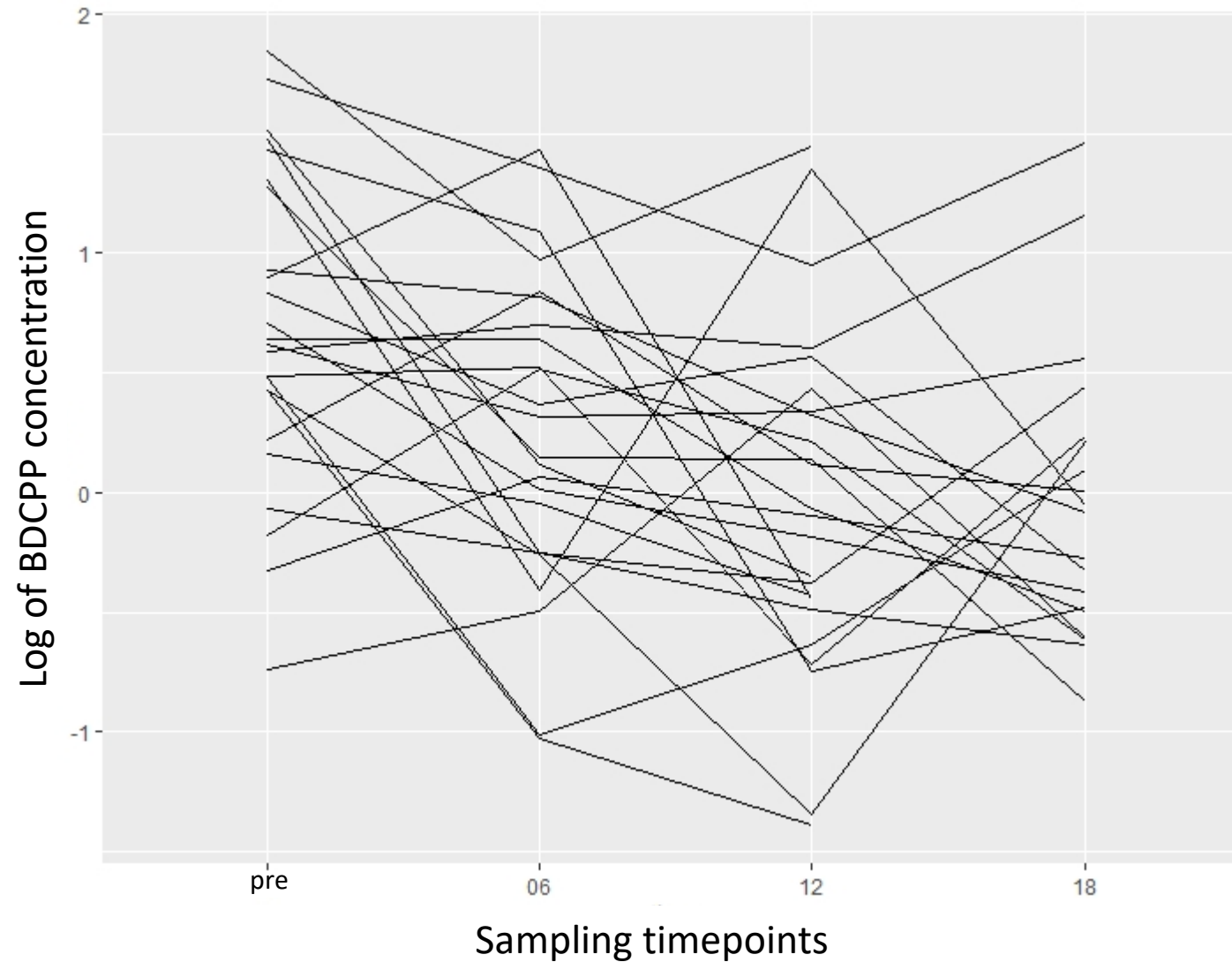
FREES levels go down relative to IPP

BDCPP – FREES participants

Low correlations over
6, 12, 18 months

ρ : 0.30-0.37

ICC: 0.42



DPP

	Timepoint	Modeled geometric mean ($\mu\text{g/L}$)	% Change	P-value for change
IPP	0 month	1.41		
IPP	12 month	0.98	-30%	<0.01
FREES	Pre-couch replacement	2.44		
FREES	12 month	2.39	-2%	0.88

