

Fate of polycyclic aromatic compounds (PACs) from diluted bitumen spilled into freshwater limnocorrals

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Supporting Information

24 pages

7 Tables

17 Figures

Tables

Table S1. Identified targets and abbreviations. Abbreviations s,w,a represent sediment, water, and air samples.

Biomarker compounds		Alkylated PACs	
^{s,w} C21 tricyclic terpene	C ₂₁ T	^{s,w,a} C ₀ -Naphthalene	C ₀ -NAP
^{s,w} C22 tricyclic terpene	C ₂₂ T	^{s,w,a} C ₁ -Naphthalenes	C ₁ -NAP
^{s,w} C23 tricyclic terpene	C ₂₃ T	^{s,w,a} C ₂ -Naphthalenes	C ₂ -NAP
^{s,w} C24 tricyclic terpene	C ₂₄ T	^{s,w,a} C ₃ -Naphthalenes	C ₃ -NAP
^{s,w} 18α(H),21β(H)-22,29,30-trinorhopane	C ₂₇ Ts	^{s,w,a} C ₄ -Naphthalenes	C ₄ -NAP
^{s,w} 17α(H),21β(H)-22,29,30-trinorhopane	C ₂₇ Tm	^{s,w,a} C ₀ -Phenanthrene/anthracene	C ₀ -PHEN
^{s,w} 17α(H),21β(H)-30-norhopane	C ₂₉ αβ H	^{s,w,a} C ₁ -Phenanthrenes/anthracenes	C ₁ -PHEN
^{s,w} 17α(H),21β(H)-hopane	C ₃₀ αβ H	^{s,w,a} C ₂ -Phenanthrenes/anthracenes	C ₂ -PHEN
^{s,w} 22S-17α(H),21β(H)-30-homohopane	C ₃₁ (S) H	^{s,w,a} C ₃ -Phenanthrenes/anthracenes	C ₃ -PHEN
^{s,w} 22R-17α(H),21β(H)-30-homohopane	C ₃₁ (R) H	^{s,w,a} C ₄ -Phenanthrenes/anthracenes	C ₄ -PHEN
^{s,w} 22S-17α(H),21β(H)-30,31-bishomohopane	C ₃₂ (S) H	^{s,w,a} C ₀ -Dibenzothiophene	C ₀ -DBT
^{s,w} 22R-17α(H),21β(H)-30,31-bishomohopane	C ₃₂ (R) H	^{s,w,a} C ₁ -Dibenzothiophenes	C ₁ -DBT
^{s,w} 22S-17α(H),21β(H)-30,31,32-trishomohopane	C ₃₃ (S) H	^{s,w,a} C ₂ -Dibenzothiophenes	C ₂ -DBT
^{s,w} 22R-17α(H),21β(H)-30,31,32-trishomohopane	C ₃₃ (R) H	^{s,w,a} C ₃ -Dibenzothiophenes	C ₃ -DBT
^{s,w} 22S-17α(H),21β(H)-30,31,32,33-tetrakishomohopane	C ₃₄ (S) H	^{s,w,a} C ₀ -Fluorene	C ₀ -FLU
^{s,w} 22R-17α(H),21β(H)-30,31,32,33-tetrakishomohopane	C ₃₄ (R) H	^{s,w,a} C ₁ -Fluorenes	C ₁ -FLU
^{s,w} 22S-17α(H),21β(H)-30,31,32,33,34-pentakishomohopane	C ₃₅ (S) H	^{s,w,a} C ₂ -Fluorenes	C ₂ -FLU
^{s,w} 22R-17α(H),21β(H)-30,31,32,33,34-pentakishomohopane	C ₃₅ (R) H	^{s,w,a} C ₃ -Fluorenes	C ₃ -FLU
^{s,w} C ₂₇ 20-5α(H),14β(H),17β(H)-cholestane	C ₂₇ αββ H	^{s,w,a} C ₀ -Fluoroanthene/pyrene	C ₀ -FLA
^{s,w} C ₂₈ 20-5α(H),14β(H),17β(H)-ergostane	C ₂₈ αββ H	^{s,w,a} C ₁ -Fluoroanthenes/pyrenes	C ₁ -FLA
^{s,w} C ₂₉ 20-5α(H),14β(H),17β(H)-stigmastane	C ₂₉ αββ H	^{s,w,a} C ₂ -Fluoroanthenes/pyrenes	C ₂ -FLA
		^{s,w,a} C ₃ -Fluoroanthenes/pyrenes	C ₃ -FLA
		^{s,w,a} C ₄ -Fluoroanthenes/pyrenes	C ₄ -FLA
		^{s,w} C ₀ -Benzonaphthothiophenes	C ₀ -BNT
		^{s,w} C ₁ -Benzonaphthothiophenes	C ₁ -BNT
		^{s,w} C ₂ -Benzonaphthothiophenes	C ₂ -BNT
		^{s,w} C ₃ -Benzonaphthothiophenes	C ₃ -BNT
		^{s,w} C ₄ -Benzonaphthothiophenes	C ₄ -BNT
		^{s,w,a} C ₀ -Chrysene	C ₀ -CHR
		^{s,w,a} C ₁ -Chrysenes	C ₁ -CHR
		^{s,w,a} C ₂ -Chrysenes	C ₂ -CHR
		^{s,w,a} C ₃ -Chrysenes	C ₃ -CHR
		^{s,w} Biphenyl	Bph
		^{s,w,a} Acenaphthylene	AcI
		^{s,w,a} Acenaphthene	Ace
		^{s,w,a} Anthracene	An
		^{s,w,a} Pyrene	Py
		^{s,w,a} Benz(a)anthracene	BaA
		^{s,w,a} Benzo(b)fluoranthene	BbF
		^{s,w,a} Benzo(k)fluoranthene	BkF
		^{s,w} Benzo(e)pyrene	BeP
		^{s,w,a} Benzo(a)pyrene	BaP
		^{s,w} Perylene	Pe
		^{s,w,a} Indeno(1,2,3-cd)pyrene	IP
		^{s,w,a} Dibenzo(a,h)anthracene	DA
		^{s,w,a} Benzo(g,h,i)perylene	BgP

Table S2. Mean percent recoveries (\pm standard deviation) for deuterated compounds in water, sediment, and air samples

Compound	% Recovery (\pm sd)		
	Water Samples	Sediment Samples	Air Samples
d ₈ -naphthalene	71.4 \pm 16.7	74.2 \pm 13.8	97.4 \pm 16.6
d ₁₀ -Acenaphthene	80.1 \pm 16.5	89.2 \pm 12.9	83.0 \pm 8.0
d ₁₀ -Phenanthrene	91.8 \pm 18.0	97.2 \pm 11.8	69.7 \pm 3.0
d ₁₂ -Benz(a)anthracene	92.4 \pm 18.7	100.0 \pm 14.0	53.1 \pm 4.0
d ₁₂ -Perylene	81.6 \pm 16.4	103.7 \pm 9.5	52.3 \pm 5.7

Table S3. Maximum concentrations of 7 different PAC groups measured in our study compared to other studies using diluted bitumen at similar oil:water ratios. The maximum concentration observed for each PAC group is presented as well as the day in which this concentration was reached during the study (T_{max}).

PAC Group	Our Study				Ortmann et al. (2020) ^a						Stoyanovich et al. (2019) ^b	
	CLB 1:10000 (18 L)		CLB 1:4600 (42 L)		AWB 1:6400		Synbit 1:6400		WCS 1:6400		CLB 1:8000	
	Max Conc (ng/L)	T_{max} (d)	Max Conc (ng/L)	T_{max} (d)	Max Conc (ng/L)	T_{max} (d)	Max Conc (ng/L)	T_{max} (d)	Max Conc (ng/L)	T_{max} (d)	Max Conc (ng/L)	T_{max} (d)
Σ NAP _{C0-C4}	299	8	619	15	1466	2	2125	2	3196	2	1005	4
Σ PHEN _{C0-C4}	197	8	284	28	26.76	4	2719	10	1974	10	362	4
Σ DBT _{C0-C3}	182	8	293	2	0	0	1550	10	1431	10	89	7
Σ FLU _{C0-C3}	280	15	466	15	130.5	4	1508	10	1021	10	251	4
Σ FLA _{C0-C4}	27.5	8	34.5	8	-	-	-	-	-	-	12	4
Σ BNT _{C0-C4}	45.9	8	61.1	8	12.14	14	1190	10	1058	10	n.d. ^c	n.d.
Σ CHR _{C0-C3}	17.3	8	21.2	8	0	0	766	10	444	10	5	4

^aOrtmann et al. (2020) spilled 3 different types of diluted bitumen (Access Wester Blend; AWB, Synbit, and Western Canadian Select; WCS) into enclosures containing seawater during the summer and sampled the water column for 14 days following the spill.

^bStoyanovich et al. (2019) spilled Cold Lake Blend (CLB) into tanks containing freshwater during the summer and samples the water column for 11 days following the spill.

^cn.d. represents concentration below method detection limits

Table S4. Comparison between sediment jar and oiled sediment core data with fold change presented in brackets.

PAC Group	Jar Sample Max Jar ^a	Oiled Core Sample					
		TB-18-1	TB-18-2	TB-18-3	TB-82-1	TB-82-2	TB-82-3
ΣPAC ₄₆	189	14646 (77X)	1696.2 (9X)	587.2 (3X)	379.2 (2X)	4708.6 (25X)	532.3 (3X)
ΣNAP _{C0-C4}	22.8	263 (12X)	78.9 (3X)	84.1 (4X)	25.8 (1X)	343.6 (15X)	63.5 (3X)
ΣPHEN _{C0-C4}	50.7	4144 (82X)	581.2 (11X)	136.1 (3X)	150.8 (3X)	1240.5 (24X)	145.0 (3X)
ΣDBT _{C0-C3}	31.9	3684 (115X)	343.6 (11X)	127.9 (4X)	62.8 (2X)	1016.6 (32X)	115.4 (4X)
ΣFLU _{C0-C3}	19.6	929 (47X)	88.7 (5X)	50.8 (3X)	22.5 (1X)	254.1 (13X)	44.8 (2X)
ΣFLA _{C0-C4}	9.65	859 (89X)	76.2 (8X)	22.6 (2X)	16.1 (2X)	222.4 (23X)	23.4 (2X)
ΣBNT _{C0-C4}	45.8	3760 (82X)	410.9 (9X)	127.4 (3X)	75.7 (2X)	1301.0 (28X)	104.9 (2X)
ΣCHR _{C0-C3}	10.5	788 (75X)	87.9 (8X)	25.3 (2X)	15.5 (1X)	249.1 (24X)	25.3 (2X)

^aThe maximum jar values were observed in the 18 L treatment for each PAC group except for ΣFLU_{C0-C3} and ΣNAP_{C0-C4} which were observed in the 180 L treatment, all on day 70.

