

Supporting information for:

How good are the predictions of mobility of aged  
polychlorinated biphenyls (PCBs) in soil? Insights from a  
soil column experiment

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## **S1. Soil sampling and treatment**

Soil used in this experiment derives from a rhizoremediation greenhouse experiment in pots treated with *Festuca arundinacea*, performed by ERSAF (Ente Regionale per i Servizi all'Agricoltura e alle Foreste) Lombardia (Brescia, Italy). After 18 months from the beginning of the experiment, soil sub-samples (250 g) were obtained from a pot using the incremental sampling methodology of the one-dimensional Japanese slab cake (JSC) (ITRC, 2012) to reduce the data variability and increases sample representativeness. Briefly, for the JSC the soil contained in the gardening pot was passed through a certified stainless-steel sieve with 2 mm mesh and was stratified on clean flat surface. A flat-bottom scoop was used to sample at least 30 fractions (randomly taken at the stratified flat surface) of soil to reach the final weight and placed in a stainless-steel bowl where it was mixed thoroughly. Then, samples were placed in glass vessel and frozen at -20 °C until column preparation or soil analysis.

## **S2. Dissolved organic carbon (DOC) determination**

Dissolved organic carbon of the 2 leaching solutions (tap water and humic acid) and of 4 samples of leachate not analysed for chemicals were determined in an external laboratory with a TOC (Total organic carbon) analyser (Skalar's Formacs<sup>HT</sup> TOC / TN Analyzer, Skalar Analytical B.V., Breda, Netherlands). These samples, together with samples prepared with potassium hydrogen biphthalate (KHP) (n=6) were used to obtain an equation to correlate DOC data and UV absorbance at 254 nm (UV-VIS Evolution 220, Thermo Scientific, Waltham, MA, U.S.A.). Since ions in solution (e.g. iron and nitrate) are known to determine UV interferences (Weishaar et al., 2003) conductivity was included in the regression to account for these effects.

The final equation used for DOC determination was:

$$[\text{DOC}] = 67.7 * \text{abs} - 0.05 * \text{CD} + 0.32 \quad (R^2 = 0.99) \quad (1)$$

Where [DOC] is the DOC concentration (mg/L), abs is the absorbance in  $\text{cm}^{-1}$ , CD is the conductivity in  $\mu\text{S cm}^{-1}$  of the sample corrected for the conductivity of the laboratory reagent water (e.g. tap water or ultrapure water).

### S3. Physico-chemical properties of the target contaminants

**Table S3.1.** Physico-chemical-properties of target contaminants. MW: molecular weight. WS: water solubility.

Vp: vapour pressure. HL: Half-life.

Congener	MW (g/mol) <sup>1</sup>	WS (mg/L) <sup>1</sup>	Vp (Pa) <sup>1</sup>	Log Kow <sup>2</sup>	HL air (d) <sup>1</sup>	HL water (d) <sup>1</sup>	HL soil (d) <sup>1</sup>
PCB 28	257.5	0.16	0.132	5.67	22.91	708.33	2291.67
PCB 52	292	0.03	0.0049	5.84	70.83	2291.67	2291.67
PCB 101	326.4	0.01	0.00109	6.38	70.83	2291.67	2291.67
PCB 138	360.9	0.001 <sup>3</sup>	0.000119 <sup>3</sup>	6.83	229.17	2291.67	2291.67
PCB 153	360.9	0.001	0.000119	6.92	229.17	2291.67	2291.67
PCB 180	395.3	0.002 <sup>4</sup>	0.0000273 <sup>4</sup>	7.36	229.17	2291.67	2291.67
PCB 209	498.7	0.000001	5.02E-08	8.18	2291.67	2291.67	2291.67

<sup>1</sup>Mackay et al. (1992). <sup>2</sup>Hawker and Connell (1988). <sup>3</sup>Assumed equal to PCB 153. <sup>4</sup>Assumed equal to PCB 171.

### S4. Limit of quantitation (LOQ)

Limit of quantitation (LOQ) of target PCBs are reported in table S4.1.

**Table S4.1.** LOQ values for soil, leachates and particles.

Congener	LOQ			
	Soil ( $\mu\text{g/kg}$ )	Leachates (PW) (ng/L)	Leachates (FF) (ng/L)	Particles (ng/L)
PCB 28/31	1.55	0.97	0.15	1.93
PCB 52	1.76	1.10	0.18	2.19
PCB 101	1.93	1.21	0.19	2.41
PCB 138	1.86	1.16	0.19	2.33
PCB 153	1.81	1.13	0.18	2.26
PCB 180	2.01	1.26	0.20	2.52
PCB 209	2.12	1.33	0.21	2.65

## S5. Leachate characteristics

**Table S5.1.** Temperature in leachates (average) (fraction not analysed for chemicals).

CT	t° C			
	TW	HA	LT	FC
2d	23	24	-	-
5d	26	26	-	-
7d	25	25	15	19
48d	26	26	-	-

Note: TW: tap water. HA: humic acid. LT: low temperature. FC: Field capacity. CT: contact time.

**Table S5.2.** pH in leachates (average) (fraction not analysed for chemicals).

CT	pH			
	TW	HA	LT	FC
2d	7	7	-	-
5d	7.5	7.5	-	-
7d	7.5	7.5	6.5	7.5
48d	7.5	7.5	-	-

Note: TW: tap water. HA: humic acid. LT: low temperature. FC: Field capacity. CT: contact time.

**Table S5.3.** Conductivity data (average and standard deviation).

CT	Conductivity $\mu\text{S cm}^{-1}$											
	PW1			PW2			FF					
	TW	HA	LT	TW	HA	LT	TW	HA	LT	FC		
2d	1307 ± 35	1399 ± 85	-	868 ± 18	929 ± 56	-	-	-	-	-	-	
5d	559 ± 13	561 ± 34	-	435 ± 6	442 ± 31	-	305 ± 20	306 ± 21	-	-	-	
7d	410 ± 15	412 ± 20	263 ± 21	374 ± 18	354 ± 26	321 ± 8	-	-	217 ± 10	469 ± 60	-	
48d	692 ± 22	750 ± 48	-	570 ± 25	645 ± 13	-	734 ± 102	773 ± 76	-	-	-	

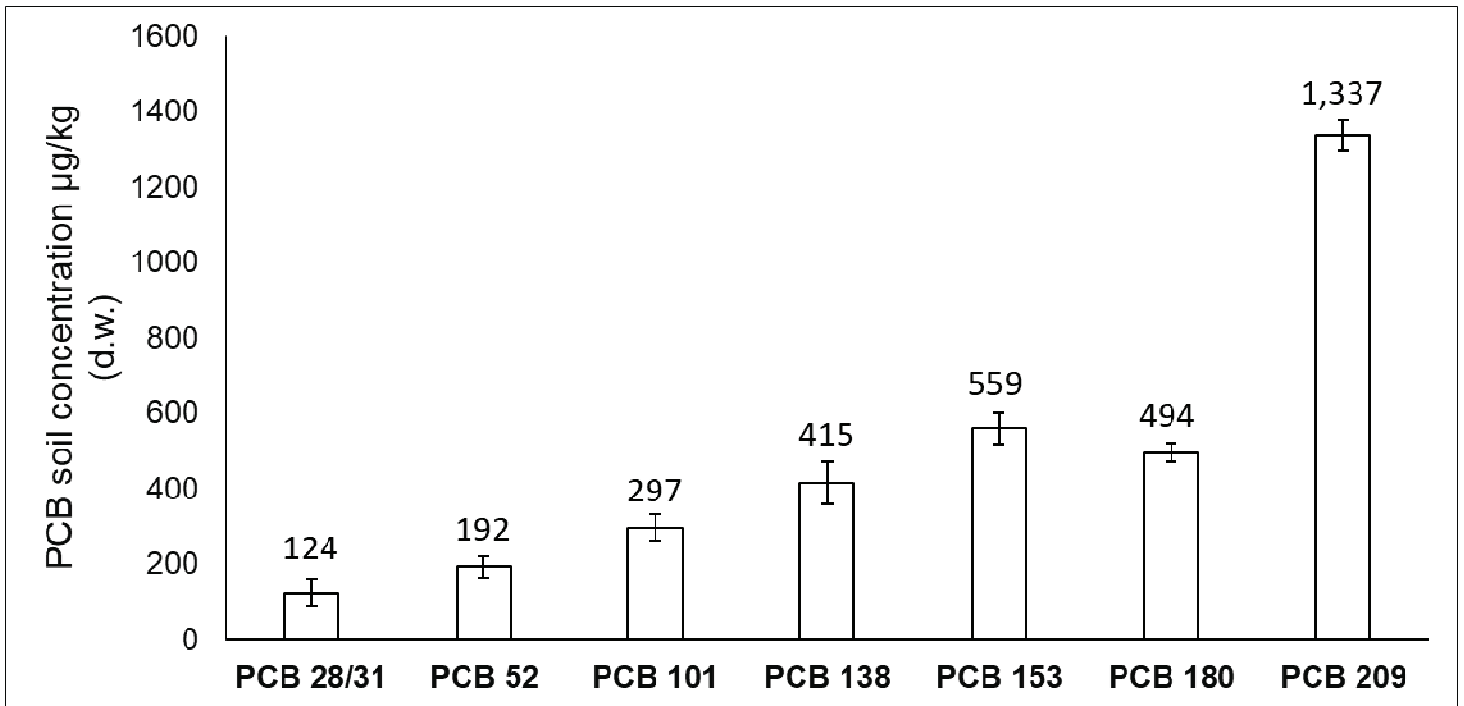
Note: PW1: pore water (fraction 1). PW2: pore water (fraction 2). FF: fast flow. TW: tap water. HA: humic acid. LT: low temperature. FC: Field capacity. CT: contact time.