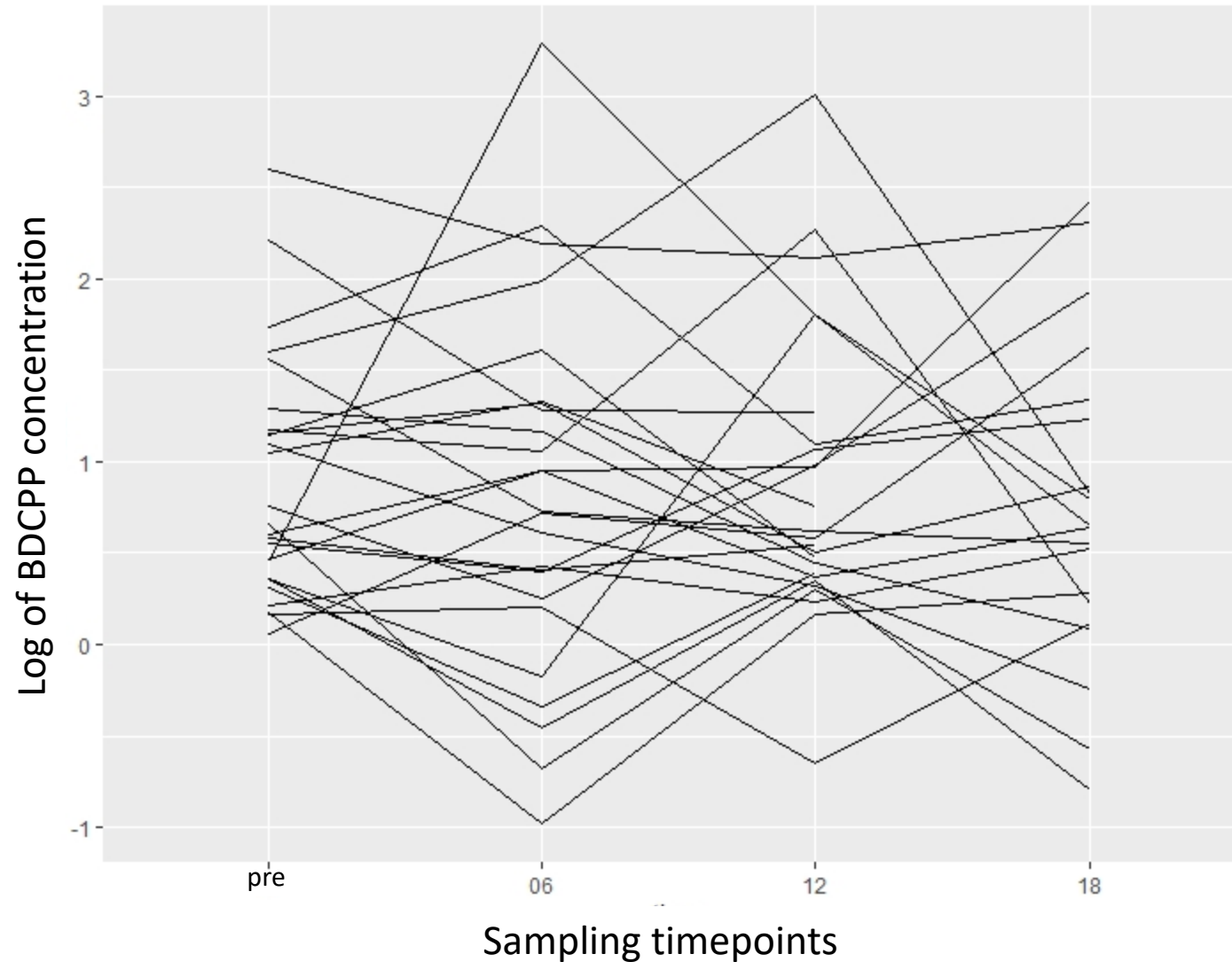
FREES levels stable while IPP levels go down