Table S5. Water column regression coefficients obtained by regressing the log₁₀ transformed concentration of each respective PAC group against the log₁₀ transformed oil:water ratio (alpha = 0.05). Certain compounds remained below detection limit during the earlier timepoints of the study and therefore did not yield sufficient regression coefficients, denoted by “NA”.

Compound group	Pre-Spill to 8 days						15 to 70 days					
	Log ₁₀ [PAC] _i = m * Log ₁₀ [oil:water] + b						Log ₁₀ [PAC] _i = m * Log ₁₀ [oil:water] + b					
	Time	m	b	R ²	p	Sig	Time	m	b	R ²	p	Sig
ΣPAC ₄₆	-3 days	-0.10	0.78	0.07	0.553	no	15 days	0.23	3.94	0.75	0.012	*
NAP (C0-C4)		-0.11	0.36	0.03	0.722	No		0.38	3.98	0.95	0.0002	***
PHEN (C0-C4)		-0.09	-0.13	0.05	0.619	No		0.09	2.69	0.34	0.168	No
DBT (C0-C3)		0.00	0.00	NA	NA	No		0.10	2.68	0.19	0.322	No
FLU (C0-C3)		-0.04	-0.12	0.17	0.357	No		0.28	3.54	0.79	0.008	*
FLA (C0-C4)		0.00	0.00	NA	NA	No		0.05	1.19	0.01	0.795	No
BNT (C0-C4)		0.00	0.00	NA	NA	No		0.51	3.04	0.59	0.045	*
CHR (C0-C3)		0.07	0.35	0.05	0.634	No		0.12	1.33	0.78	0.009	*
ΣPAC ₄₆	0.25 days	0.64	3.75	0.50	0.076	No	22 days	0.35	4.26	0.65	0.029	*
NAP (C0-C4)		0.65	3.53	0.62	0.034	*		0.48	4.24	0.84	0.004	**
PHEN (C0-C4)		0.35	1.99	0.18	0.339	No		0.23	3.03	0.53	0.062	No
DBT (C0-C3)		0.64	2.81	0.66	0.027	*		0.19	2.85	0.25	0.256	No
FLU (C0-C3)		0.40	2.16	0.25	0.254	No		0.40	3.84	0.63	0.033	*
FLA (C0-C4)		0.00	0.00	NA	NA	No		0.40	2.15	0.94	0.0003	***
BNT (C0-C4)		0.00	0.00	NA	NA	No		0.47	2.62	0.77	0.010	*
CHR (C0-C3)		0.00	0.00	NA	NA	No		0.13	1.19	0.72	0.015	*
ΣPAC ₄₆	1 day	0.27	3.67	NA	0.006	*	28 days	0.44	4.50	0.66	0.027	*
NAP (C0-C4)		0.38	3.85	0.89	0.002	**		0.50	4.28	0.79	0.007	*
PHEN (C0-C4)		0.07	2.02	0.24	0.269	No		0.34	3.36	0.56	0.052	No
DBT (C0-C3)		0.10	2.14	0.21	0.304	No		0.31	3.26	0.41	0.123	No
FLU (C0-C3)		0.15	2.22	0.76	0.011	*		0.50	4.14	0.63	0.033	*
FLA (C0-C4)		-0.03	-0.04	0.21	0.303	No		0.45	2.24	0.89	0.001	**
BNT (C0-C4)		-0.01	-0.03	0.35	0.160	No		0.52	2.46	0.70	0.018	*

CHR (C0-C3)		-0.07	-0.21	0.53	0.062	No		0.22	1.42	0.93	0.0004	***
Σ PAC ₄₆	2 days	0.21	3.25	0.12	0.442	No	42 days	0.35	4.02	0.67	0.025	*
NAP (C0-C4)		0.14	2.66	0.04	0.661	No		0.39	3.75	0.88	0.002	**
PHEN (C0-C4)		0.27	2.49	0.09	0.509	No		0.22	2.76	0.43	0.107	No
DBT (C0-C3)		0.44	3.33	0.20	0.313	No		0.30	3.03	0.56	0.054	No
FLU (C0-C3)		0.42	3.10	0.23	0.277	No		0.42	3.69	0.50	0.074	No
FLA (C0-C4)		-0.14	-0.36	0.21	0.298	No		0.47	2.37	0.64	0.030	*
BNT (C0-C4)		-0.02	-0.07	0.35	0.160	No		0.70	3.20	0.80	0.007	*
CHR (C0-C3)		-0.02	0.01	0.03	0.720	No		0.34	1.86	0.76	0.011	*
Σ PAC ₄₆		4 days	-0.03	2.44	0.02	0.769		No	56 days	0.60	4.77	0.97
NAP (C0-C4)	0.08		2.40	0.20	0.309	No	0.64	4.57		0.95	0.0002	***
PHEN (C0-C4)	-0.11		1.38	0.19	0.322	No	0.43	3.44		0.94	0.0003	***
DBT (C0-C3)	-0.19		1.05	0.27	0.228	No	0.56	3.73		0.98	0.00002	****
FLU (C0-C3)	0.06		2.20	0.14	0.415	No	0.66	4.31		0.87	0.002	**
FLA (C0-C4)	0.18		1.03	0.72	0.016	No	0.47	2.10		0.79	0.008	*
BNT (C0-C4)	-0.25		-0.06	0.21	0.305	No	0.57	2.54		0.88	0.002	**
CHR (C0-C3)	-0.20		-0.33	0.36	0.157	No	0.32	1.58		0.97	0.00004	****
Σ PAC ₄₆	8 days		0.14	3.58	0.63	0.033	*	70 days		0.51	4.50	0.78
NAP (C0-C4)		0.26	3.47	0.92	0.001	**	0.61		4.39	0.90	0.001	**
PHEN (C0-C4)		0.08	2.66	0.50	0.075	No	0.47		3.58	0.80	0.006	*
DBT (C0-C3)		0.03	2.46	0.02	0.736	No	0.42		3.31	0.63	0.033	*
FLU (C0-C3)		0.15	2.96	0.69	0.021	*	0.54		4.00	0.57	0.048	*
FLA (C0-C4)		0.22	2.26	0.87	0.002	**	0.67		2.96	0.77	0.010	*
BNT (C0-C4)		0.13	2.19	0.41	0.122	No	0.87		3.87	0.77	0.009	*
CHR (C0-C3)		0.24	2.20	0.68	0.023	*	0.54		2.53	0.88	0.002	**

Table S6. Sediment regression coefficients obtained by regressing the log₁₀ transformed concentration of each respective PAC group against the log₁₀ transformed oil:water ratio (alpha = 0.05). Certain compounds remained below detection limit during the earlier timepoints of the study and therefore did not yield sufficient regression coefficients, denoted by “NA”.

Compound group	Pre-Spill to 15 days						22 to 70 days					
	Log ₁₀ [PAC] _i = m * Log ₁₀ [oil:water] + b						Log ₁₀ [PAC] _i = m * Log ₁₀ [oil:water] + b					
	Time	m	b	R ²	p	Sig	Time	m	b	R ²	p	Sig
ΣPAC ₄₆	-3 days	0.01	1.44	0.01	0.831	No	22 days	0.16	2.31	0.18	0.404	No
NAP (C0-C4)		0.00	0.96	0.0001	0.985	No		0.08	1.25	0.18	0.407	No
PHEN (C0-C4)		0.02	1.10	0.04	0.676	No		0.09	1.49	0.09	0.555	No
DBT (C0-C3)		0.01	0.12	0.12	0.439	No		0.19	1.73	0.31	0.253	No
FLU (C0-C3)		0.00	0.19	0.002	0.918	No		0.20	1.41	0.20	0.368	No
FLA (C0-C4)		0.02	0.48	0.09	0.517	No		0.18	1.28	0.33	0.230	No
BNT (C0-C4)		0.00	0.00	NA	NA	No		0.29	2.03	0.18	0.406	No
CHR (C0-C3)		0.01	0.16	0.25	0.252	No		0.11	0.90	0.11	0.524	No
ΣPAC ₄₆	1 day	0.02	1.60	0.03	0.698	No	28 days	0.07	1.95	0.08	0.541	No
NAP (C0-C4)		0.01	1.18	0.01	0.876	No		0.13	1.44	0.56	0.052	No
PHEN (C0-C4)		0.03	1.26	0.15	0.389	No		0.00	1.17	0.00002	0.993	No
DBT (C0-C3)		0.01	0.13	0.05	0.619	No		0.16	1.50	0.13	0.419	No
FLU (C0-C3)		0.00	0.25	0.001	0.943	No		0.15	1.12	0.32	0.187	No
FLA (C0-C4)		0.01	0.46	0.04	0.651	No		0.09	0.81	0.31	0.195	No
BNT (C0-C4)		0.00	0.00	NA	NA	No		0.09	1.16	0.02	0.736	No
CHR (C0-C3)		0.00	0.11	0.05	0.621	No		-0.06	0.26	0.06	0.610	No
ΣPAC ₄₆	2 days	-0.11	0.78	0.09	0.520	No	42 days	0.12	2.28	0.12	0.448	No
NAP (C0-C4)		-0.12	0.36	0.12	0.449	No		0.13	1.47	0.70	0.020	*
PHEN (C0-C4)		-0.11	0.30	0.09	0.508	No		0.07	1.54	0.06	0.593	No
DBT (C0-C3)		0.00	0.00	NA	NA	No		0.17	1.69	0.20	0.308	No
FLU (C0-C3)		-0.03	0.16	0.01	0.843	No		0.21	1.52	0.41	0.124	No
FLA (C0-C4)		-0.06	0.07	0.12	0.451	No		0.16	1.23	0.31	0.193	No
BNT (C0-C4)		0.00	0.00	NA	NA	No		0.09	1.39	0.01	0.795	No

CHR (C0-C3)		-0.01	0.03	0.07	0.566	No		0.11	1.07	0.10	0.484	No
ΣPAC ₄₆	8 days	0.00	1.17	0.00003	0.991	No	70 days	0.21	2.76	0.53	0.063	No
NAP (C0-C4)		-0.02	0.67	0.01	0.874	No		0.09	1.59	0.83	0.004	**
PHEN (C0-C4)		0.01	0.77	0.001	0.958	No		0.17	2.05	0.42	0.114	No
DBT (C0-C3)		0.00	0.00	NA	NA	No		0.29	2.18	0.55	0.058	No
FLU (C0-C3)		0.00	0.09	0.0002	0.977	No		0.17	1.71	0.54	0.061	No
FLA (C0-C4)		0.01	0.33	0.01	0.822	No		0.20	1.50	0.52	0.068	No
BNT (C0-C4)		0.00	0.00	NA	NA	No		0.42	2.67	0.56	0.053	No
CHR (C0-C3)		0.00	0.09	0.01	0.847	No		0.19	1.43	0.34	0.171	No
ΣPAC ₄₆		15 days	-0.04	1.41	0.24	0.267		No				
NAP (C0-C4)	0.01		1.01	0.002	0.928	No						
PHEN (C0-C4)	-0.04		0.88	0.27	0.237	No						
DBT (C0-C3)	-0.04		0.58	0.16	0.374	No						
FLU (C0-C3)	0.00		0.47	0.001	0.954	No						
FLA (C0-C4)	0.00		0.39	0.003	0.903	No						
BNT (C0-C4)	-0.11		0.16	0.46	0.096	No						
CHR (C0-C3)	-0.04		0.04	0.19	0.335	No						

Table S7. CCME guidelines for protection of aquatic life in water. Only PACs with available data are presented. Max concentrations presented across range of dilbit treatments.