**Table S5.4.** DOC data (average and standard deviation).

CT	DOC concentration (mg/L)											
	PW1			PW2			FF					
	TW	HA	LT	TW	HA	LT	TW	HA	LT	FC		
2d	31.30 ± 2.22	22.44 ± 3.33	-	28.84 ± 1.52	22.70 ± 2.19	-	-	-	-	-	-	
5d	35.04 ± 1.49	38.26 ± 0.43	-	36.31 ± 1.48	36.15 ± 1.05	-	33.76 ± 6.74	38.07 ± 7.50	-	-	-	
7d	28.48 ± 2.38	38.28 ± 1.97	12.05 ± 3.86	25.73 ± 1.52	34.31 ± 1.56	7.12 ± 4.16	-	-	9.62 ± 4.38	15.33 ± 7.28	-	
48d	52.37 ± 3.15	58.30 ± 5.12	-	47.71 ± 2.70	53.12 ± 2.43	-	33.73 ± 7.53	42.51 ± 3.89	-	-	-	

Note: PW1: pore water (fraction 1). PW2: pore water (fraction 2). FF: fast flow. TW: tap water. HA: humic acid. LT: low temperature. FC: Field capacity. CT: contact time.

**S6. PCB concentrations in soil**



**Figure S6.1.** PCB concentrations in soil (average and standard deviation).

## S7. PCB bulk concentrations in leachate

**Table S7.1.** PCB bulk concentrations in leachates (average and standard deviation).

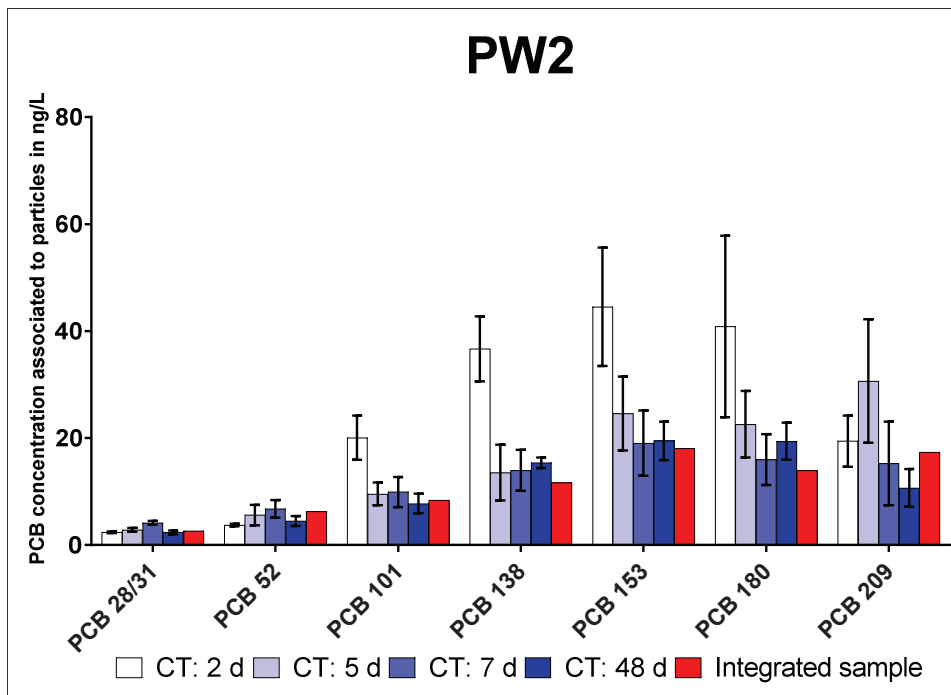
Congener	CT	PCB bulk water concentration (ng/L)									
		PW1			PW2			FF			
		TW	HA	LT	TW	HA	LT	TW	HA	LT	FC
28/31	2d	23.40 ± 0.56	25.42 ± 1.52	-	20.29 ± 2.89	21.15 ± 3.80	-	-	-	-	-
	5d	22.57 ± 2.14	24.29 ± 0.94	-	16.22 ± 2.71	16.55 ± 1.31	-	5.40 ± 0.31	10.55 ± 2.00	-	-
	7d	19.57 ± 0.84	22.71 ± 3.30	3.72 ± 0.80	10.18 ± 0.47	11.14 ± 2.42	3.96 ± 0.33	-	-	5.50 ± 1.49	8.99 ± 0.65
	48d	17.27 ± 0.95	18.53 ± 1.15	-	7.84 ± 1.06	8.66 ± 1.86	-	4.25 ± 0.38	9.47 ± 2.71	-	-
52	2d	34.63 ± 1.73	41.32 ± 4.60	-	34.54 ± 4.55	37.78 ± 6.04	-	-	-	-	-
	5d	41.13 ± 5.94	51.27 ± 5.00	-	33.36 ± 1.84	35.69 ± 1.92	-	10.27 ± 0.43	22.16 ± 2.46	-	-
	7d	33.88 ± 2.94	45.96 ± 9.06	7.24 ± 1.49	18.95 ± 1.46	23.89 ± 7.98	6.81 ± 1.75	-	-	11.53 ± 3.26	13.00 ± 1.80
	48d	35.80 ± 3.31	35.14 ± 1.39	-	14.52 ± 2.65	17.59 ± 5.09	-	7.35 ± 1.42	19.07 ± 7.02	-	-
101	2d	55.48 ± 2.85	54.66 ± 5.49	-	41.81 ± 9.79	46.26 ± 7.11	-	-	-	-	-
	5d	63.11 ± 5.35	73.66 ± 8.73	-	47.89 ± 3.04	57.71 ± 2.49	-	12.02 ± 0.59	35.95 ± 2.19	-	-
	7d	68.45 ± 9.56	91.61 ± 4.82	10.87 ± 0.58	34.54 ± 5.67	47.37 ± 15.54	7.28 ± 1.87	-	-	8.60 ± 0.71	17.69 ± 1.65
	48d	69.24 ± 4.88	69.55 ± 6.76	-	22.85 ± 5.95	31.78 ± 10.11	-	8.50 ± 0.59	36.57 ± 8.67	-	-
138	2d	66.02 ± 2.22	66.61 ± 4.78	-	41.56 ± 6.26	44.91 ± 3.46	-	-	-	-	-
	5d	78.46 ± 3.51	85.24 ± 8.70	-	55.23 ± 0.32	58.62 ± 2.50	-	13.06 ± 0.42	39.79 ± 6.86	-	-
	7d	92.78 ± 10.48	129.74 ± 5.88	12.27 ± 2.76	45.35 ± 10.13	68.89 ± 15.04	7.48 ± 0.98	-	-	5.20 ± 0.90	22.48 ± 1.83
	48d	100.75 ± 5.58	101.57 ± 20.05	-	32.25 ± 11.26	55.21 ± 15.63	-	12.56 ± 0.77	50.50 ± 7.86	-	-
153	2d	86.39 ± 4.33	85.36 ± 1.89	-	67.65 ± 13.07	62.82 ± 6.88	-	-	-	-	-
	5d	98.05 ± 3.89	106.18 ± 10.42	-	71.11 ± 1.13	78.30 ± 2.65	-	15.86 ± 0.36	47.92 ± 6.28	-	-
	7d	111.30 ± 12.69	154.29 ± 8.27	15.37 ± 1.62	57.31 ± 12.35	86.25 ± 21.46	9.11 ± 1.67	-	-	7.15 ± 1.15	27.69 ± 1.59
	48d	118.60 ± 2.68	124.18 ± 21.87	-	39.13 ± 11.53	63.66 ± 16.88	-	14.66 ± 0.63	58.71 ± 9.29	-	-
180	2d	47.75 ± 5.39	52.33 ± 1.17	-	55.90 ± 13.58	48.37 ± 10.44	-	-	-	-	-
	5d	67.82 ± 2.24	75.20 ± 9.74	-	48.89 ± 7.09	43.16 ± 3.50	-	10.54 ± 0.77	29.84 ± 5.55	-	-
	7d	68.43 ± 7.59	106.85 ± 8.10	11.67 ± 2.99	36.68 ± 8.97	55.34 ± 12.28	4.19 ± 1.81	-	-	3.14 ± 0.69	17.00 ± 2.19
	48d	79.30 ± 5.61	83.97 ± 21.96	-	25.08 ± 7.31	51.35 ± 6.63	-	12.02 ± 0.64	39.62 ± 4.37	-	-
209	2d	22.04 ± 1.35	30.89 ± 5.73	-	30.98 ± 9.47	25.84 ± 3.72	-	-	-	-	-
	5d	46.55 ± 2.96	47.50 ± 8.96	-	38.40 ± 14.52	33.54 ± 6.56	-	5.97 ± 0.31	9.31 ± 3.09	-	-
	7d	39.29 ± 1.66	58.04 ± 6.94	6.72 ± 1.31	25.06 ± 8.64	38.27 ± 7.80	3.27 ± 1.23	-	-	1.46 ± 0.56	8.94 ± 1.54
	48d	44.73 ± 5.14	46.19 ± 18.02	-	13.38 ± 4.40	31.14 ± 7.88	-	7.86 ± 0.59	10.98 ± 2.39	-	-