DPP – FREES participants

Low to mid correlations
over 6, 12, 18 months

ρ : 0.37-0.55

ICC: 0.42



Associations with behaviors - FREES

- Initial handwashing frequency
 - No associations with initial concentrations or change over time
 - Few reported any change in handwashing frequency over time
- Vegetarians vs. meat eaters
 - No associations
- Hours at work computer
 - No associations
- Sleeping on a foam mattress
 - Associated with increased initial PBDE levels, no association with change over time

Sensitivity tests

- Influence of sex and race
 - No differences by race
 - Females had greater change in BDE-99
- Different beginning ranges of flame retardants in the two groups
 - Limited FREES to only those in same range of PBDE concentrations as IPP
 - No difference
- Clustering of people in same homes did not affect chemical level changes

Future work

- Biomarker levels may not be sufficient to prove intervention was the source of any particular change
- Ways to address this further will include:
 - Coordinated analyses of dust, foam, and biomarker data
 - Further review of questionnaire data

Limitations

- Limited availability of information on behavior and behavior change for IPP
- Questionnaires for FREES may not have captured all behavior changes
- Small sample sizes reduce our confidence in assessing other sources of variability and sources of possible confounding

Compared to other intervention or time change studies

- Handwashing and house cleaning intervention (1 week each)
 - Up to 52% decreases in some OPFRs for individual intervention, increases in others
- Foam/dust exposures before and after gymnastics practice
 - 50% increase in DPP after practice
- Within-person OPFR variability over 5 weeks
 - Interclass correlations of 0.54-0.67
- Within-person PBDE variability over a year (2010-2011)
 - Interclass correlations of 0.91-0.98



Conclusions

- PBDE measurements decreased at a greater rate in FREES compared to IPP group, except for BDE-153
- OPFR measurements showed differing patterns and may be complicated by their short half-lives
- Attributing this intervention to any particular chemical change requires further interrogation of dust, foam, and questionnaire items

Acknowledgements

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- UC Davis – Bennett lab
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- Environmental Working Group
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References

- Carignan CC, Fang M, Stapleton HM, Heiger-Bernays W, McClean MD, Webster TF. Urinary biomarkers of flame retardant exposure among collegiate U.S. gymnasts. *Environ Int.* 2016 Sep;94:362-368.
- Gibson EA, Stapleton HM, Calero L, Holmes D, Burke K, Martinez R, Cortes B, Nematollahi A, Evans D, Herbstman JB. Flame retardant exposure assessment: findings from a behavioral intervention study. *J Expo Sci Environ Epidemiol.* 2019 Jan;29(1):33-48.
- Guo W, Holden A, Smith SC, Gephart R, Petreas M, Park JS. PBDE levels in breast milk are decreasing in California. *Chemosphere.* 2016 May;150:505-513.
- Gyalpo T, Scheringer M, Hungerbühler K. Recommendations for Evaluating Temporal Trends of Persistent Organic Pollutants in Breast Milk. *Environ Health Perspect.* 2016 Jul;124(7):881-5.
- Hurley S, Goldberg D, Nelson DO, Guo W, Wang Y, Baek HG, Park JS, Petreas M, Bernstein L, Anton-Culver H, Reynolds P. Temporal Evaluation of Polybrominated Diphenyl Ether (PBDE) Serum Levels in Middle-Aged and Older California Women, 2011-2015. *Environ Sci Technol.* 2017 Apr 18;51(8):4697-4704.
- Makey CM, McClean MD, Sjödin A, Weinberg J, Carignan CC, Webster TF. Temporal variability of polybrominated diphenyl ether (PBDE) serum concentrations over one year. *Environ Sci Technol.* 2014 Dec 16;48(24):14642-9.
- Ospina M, Jayatilaka NK, Wong LY, Restrepo P, Calafat AM. Exposure to organophosphate flame retardant chemicals in the U.S. general population: Data from the 2013-2014 National Health and Nutrition Examination Survey. *Environ Int.* 2018 Jan;110:32-41.
- Wang Y, Li W, Martínez-Moral MP, Sun H, Kannan K. Metabolites of organophosphate esters in urine from the United States: Concentrations, temporal variability, and exposure assessment. *Environ Int.* 2019 Jan;122:213-221



Thank you!

Questions?

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