Compound	Conc (ng/L)	Maximum concentrations observed in timeseries samples of present study (ng/L)
Acenaphthene	5800	3.22 – 10.8
Anthracene	12	2.10 – 9.69
Benz(a)anthracene	18	0.72 – 2.14
Benzo(a)pyrene	15	0.40 – 3.41
Fluoranthene	40	1.74 – 5.40
Fluorene	3000	5.98 – 20.0
Naphthalene	1100	8.10 – 98.2
Phenanthrene	400	10.9 – 35.2
Pyrene	25	1.56 – 13.3

Table S8. CCME guidelines for protection of aquatic life in sediments. Only PACs with available data are presented. Max concentrations presented across range of dilbit treatments.

Compound	ISQG ^a (ng/g d.w.)	PEL ^a (ng/g d.w.)	Maximum concentrations observed in timeseries samples of present study (ng/g d.w.)
2-Methylnaphthalene	20.2	201	2.54 – 4.63
Acenaphthene	6.71	88.9	0.15 – 0.26
Acenaphthylene	5.87	128	0.097 – 0.18
Anthracene	46.9	245	0.26 – 0.38
Benz(a)anthracene	31.7	385	0.18 – 0.31
Benzo(a)pyrene	31.9	782	0.064 – 0.15
Chrysene	57.1	862	0.51 – 1.43
Dibenz(a,h)anthracene	6.22	135	0.025 – 0.10
Fluoranthene	111	2355	1.39 – 2.02
Fluorene	21.2	144	0.69 – 1.20
Naphthalene	34.6	391	1.54 – 3.23
Phenanthrene	41.9	515	3.86 – 5.67
Pyrene	53	875	1.09 – 1.89

^aISQG: Interim Sediment Quality Guideline

^bPEL: Probable Effect Level

Figures

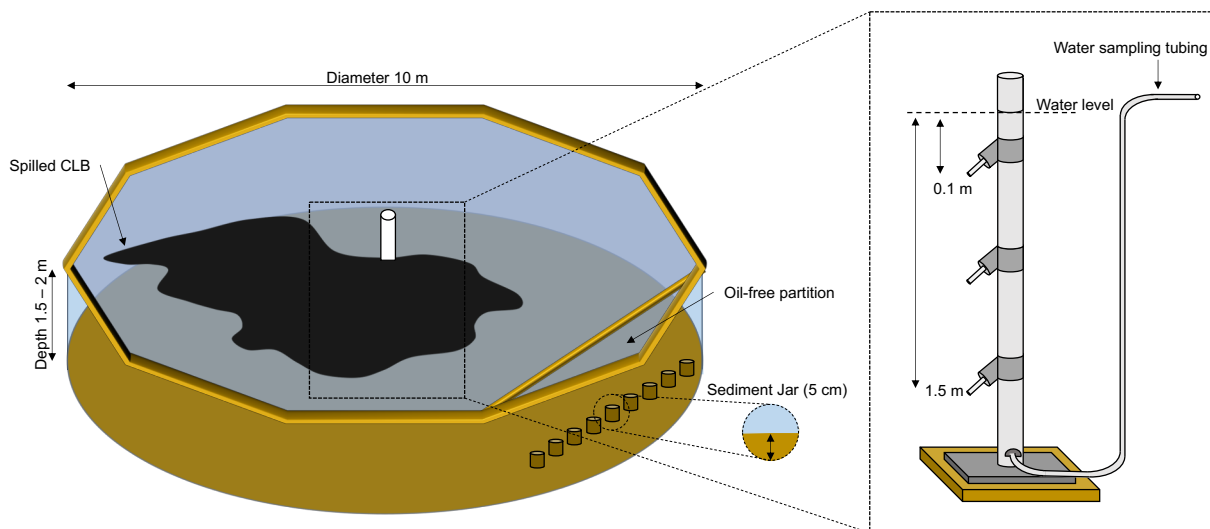


Figure S1. Schematic of limnocorral set up showing water and sediment sampling methods.



Figure S2. The addition of 180 L of CLWB (1:500 oil:water, v/v) onto the water surface of the limnocorral. (A) Overview of addition method and visible sandbags used to seal the membrane to the sediments, (B) View of telescopic arm and pumping system being held at approximately 5–10 cm from the water's surface. The oil exited the hose parallel to the water's surface to avoid forcing oil below the water surface and allowing it to spread out during the addition.

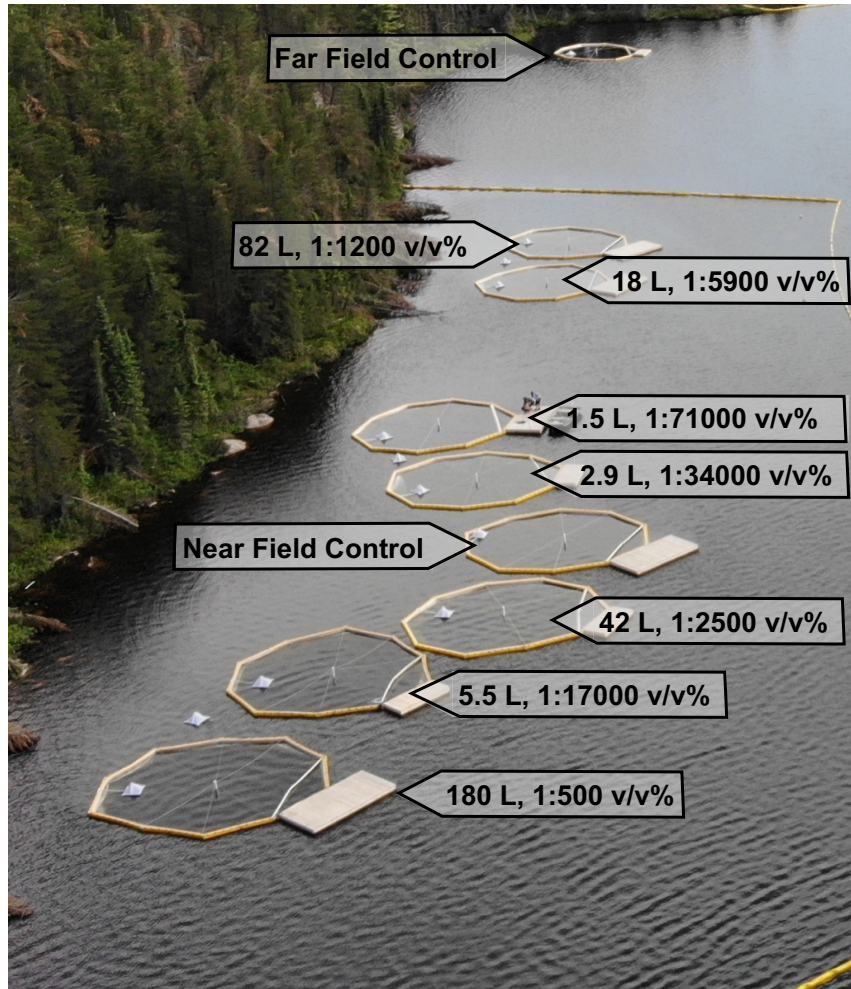


Figure S3. Distribution of spill volumes and resulting oil:water ratios



Figure S4. Example of a tarball sitting on top of the sediment in a coring tube. The tarball was removed and then the top 1cm of the core was processed for PAC analysis.