Note: PW1: pore water (fraction 1). PW2: pore water (fraction 2). FF: fast flow. TW: tap water. HA: humic acid. LT: low temperature. FC: Field capacity. CT: contact time.



## S8. PCB concentrations in particles

To reduce the number of the analysis, the fraction of PCBs associated to particles in the experiment “TW vs. HA” was monitored just in the second pore water fraction (PW2) while all the fractions collected in “saturated vs. unsaturated” and in “HT vs. LT” experiments were analysed also for particle concentrations. Specifically, the samples were split in two fractions, one fraction was used for bulk concentration extraction and the other one was filtered and used for the particle measurement (Table S8.1). After the first sampling (contact time, CT: 2 days) where the particle concentrations were higher compared to the others CT, the transport associated to particles was stable with time (Figure S8.1). An additional sampling was therefore performed after a CT of 44 days at the same temperature conditions (25 °C) to attempt an estimation of the fraction associated to particles also for the first pore water fraction (PW1) and the fast flow (FF), wherever these data were missing. Since, particle concentrations were close to MDL for some congeners (PCB 28/31, 52), an integrated sample was performed, mixing leachate from 3 replicate columns to increase the signal. Data from this additional sampling were quite in agreement with the other samplings (Figure S8.1), therefore data from this experiment (Table S8.2) were used to estimate PCB distributions (S9).



**Figure S8.1.** Temporal trend (contact time (CT): 2, 5, 7, 48 days) of PCB fractions associated to particles (average and standard deviation) compared to the integrated samples performed after a CT of 44 days in PW2 (pore water, fraction 2).

**Table S8.1.** PCB concentrations in mobile particles (average and standard deviation).

Congener	CT	PCB associated to particle concentration (ng/L)										
		PW1			PW2			FF				
		TW	HA	LT	TW	HA	LT	TW	HA	LT	FC	
28/31	2d	-	-	-	2.39 ± 0.14	4.28 ± 1.40	-	-	-	-	-	-
	5d	-	-	-	2.86 ± 0.35	2.72 ± 0.46	-	-	-	-	-	
	7d	-	-	2.57 ± 0.10	4.18 ± 0.35	4.59 ± 1.07	2.68 ± 0.40	-	-	2.62 ± 0.38	<1.93	
	48d	-	-	-	2.39 ± 0.37	3.10 ± 1.10	-	-	-	-	-	
52	2d	-	-	-	3.75 ± 0.22	11.20 ± 4.57	-	-	-	-	-	
	5d	-	-	-	5.63 ± 1.91	6.12 ± 2.58	-	-	-	-	-	
	7d	-	-	4.75 ± 0.14	6.77 ± 1.67	11.24 ± 0.56	3.94 ± 0.64	-	-	4.17 ± 0.92	<2.19	
	48d	-	-	-	4.49 ± 0.92	6.44 ± 2.23	-	-	-	-	-	
101	2d	-	-	-	20.06 ± 4.10	20.82 ± 7.93	-	-	-	-	-	
	5d	-	-	-	9.57 ± 2.14	9.24 ± 4.16	-	-	-	-	-	
	7d	-	-	6.25 ± 0.98	9.93 ± 1.83	19.95 ± 3.32	4.31 ± 0.67	-	-	4.96 ± 1.17	2.61 ± 0.31	
	48d	-	-	-	7.75 ± 2.81	11.20 ± 4.64	-	-	-	-	-	
138	2d	-	-	-	32.76 ± 2.16	32.41 ± 5.58	-	-	-	-	-	
	5d	-	-	-	13.57 ± 5.19	15.24 ± 6.89	-	-	-	-	-	
	7d	-	-	8.74 ± 1.88	13.98 ± 3.84	34.84 ± 4.96	4.18 ± 1.11	-	-	1.86 ± 0.17	4.53 ± 2.03	
	48d	-	-	-	15.38 ± 1.00	22.02 ± 7.16	-	-	-	-	-	
153	2d	-	-	-	40.50 ± 4.06	44.38 ± 11.27	-	-	-	-	-	
	5d	-	-	-	24.61 ± 6.91	20.69 ± 6.32	-	-	-	-	-	
	7d	-	-	11.23 ± 3.69	19.07 ± 6.10	44.26 ± 9.12	4.72 ± 0.76	-	-	3.08 ± 0.64	5.40 ± 1.31	
	48d	-	-	-	19.49 ± 3.59	28.17 ± 13.25	-	-	-	-	-	
180	2d	-	-	-	31.87 ± 1.58	38.46 ± 3.79	-	-	-	-	-	
	5d	-	-	-	22.60 ± 6.21	19.83 ± 6.21	-	-	-	-	-	
	7d	-	-	9.31 ± 3.81	15.99 ± 4.74	40.86 ± 10.36	2.97 ± 1.60	-	-	2.51 ± 0.55	7.14 ± 3.93	
	48d	-	-	-	19.41 ± 3.45	27.52 ± 9.04	-	-	-	-	-	
209	2d	-	-	-	19.43 ± 4.80	20.67 ± 2.98	-	-	-	-	-	
	5d	-	-	-	30.67 ± 11.53	23.76 ± 2.01	-	-	-	-	-	
	7d	-	-	5.38 ± 1.05	15.28 ± 7.79	30.61 ± 6.24	2.22 ± 0.31	-	-	1.01 ± 0.59	6.54 ± 0.36	
	48d	-	-	-	10.71 ± 3.52	24.91 ± 6.31	-	-	-	-	-	

Note: PW1: pore water (fraction 1). PW2: pore water (fraction 2). FF: fast flow. TW: tap water. HA: humic acid. LT: low temperature. FC: Field capacity. CT: contact time.

**Table S8.2.** PCB concentrations in mobile particles measured after a CT of 44 days (integrated samples from different columns).

<b>Congener</b>	<b>Concentration of PCBs associated to particles (ng/L)</b>		
	<b>PW 1</b>	<b>PW 2</b>	<b>FF</b>
PCB 28/31	3.22	2.64	1.70
PCB 52	10.32	6.31	4.16
PCB 101	18.17	8.39	4.24
PCB 138	31.28	11.70	5.35
PCB 153	47.81	18.07	8.03
PCB 180	39.20	13.96	4.21
PCB 209	51.87	17.35	6.53

Note: PW1: pore water (fraction 1). PW2: pore water (fraction 2). FF: fast flow.

S9. PCB distribution among freely dissolved, DOC and particles

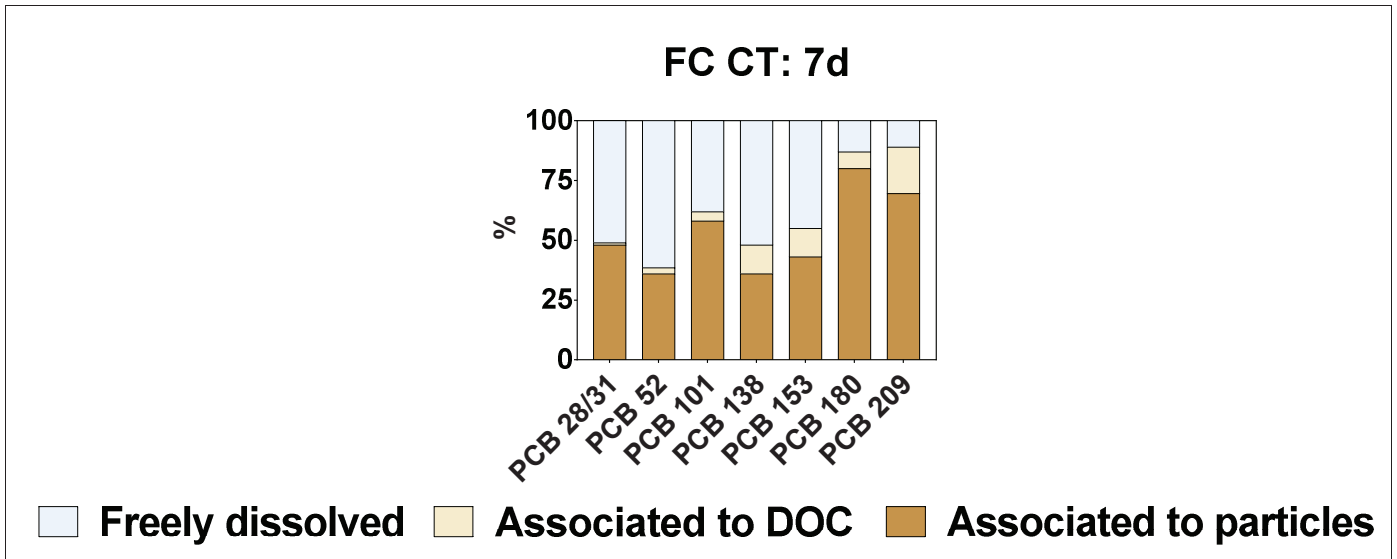


Figure S9.1. PCB distribution among freely dissolved, DOC and mobile particles in TW (tap water) samples after a contact time (CT) of 7 days in field capacity (FC). FF: fast flow.

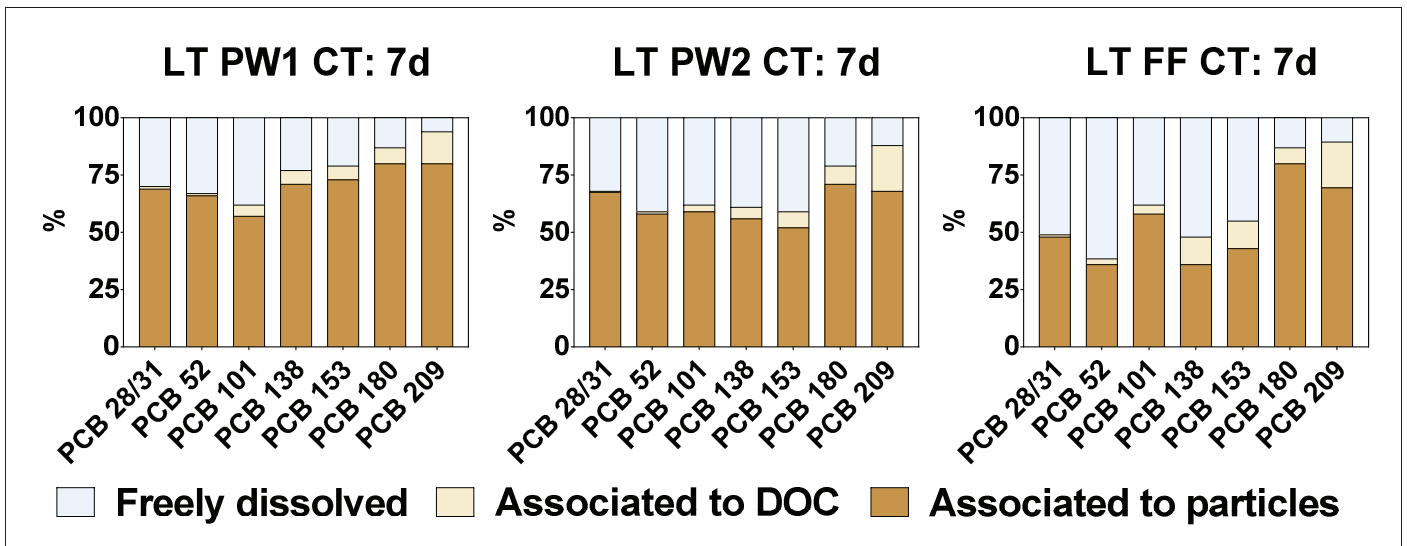


Figure S9.2. PCB distribution among freely dissolved, DOC and mobile particles in TW (tap water) samples at 15° C after a contact time (CT) of 7 days. PW1: pore water (fraction 1). PW2: pore water (fraction 2). FF: fast flow. LT: low temperature.