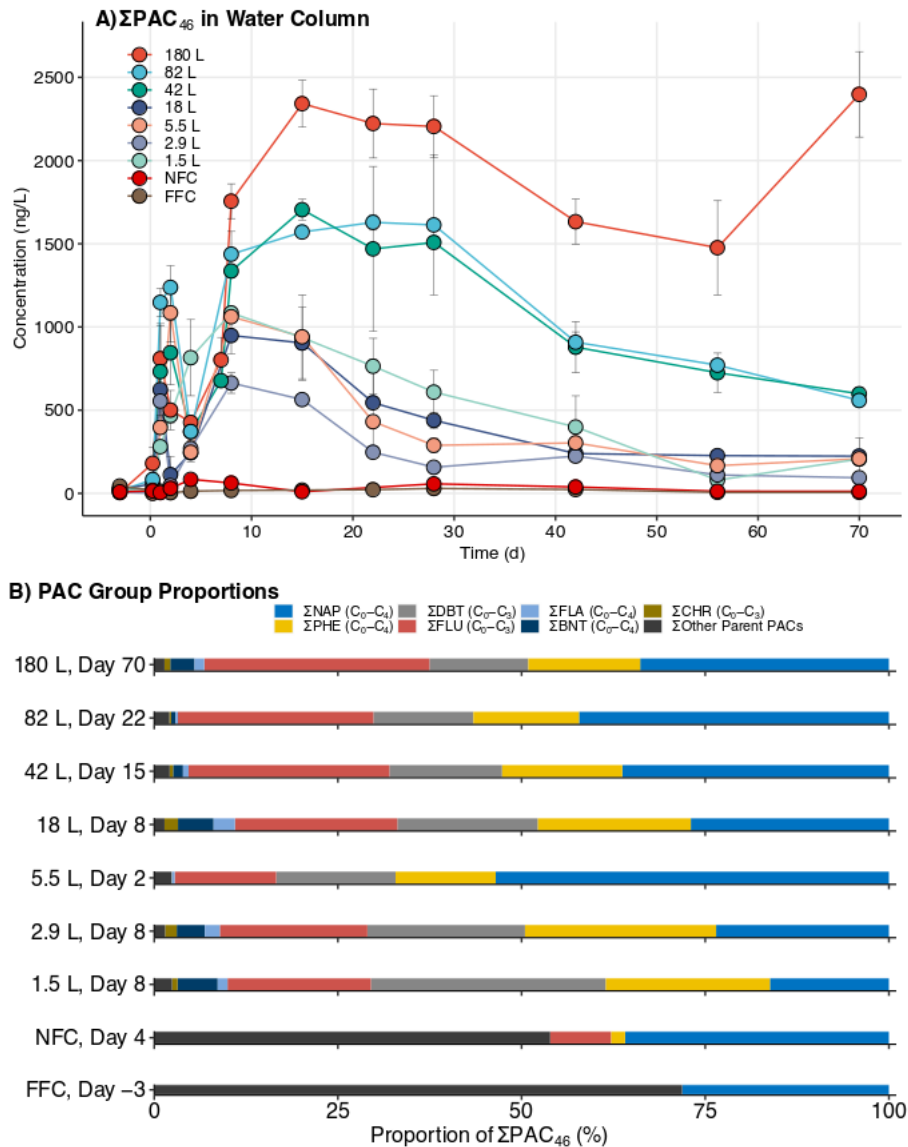


Figure S5. Temporal progression of mean ΣPAC_{46} concentrations in the water column, error bars represent standard deviation (A) and the resulting proportions of each PAC group during the period of maximum ΣPAC_{46} concentration in each treatment (B). ΣPAC_{46} concentrations have been previously reported by Rodriguez-Gil et al. (2021). “ Σ Other Parent PACs” represents the sum of Biphenyl, Acenaphthylene, Acenaphthene, Anthracene, Pyrene, Benz(a)anthracene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(e)pyrene, Benzo(a)pyrene, Perylene, Ideno(1,2,3-cd)pyrene, Dibenzo(a,h)anthracene, and Benzo(g,h,i)perylene.

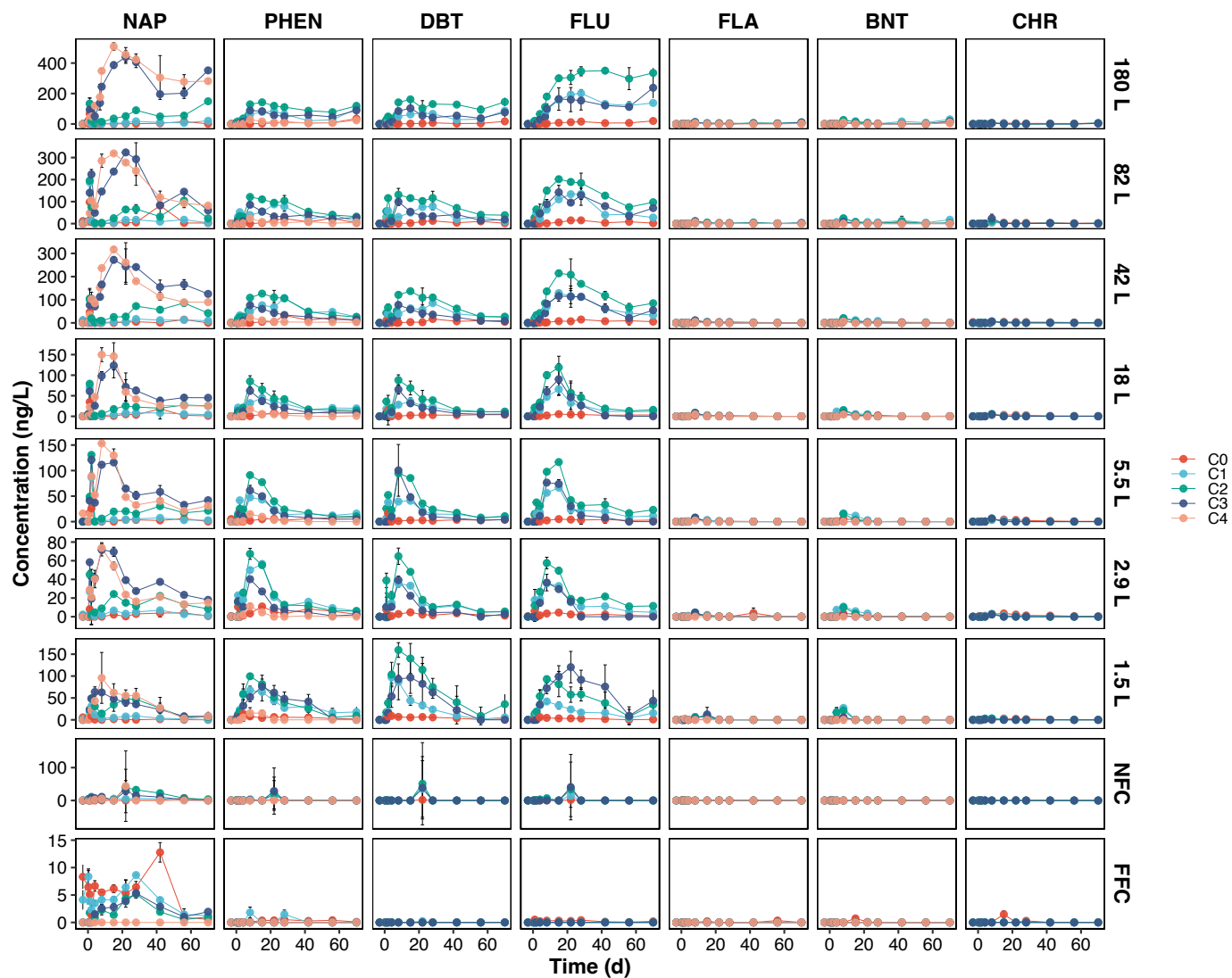


Figure S6. Temporal progression of alkylated PAC concentrations in the water columns of each treatment and control, across each parent PAC group. The error bars represent standard deviation about the mean.

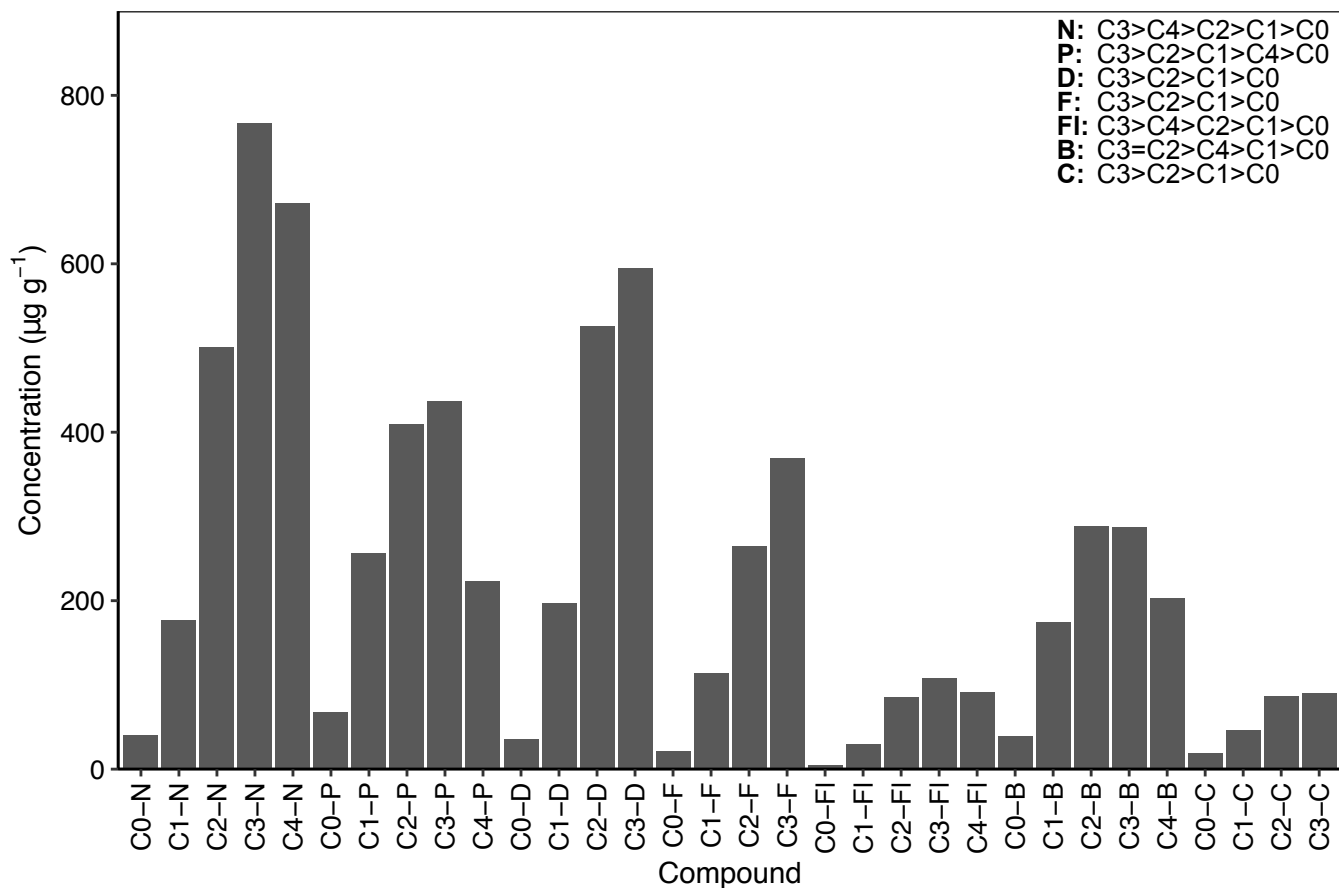


Figure S7. Distribution of parent and alkylated PACs in the source CLWB dilbit. The upper right quadrant of the plot shows the within group abundance distributions of each alkylated compound. N: naphthalene, P: phenanthrene, D: dibenzothiophene, F: fluorene, Fl: fluoranthene, B: benzonaphthothiophene, and C: chrysene.