## S10. Cumulative leaching fluxes vs. Log K<sub>ow</sub>

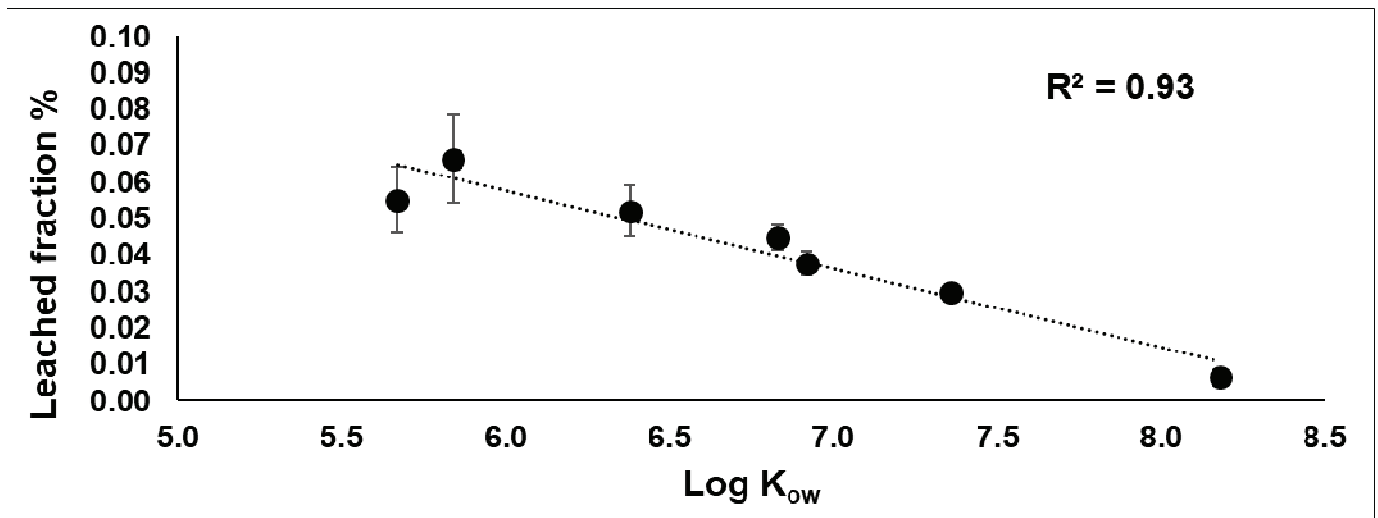
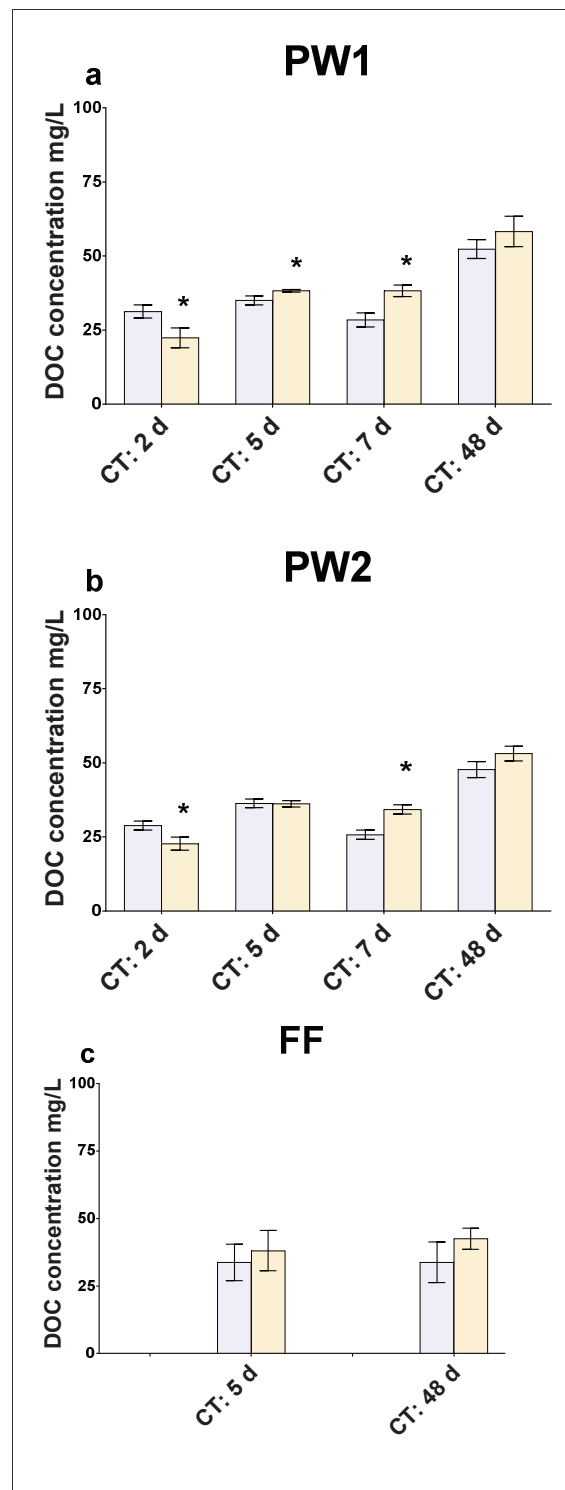
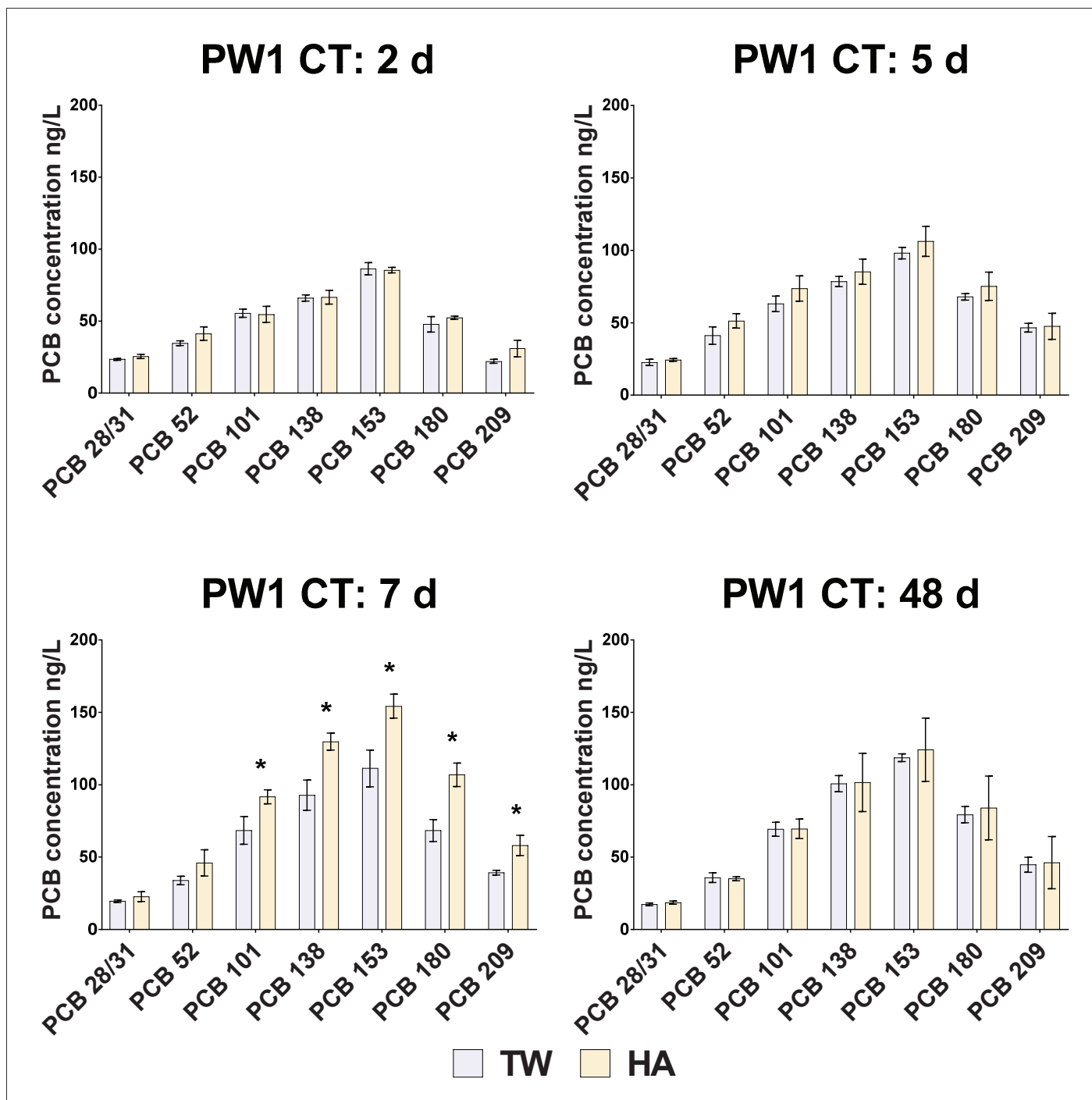


Figure S10.1. Cumulative percentage of PCBs leached from initial soil content toward Log K<sub>ow</sub> values (average and standard deviation).

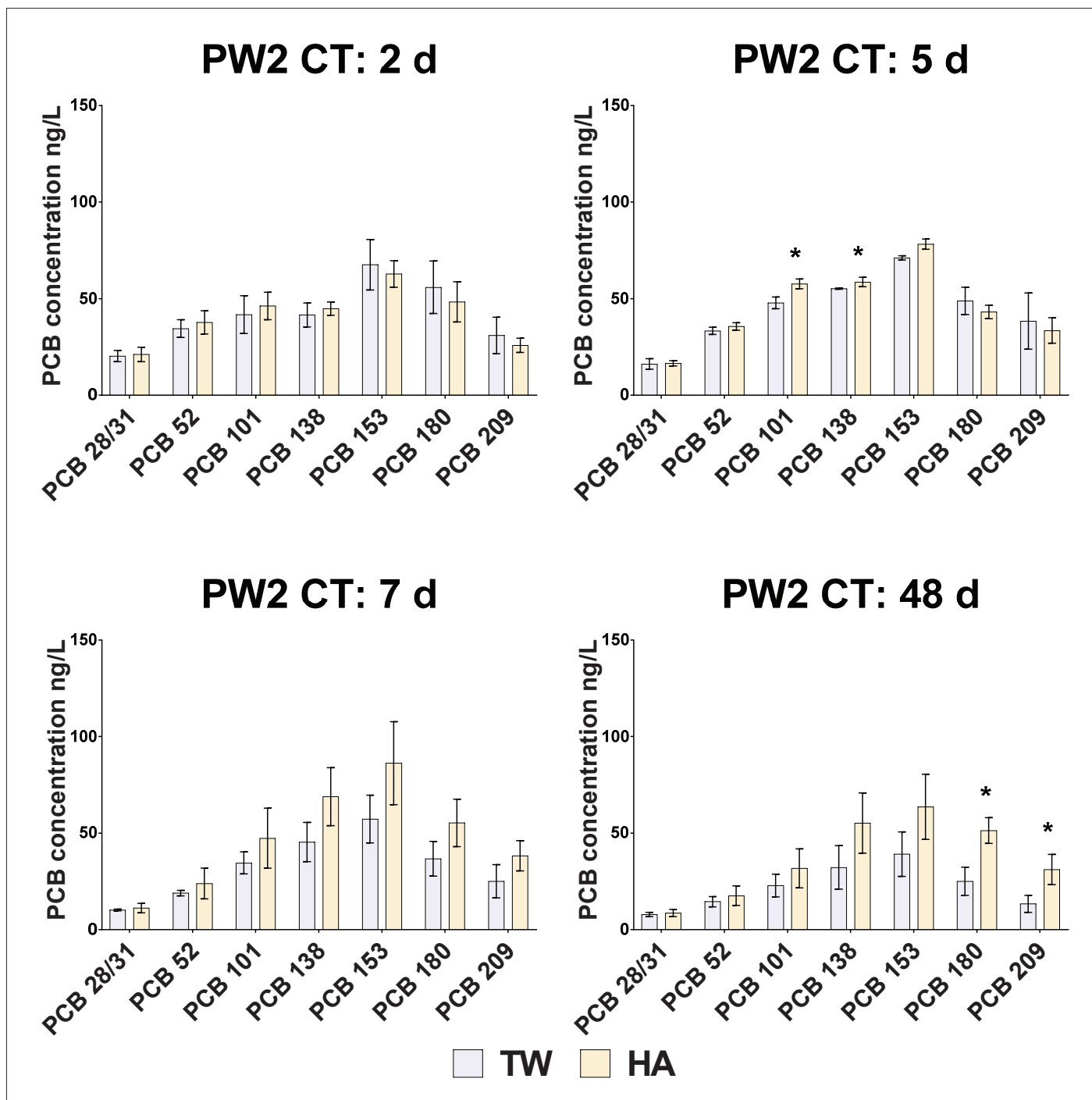
## S11. Comparison between TW and HA samples



**Figure S11.1.** Comparison between DOC concentrations in TW (tap water) and HA (humic acid) columns at different contact time (CT) (average and standard deviation). Data labelled with \* were significantly different (Student's t test,  $\alpha=0.05$ ). PW1: pore water (fraction 1). PW2: pore water (fraction 2). FF: fast flow.

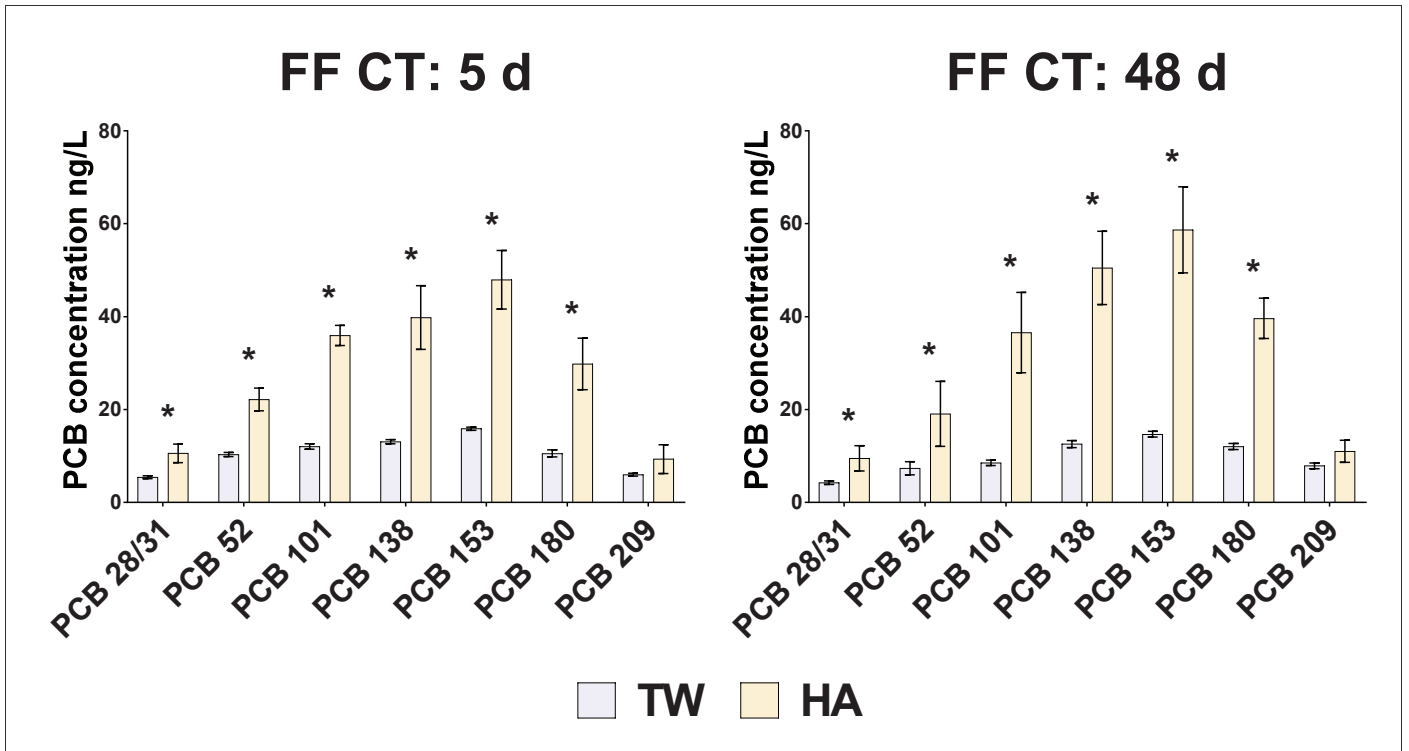


**Figure S11.2.** Comparison between PCB bulk concentrations in TW (tap water) and HA (humic acid) columns in PW1 (pore water, fraction 1) samples at different contact time (CT) (average and standard deviation). Data labelled with \* were significantly different (Student's t test,  $\alpha=0.05$ ).