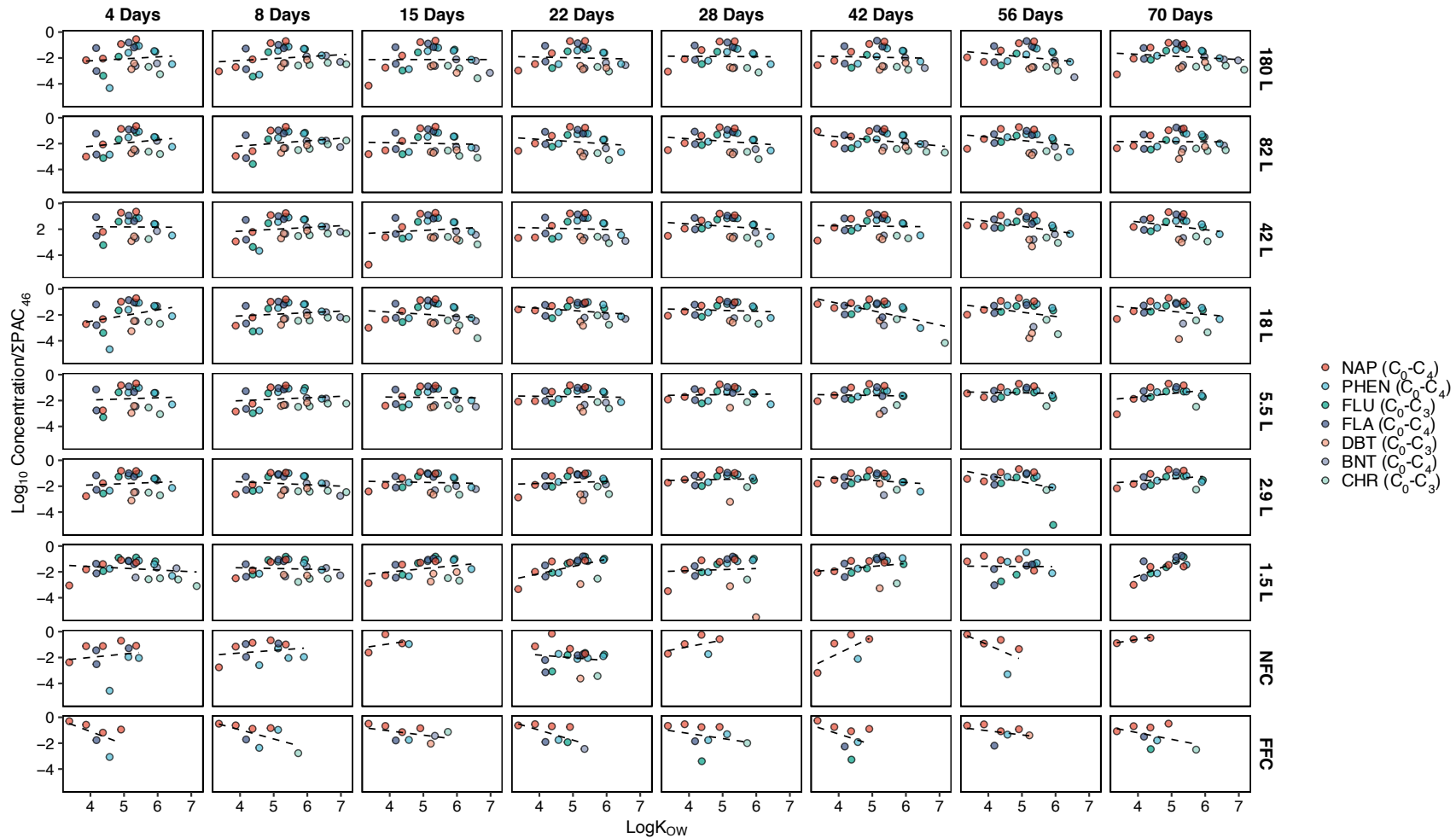


Figure S8. The relationship between water column concentration of individual PACs and $\log K_{ow}$. The dashed lines represent linear regression outputs. Individual compound concentrations were normalized to the ΣPAC_{46} concentration of each respective sample. Compounds whose concentrations were below MDL were omitted from this analysis.

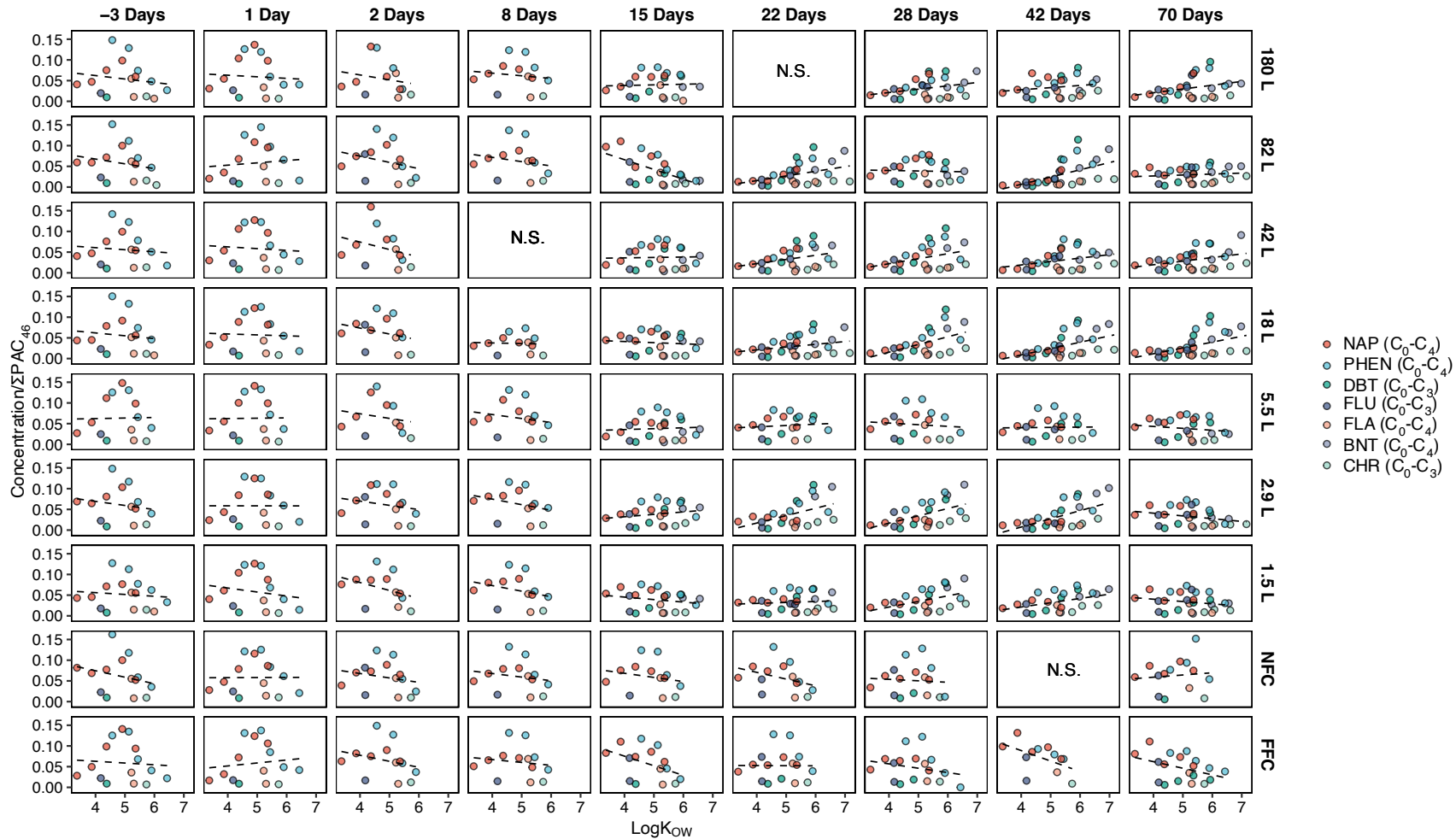


Figure S9. The relationship between sediment concentration of individual PACs and $\log K_{ow}$. The dashed lines represent linear regression outputs. Individual compound concentrations were normalized to the ΣPAC_{46} concentration of each respective sample. Compounds whose concentrations were below MDL were omitted from this analysis.

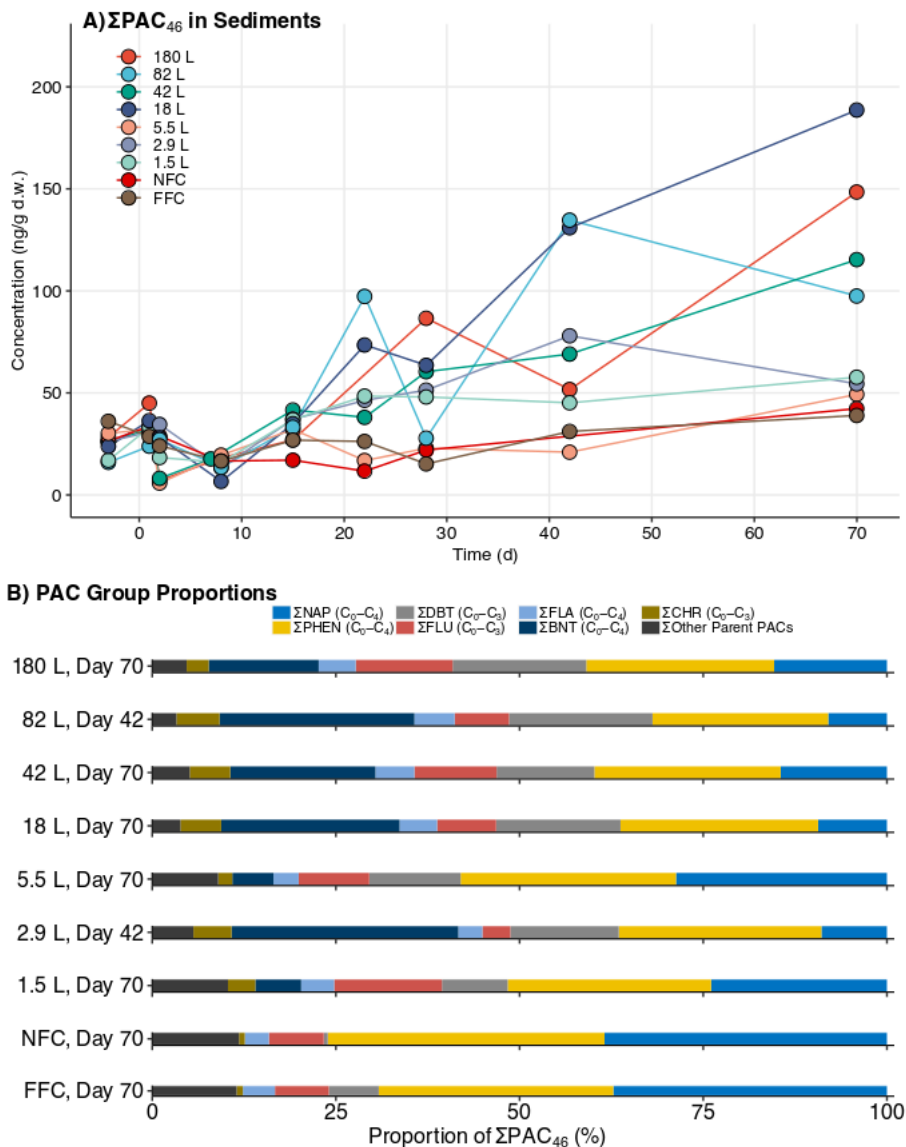


Figure S10. Temporal progression of mean Σ PAC₄₆ concentrations in the sediments (A) and the resulting proportions of each PAC group during the period of maximum Σ PAC₄₆ concentration in each treatment (B). Σ PAC₄₆ concentrations have been previously reported by Rodriguez-Gil et al. (2021). “ Σ Other Parent PACs” represents the sum of Biphenyl, Acenaphthylene, Acenaphthene, Anthracene, Pyrene, Benz(a)anthracene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(e)pyrene, Benzo(a)pyrene, Perylene, Ideno(1,2,3-cd)pyrene, Dibenzo(a,h)anthracene, and Benzo(g,h,i)perylene.