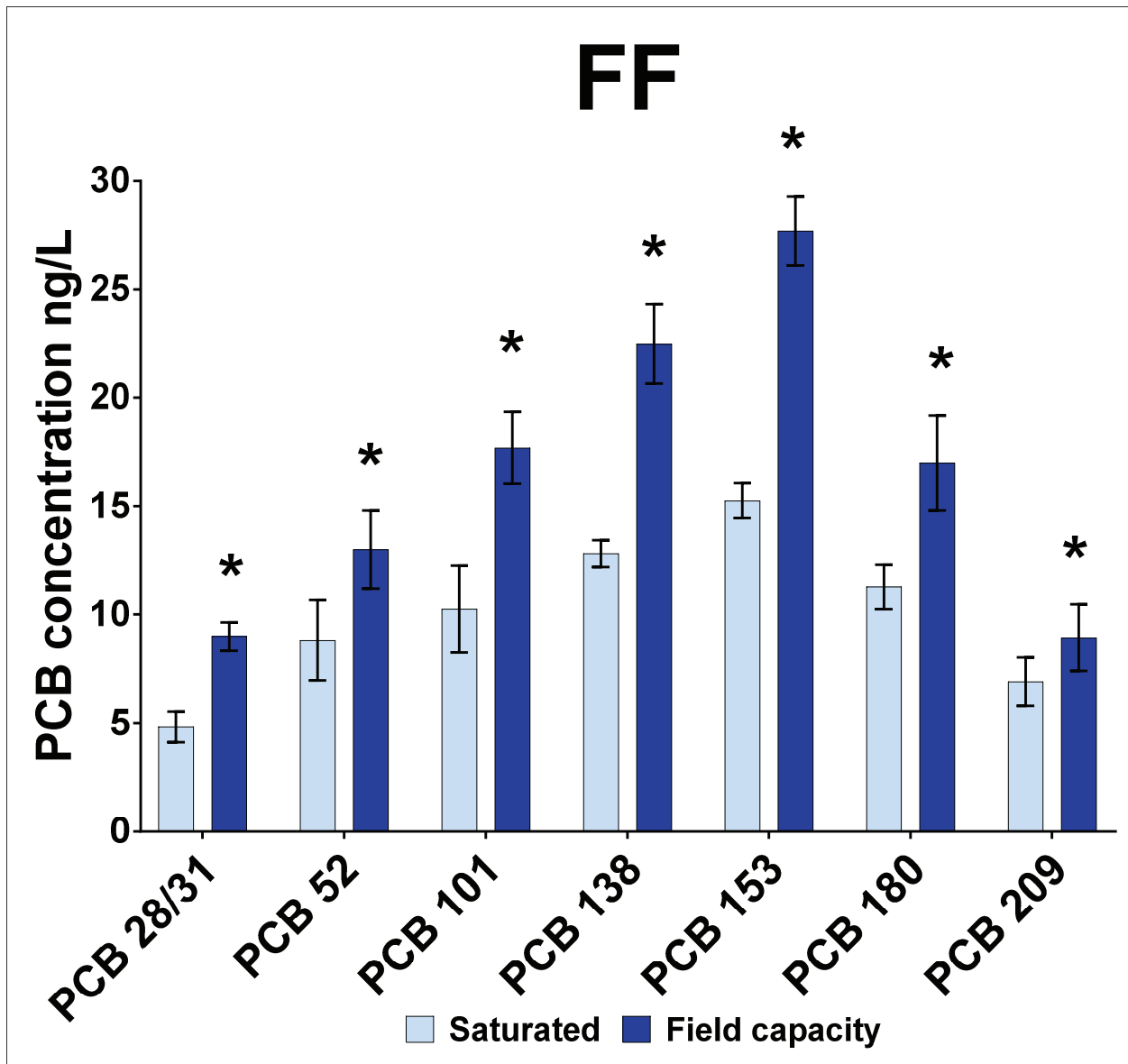
**Figure S11.3.** Comparison between PCB bulk concentrations in TW and HA columns in PW2 (pore water, fraction 2) samples at different contact time (CT) (average and standard deviation). Data labelled with \* were significantly different (Student's t test,  $\alpha=0.05$ ).





**Figure S11.4.** Comparison between PCB concentrations in TW and HA columns in FF (fast flow) samples at different contact time (CT) (average and standard deviation). Data labelled with \* were significantly different (Student's t test,  $\alpha=0.05$ ).

S12. Comparison between samples collected in saturated and field capacity conditions



**Figure S12.1.** Comparison between PCB concentrations in saturated towards field capacity conditions in FF (fast flow) samples (average and standard deviation). FF samples in field capacity were collected after a contact time of 7 days. For saturated conditions average values between the two FF sampling after contact time of 5 and 48 days were used. Data labelled with \* were significantly different (Student's t test,  $\alpha=0.05$ ).

S13. Comparison between samples collected at 25 °C and 15 °C temperatures

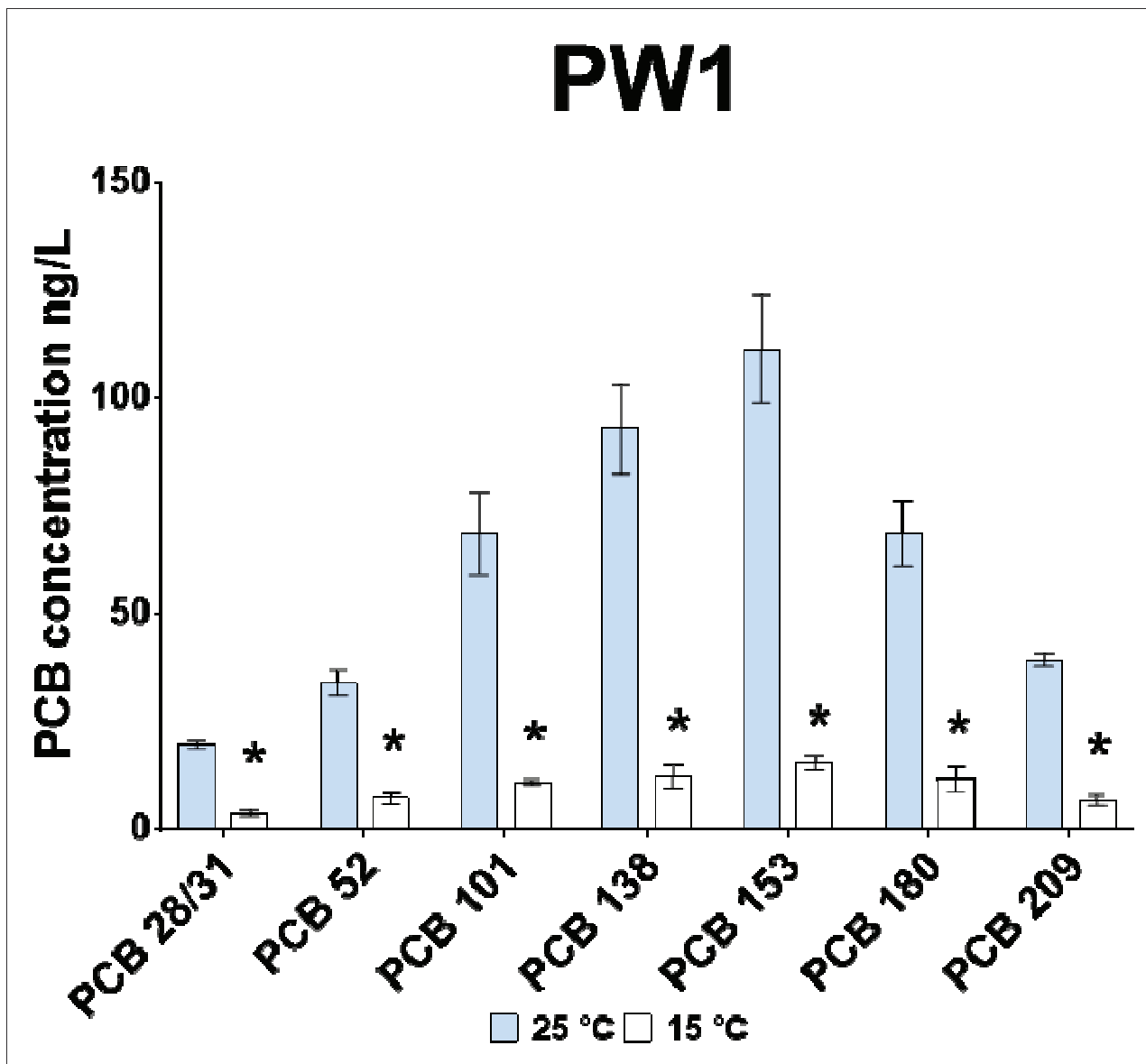
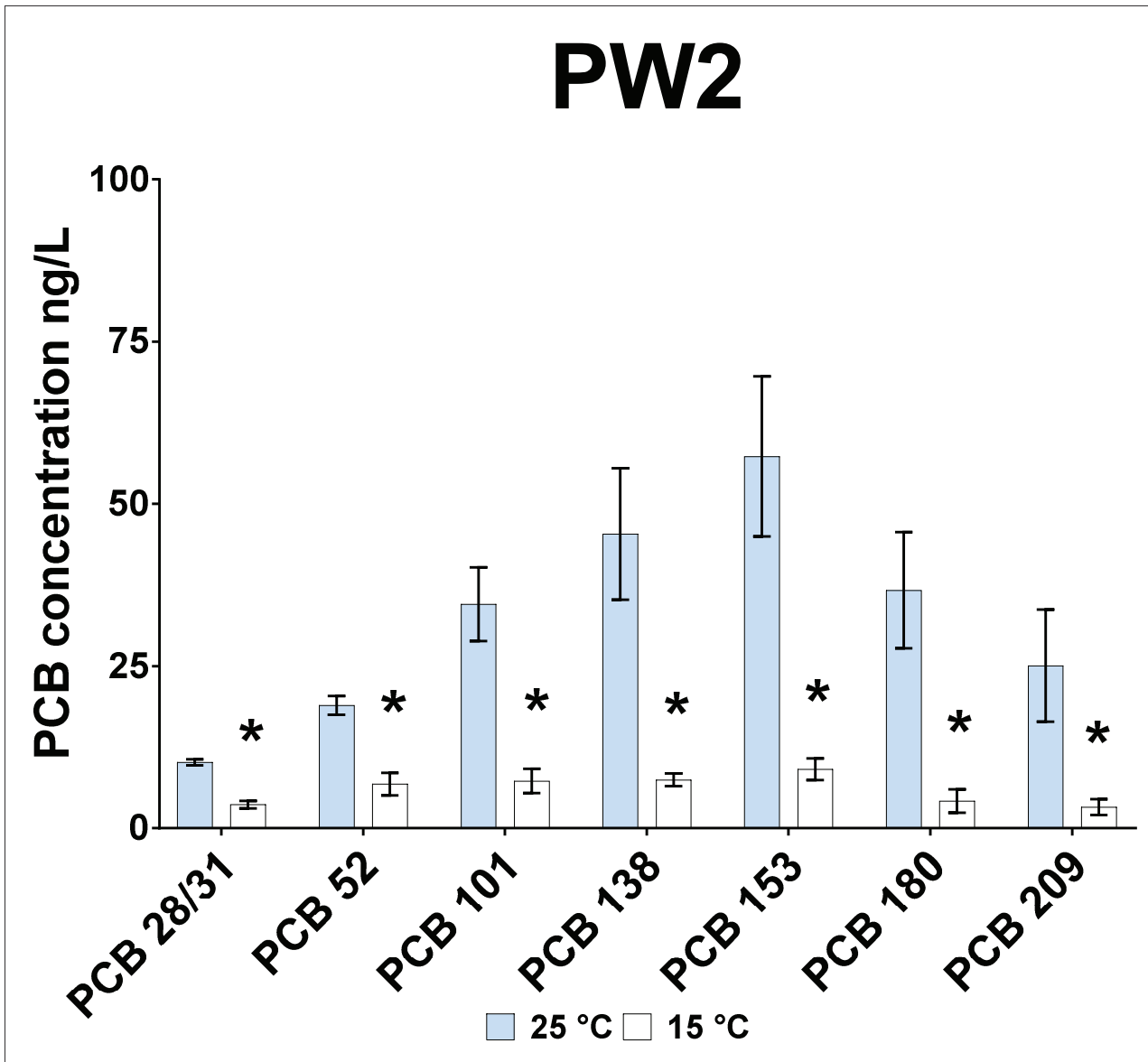
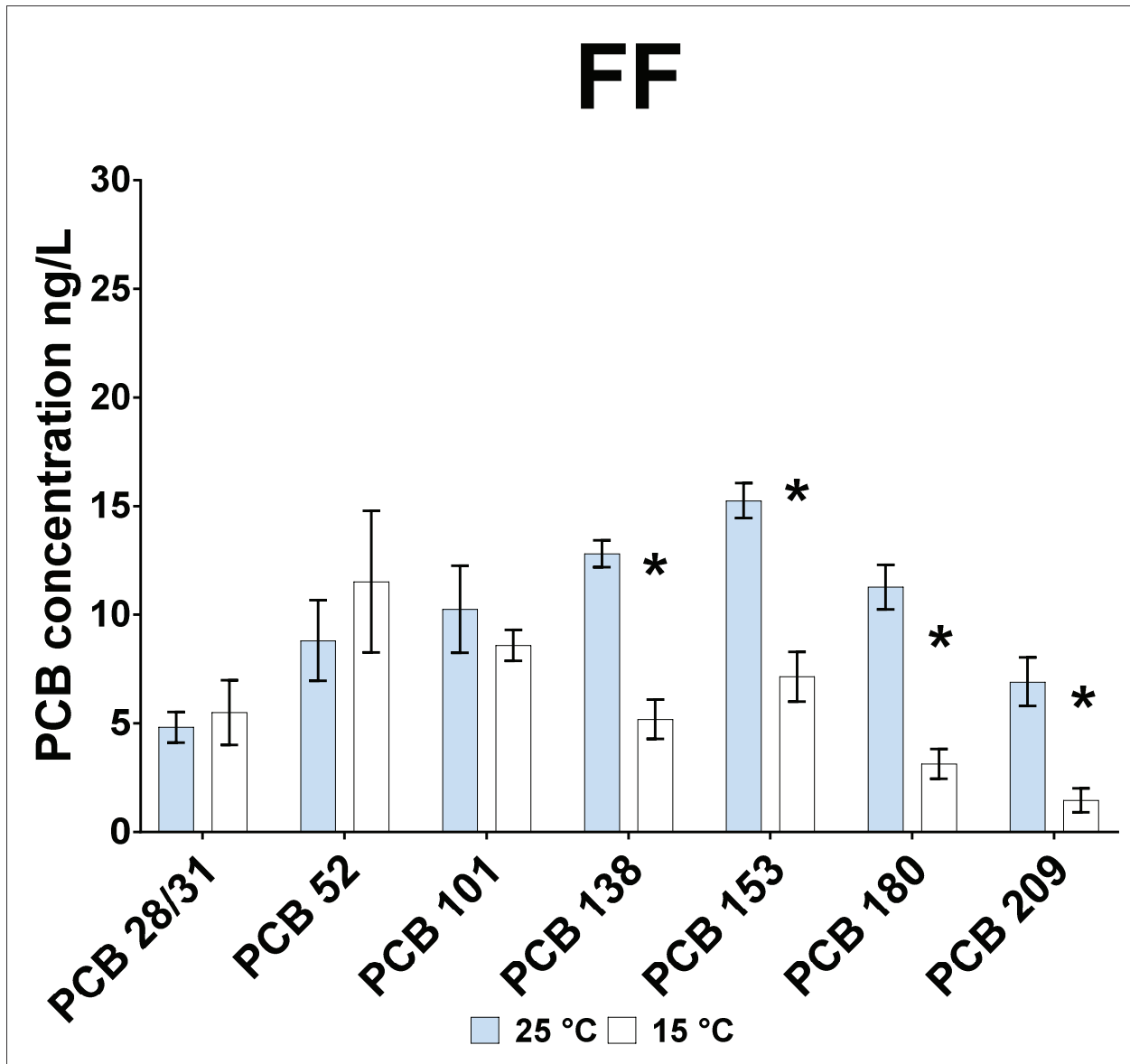


Figure S13.1. Comparison between PCB concentration at 25 °C towards 15 °C at equivalent contact time (7 days) for PW1 (pore water, fraction 1) (average and standard deviation).



**Figure S13.2.** Comparison between PCB concentration at 25 °C towards 15 °C at equivalent contact time (7 days) for PW2 (pore water, fraction 2) (average and standard deviation).



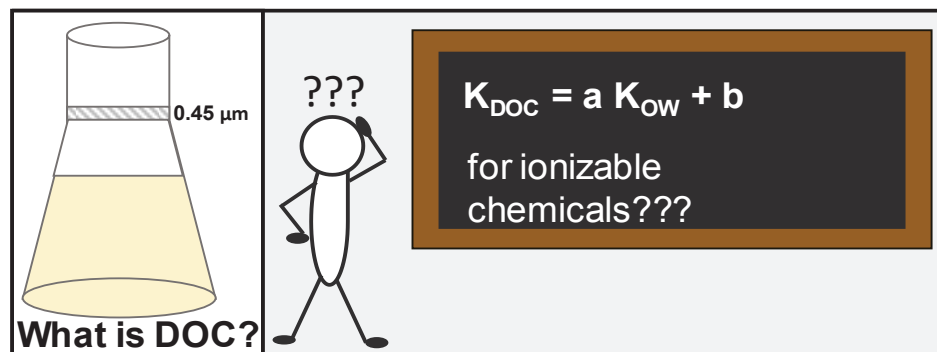
**Figure S13.3.** Comparison between PCB concentrations at 25 °C (left) towards 15 °C (right) for FF (fast flow) samples (average and standard deviation). FF samples at 15 °C were collected after a contact time of 7 days. For FF samples at 25 °C average values between the two FF sampling after contact time of 5 and 48 days were used. Data labelled with \* were significantly different (Student's t test,  $\alpha=0.05$ ).

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**Paper III: A review of the predictive models estimating association of neutral and ionizable organic chemicals with dissolved organic carbon**



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