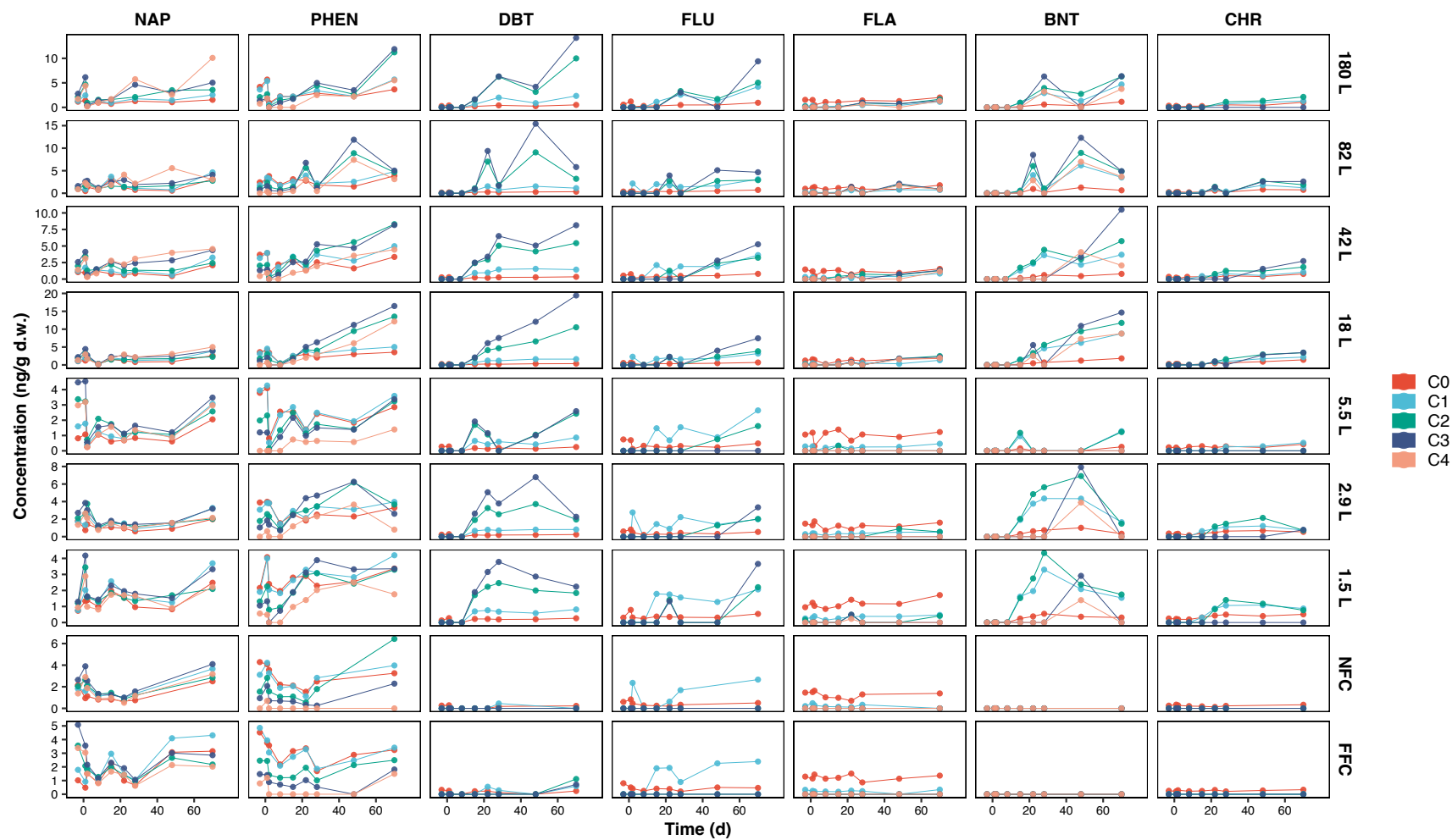


Figure S11. Temporal progression of alkylated PAC concentrations in sediment samples collected from each treatment and control, across each parent PAC group.

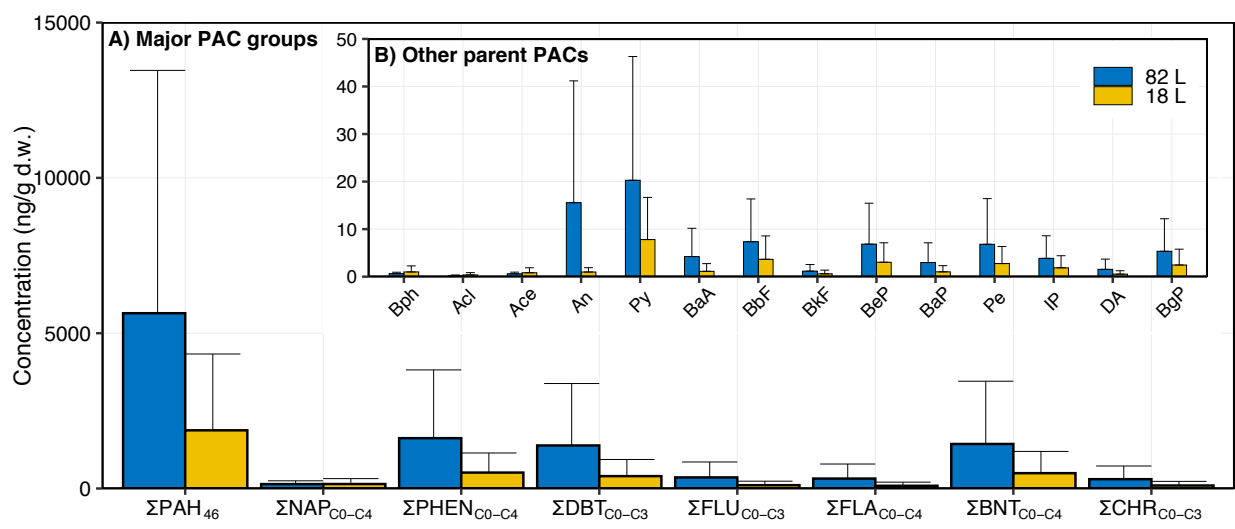


Figure S12. Mean concentrations of polycyclic aromatic compound (PAC) groups in sediment samples in direct contact with sunken dilbit. The sum of the parent and each alkylated homolog in each group is in (A). Other parent PACs are in panel (B). Error bars are standard deviations (N= 3)

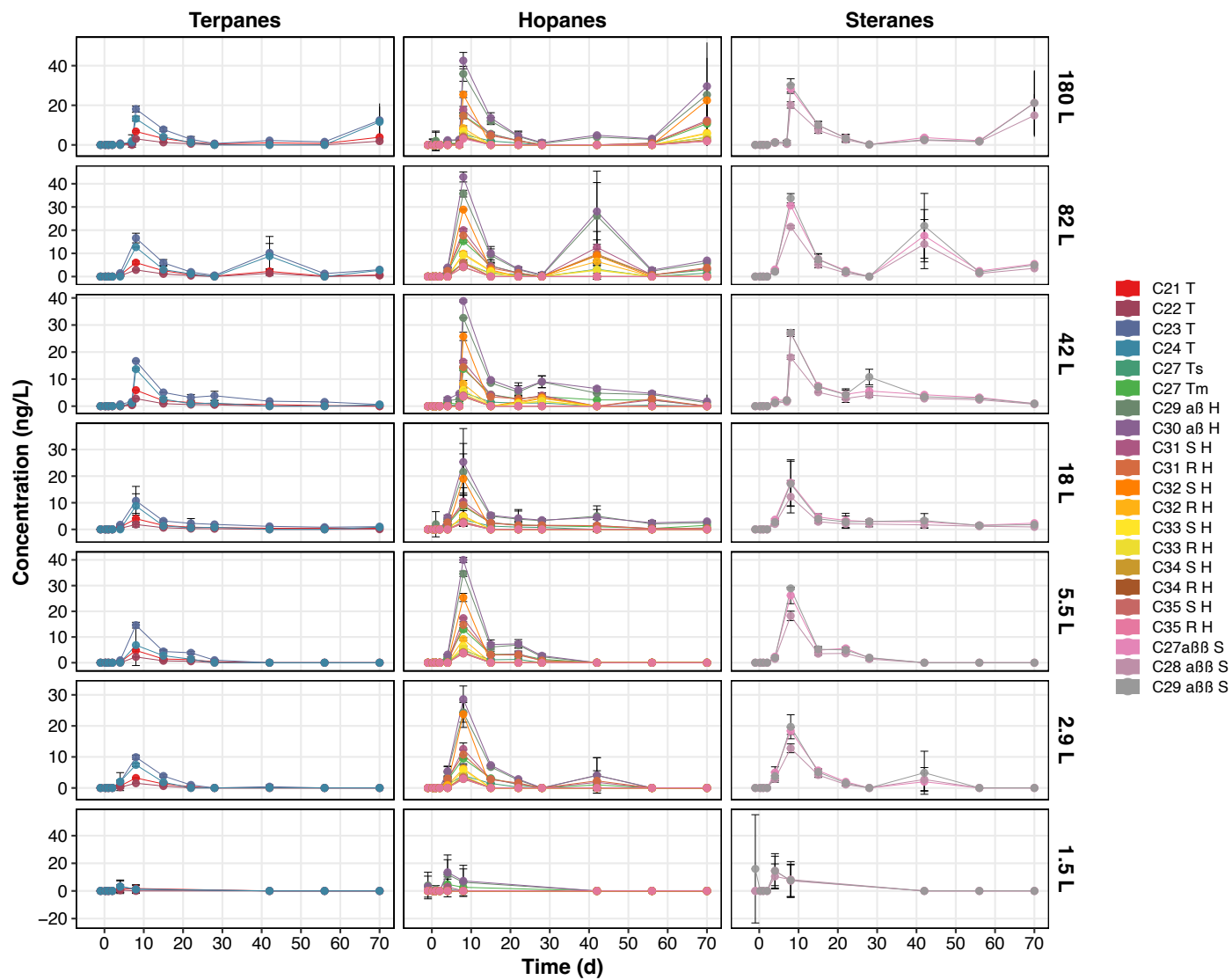


Figure S13. Temporal progression of petroleum biomarker compound concentrations (mean \pm standard deviation) in the water column.

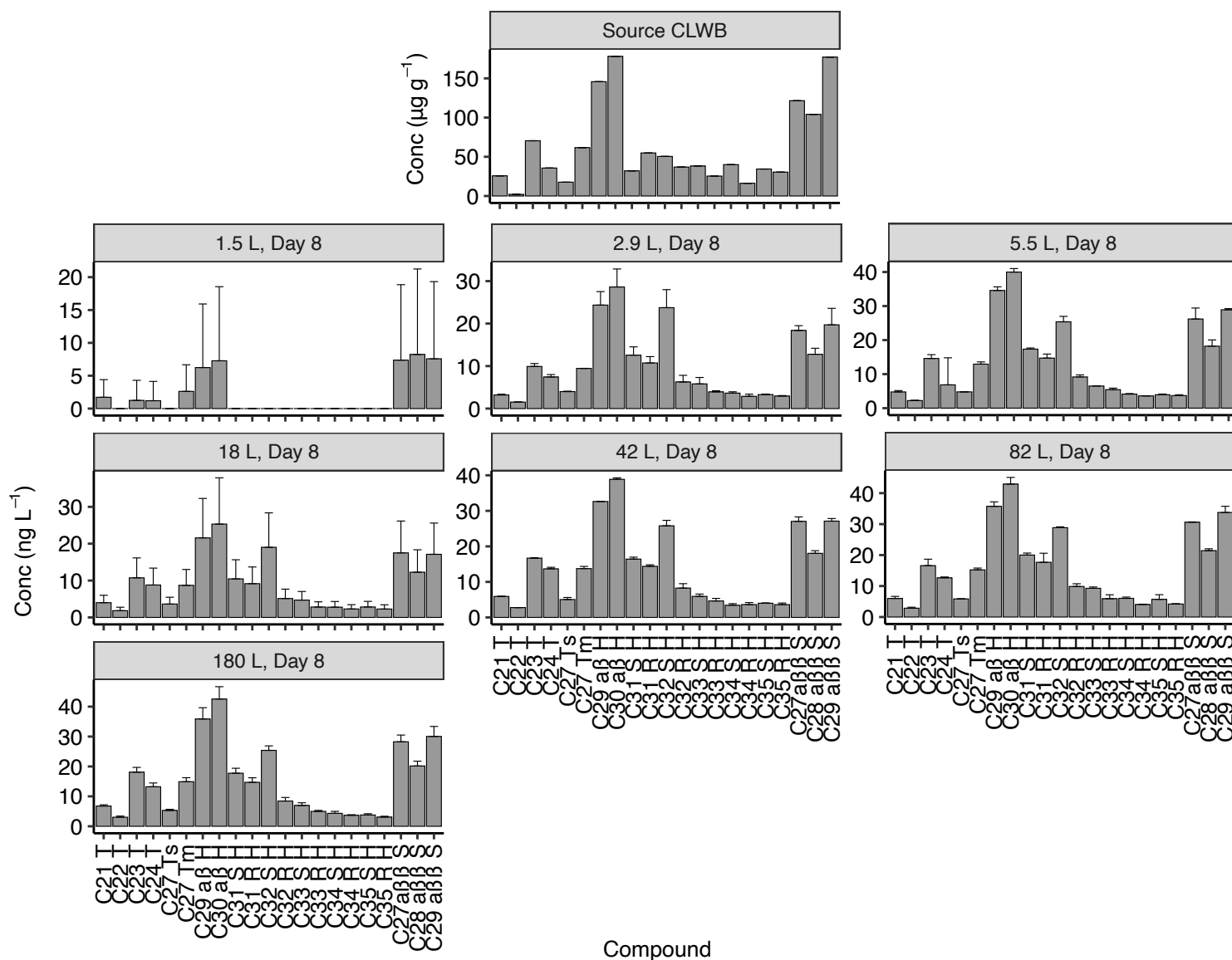


Figure S14. Concentration profiles of petroleum biomarkers in the source CLWB and water samples collected on day 8 from each dilbit amended treatment. Error bars represent standard deviation about the mean.

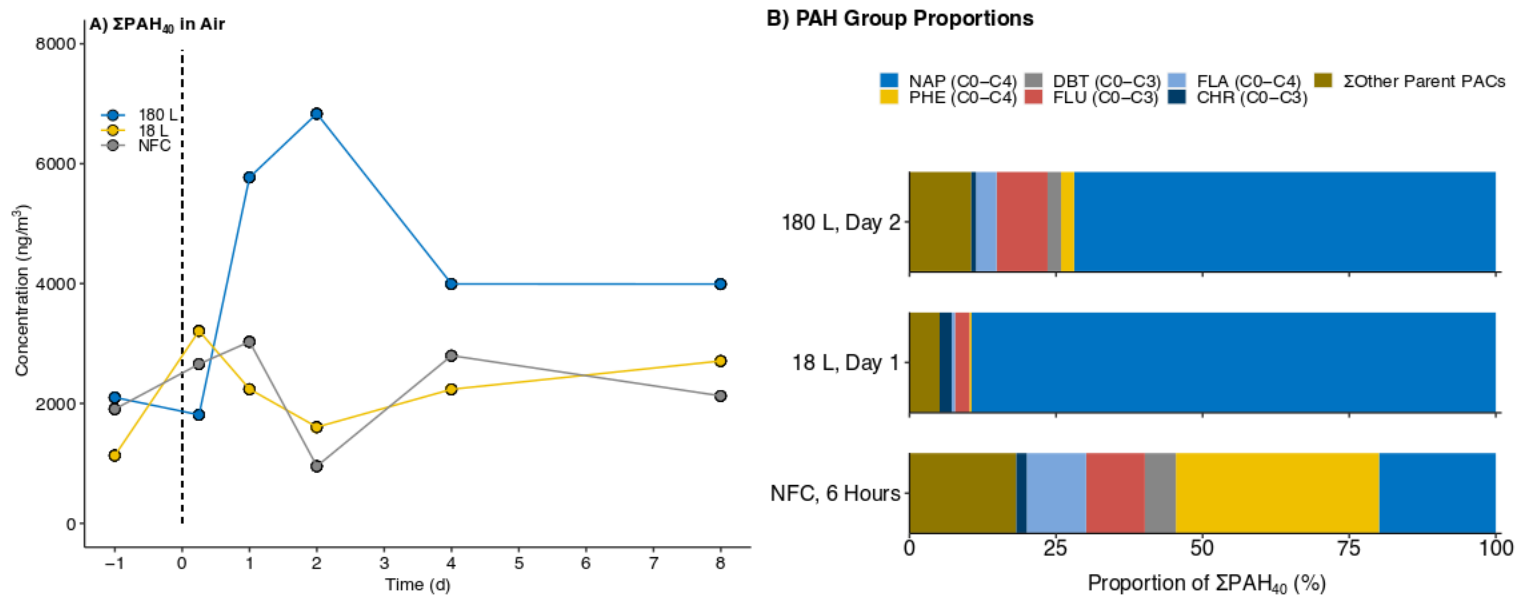


Figure S15. Temporal progression of ΣPAC₄₀ concentrations in the sediments (A) and the resulting proportions of each PAC group during the period of maximum ΣPAC₄₀ concentration in each treatment (B). “Σ Other Parent PACs” represents the sum of Biphenyl, Acenaphthylene, Acenaphthene, Anthracene, Pyrene, Benz(a)anthracene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(e)pyrene, Benzo(a)pyrene, Perylene, Ideno(1,2,3-cd)pyrene, Dibenzo(a,h)anthracene, and Benzo(g,h,i)perylene.

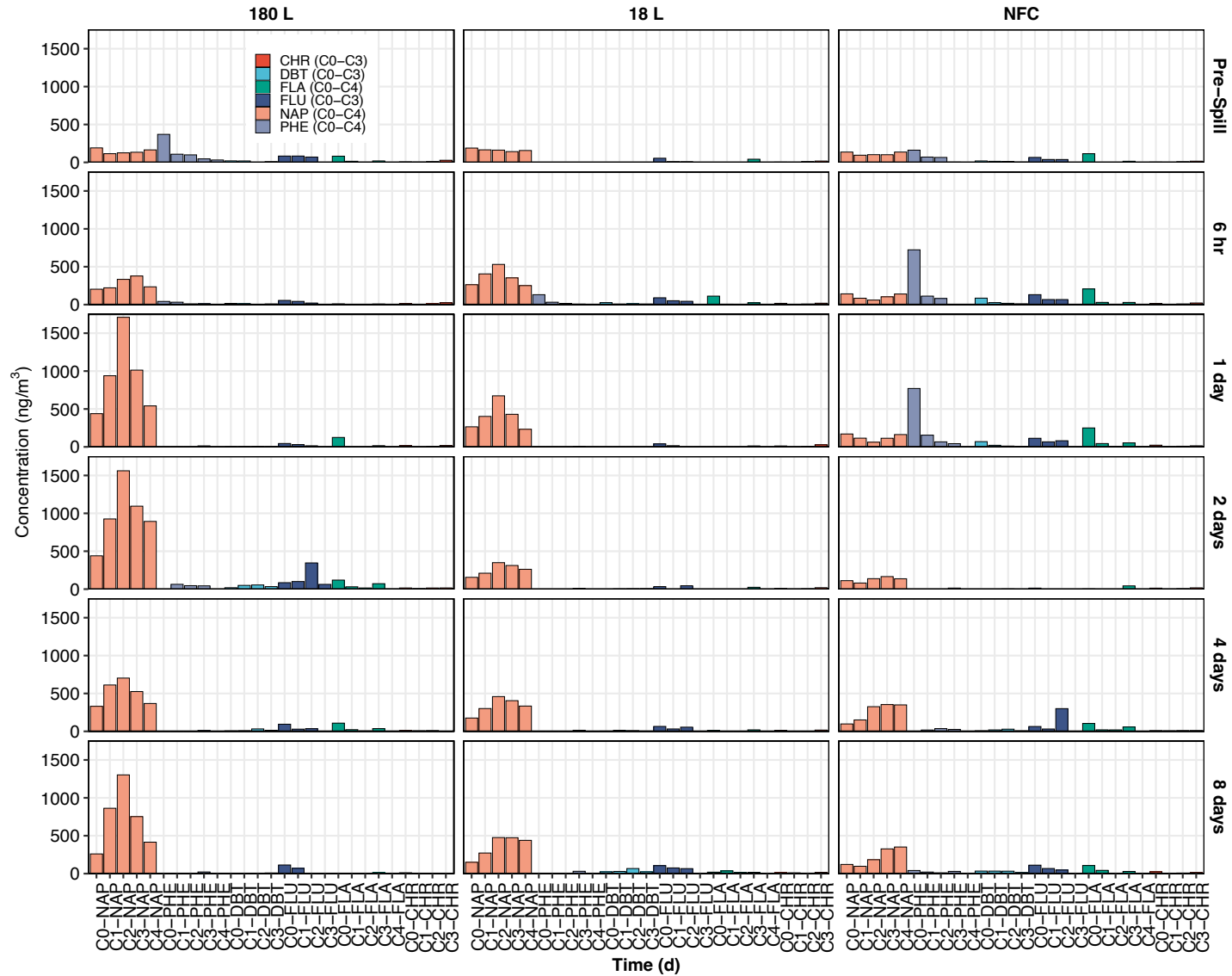


Figure S16. Alkylated PAC profiles of each air sample collected over time during the first 8 days of the study.

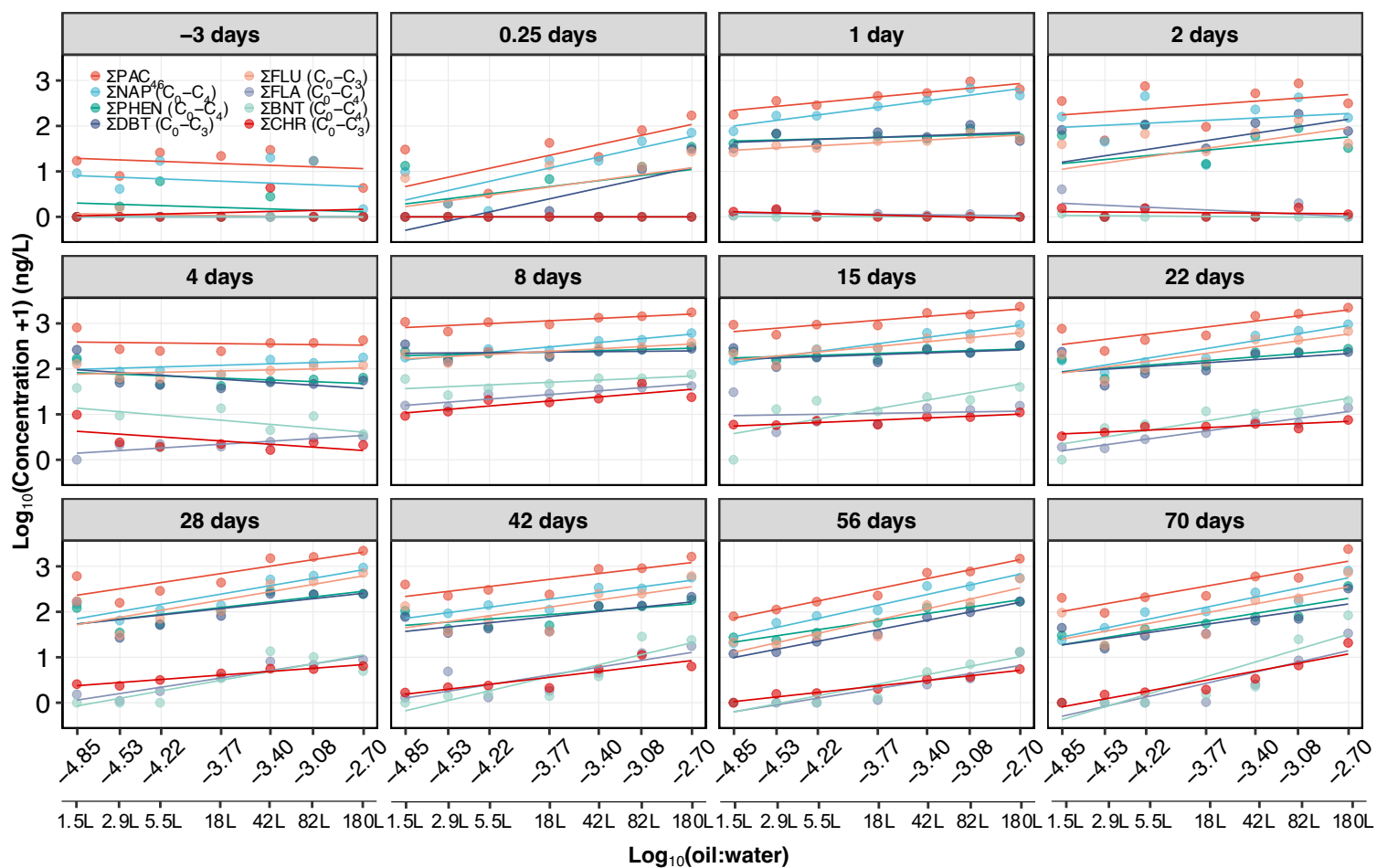


Figure S17. Individual linear regressions comparing concentration in the water column and spill volume at each timepoint for each PAC group. Both axes were logarithmically transformed. To account for cases where concentrations were 0, a constant of 1 was added to each measurement prior to transformation. The controls were excluded from the regressions.

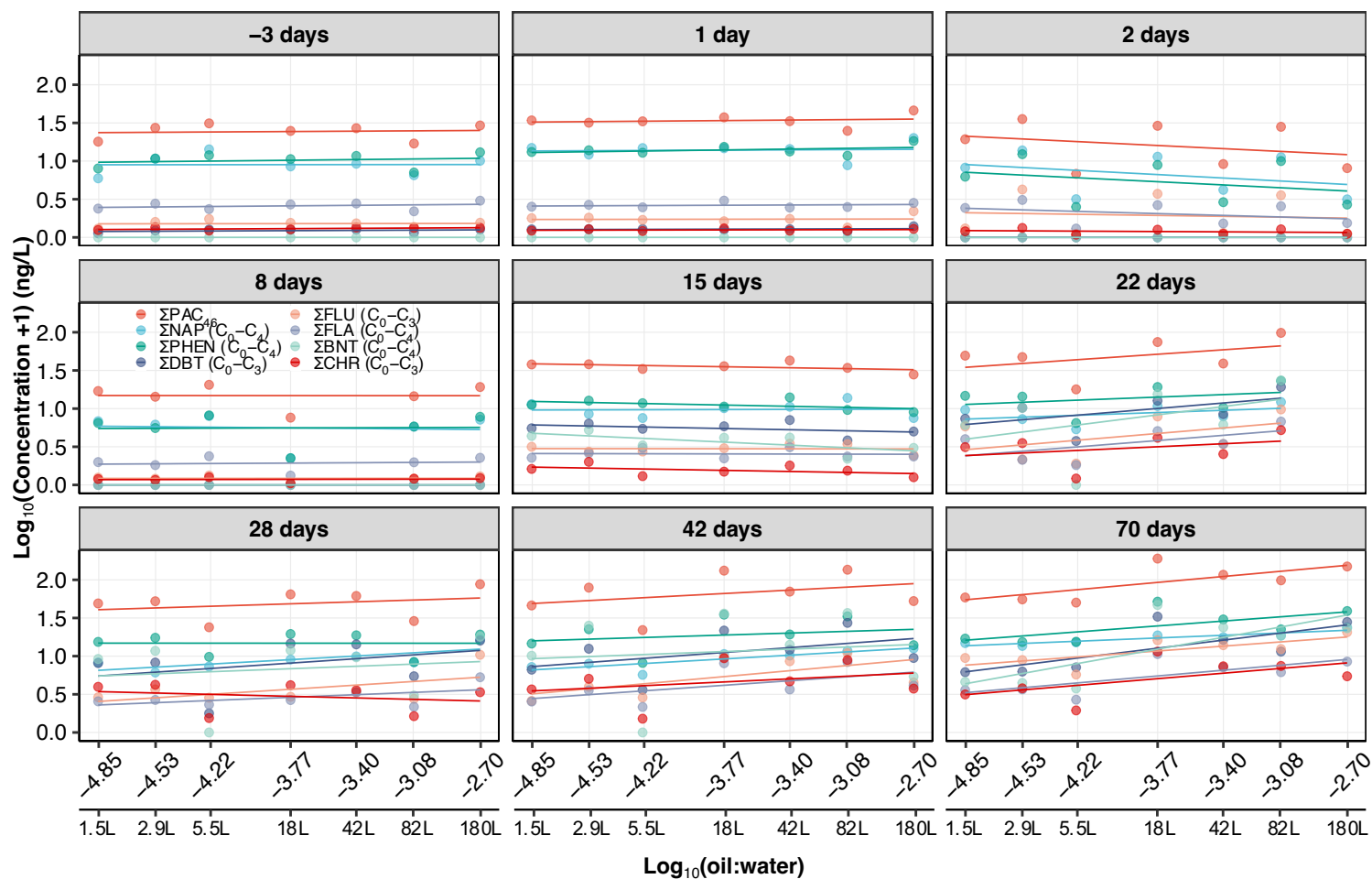


Figure S17. Individual linear regressions comparing concentration in the sediments and spill volume at each timepoint for each PAC group. Both axes were logarithmically transformed. To account for cases where concentrations were 0, a constant of 1 was added to each measurement prior to transformation. The controls were excluded from the regressions.

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