

Supporting Information

Empirical investigation of the Junge variability-lifetime relationship using long-term monitoring data on PCB concentrations in air

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NOTE: An Excel spreadsheet providing a step-by-step procedure of data handling for an example congener and a full list of MDLs used for each congener at each site is available from the corresponding author.
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Table S1: Summary of Method Detection Limits (MDLs) and data selection. Average homologue MDLs and number of ‘congener years’ available as well as excluded from the analysis. Congeners were excluded due to: (a) missing datapoints (i.e. if there were <15 datapoints (Great Lakes sites) or < 20 (Alert), resulting in <10 months of data coverage for a given year); (b) the majority of data for a given year falling below the MDL.

Site	PCB Homologue (no. of chlorines)	No. of congeners	Average MDL (pg m ⁻³)	No. of congener years used in analysis ^a	Congener years removed due to missing data	Congener years removed due to data <MDL	Overall % removed due to missing data	Overall % removed due to below MDLs
Alert	2	4	0.17	32 (32)	0	0	0	0.2
	3	9	0.08	72 (71)	0	1		
	4	12	0.03	96 (96)	0	0		
	5	12	0.02	96 (96)	0	0		
	6	14	0.02	112 (112)	0	0		
	7	13	0.02	104 (104)	0	0		
	8	6	0.01	48 (48)	0	0		
	9	2	0.01	16 (16)	0	0		
EGH	10	1	0.02	8 (8)	0	0	31	9.5
	2	1	0.13	10	0	0		
	3	13	0.29	101 (130)	27	2		
	4	16	0.28	127 (160)	29	4		
	5	15	0.25	120 (150)	22	8		
	6	11	0.09	70 (110)	31	9		
	7	5	0.14	19 (50)	15	16		
	8	3	0.13	8 (30)	18	4		
BNT	9	0	-	0	0	0	10	9
	10	0	-	0	0	0		
	2	3	1.28	23 (24)	1	0		
	3	7	0.96	49 (56)	1	6		
	4	7	0.48	50 (56)	3	3		
	5	8	0.19	50 (64)	6	8		
	6	4	0.07	31 (32)	1	0		
	7	2	0.05	12 (16)	3	1		
SBD	8	1	0.08	1 (8)	6	1	25	16
	9	0	-	0	0	0		
	10	0	-	0	0	0		
	2	1	0.23	8 (9)	1	0		
	3	11	0.46	87 (99)	12	0		
	4	12	0.38	92 (108)	10	6		
	5	14	0.38	88 (126)	26	12		
	6	8	0.12	43 (72)	20	9		
PPT	7	5	0.14	16 (45)	11	18	16	0.7
	8	2	0.13	4 (18)	5	9		
	9	0	-	0	0	0		
	10	0	-	0	0	0		
	2	3	0.82	27 (27)	0	0		
	3	7	0.81	63 (63)	0	0		
	4	7	0.43	63 (63)	0	0		
	5	10	0.11	77 (99)	12	0		
STP	6	5	0.08	39 (45)	6	0	20	10
	7	4	0.05	26 (36)	9	1		
	8	2	0.07	10 (18)	8	0		
	9	2	0.07	5 (18)	13	0		
	10	0	-	0	0	0		
	2	1	0.13	9 (9)	0	0		
	3	14	0.31	103 (126)	20	3		
	4	16	0.19	124 (144)	17	3		
5	15	0.38	107 (135)	14	14			
6	11	0.08	61 (99)	19	19			
7	8	0.14	28 (72)	20	24			
8	5	0.06	24 (45)	18	3			
9	2	0.02	2 (18)	15	1			
10	1	0.04	0 (9)	9	0			

^a

Numbers in parentheses represent the total number of ‘congener years’ available. For example, at Alert, there were four congeners in the dichlorinated PCB homologue reported over eight years, thus providing a total of 32 congener years.

Numbers

Table S2: Scenario I behaviour. Details of years at all sites that show a positive relationship between log RSD and degree of chlorination together with correlation coefficients (r^2) and statistical significance (p -value). The n - value is the number of homologue groups included for that year. Statistically significant p -values (<0.05) for years showing Scenario I behaviour are highlighted in red.

Site		1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	All years
Alert	Scenario I behaviour?			NO	NO	NO	NO	NO	NO	NO		NO
	n	no data	not used	8	8	8	8	8	8	8	not used	8
	r^2			N/A	N/A	N/A	N/A	N/A	N/A	N/A		N/A
	p value											
EGH	Scenario I behaviour?	NO	NO	NO	YES	NO	YES	NO	NO	NO	NO	NO
	n	5	5	5	4	5	4	4	4	5	4	4
	r^2	N/A	N/A	N/A	0.09	N/A	0.29	N/A	N/A	N/A	N/A	N/A
	p value				0.71		0.35					
BNT	Scenario I behaviour?			NO	YES	YES	NO	NO	YES	YES	YES	NO
	n	no data	no data	6	6	5	6	5	5	6	5	5
	r^2			N/A	0.02	0.39	N/A	N/A	0.77	0.09	0.76	N/A
	p value				0.78	0.26			0.05	0.57	0.05	
SBD	Scenario I behaviour?		NO	NO	NO	NO	NO	NO	NO	NO	NO	NO
	n	no data	6	6	6	6	6	5	6	6	5	5
	r^2		N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
	p value											
PPT	Scenario I behaviour?		YES	YES	YES	YES	NO	NO	YES	YES	YES	YES
	n	no data	8	7	8	6	6	6	6	6	6	7
	r^2		0.50	0.10	0.12	0.54	N/A	N/A	0.44	0.73	0.36	0.34
	p value		0.05	0.50	0.41	0.09			0.15	0.03	0.21	0.13
STP	Scenario I behaviour?		NO	NO	YES	YES	NO	YES	NO	NO	NO	NO
	n	no data	6	6	7	7	6	7	7	7	6	6
	r^2		N/A	N/A	0.84	0.15	N/A	0.08	N/A	N/A	N/A	N/A
	p value				0.01	0.39		0.55				

Table S3: Scenario II behaviour. Details of years at all sites that demonstrate an inverse relationship between log RSD and log CTD together with correlation coefficients (r^2), statistical significance (p -value) and Junge parameters ($-b$ and a). The n - value is the number of homologue groups included for that year. Statistically significant (p -values <0.05) years are highlighted in red.

Site		1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	All years
Alert	Scenario II behaviour?			YES	NO	YES	YES	YES	YES	NO		YES
	n			8	8	8	8	8	8	8		8
	R^2	no data	not used	0.05		0.35	0.15	0.26	0.31		not used	0.01
	p value			0.62	N/A	0.16	0.39	0.24	0.19	N/A		0.81
	b			0.08		0.40	0.26	0.42	0.33			0.03
	a			0.41		0.03	0.08	0.02	0.04			0.80
EGH	Scenario II behaviour?	YES	YES	YES	NO	YES	NO	YES	YES	YES	YES	YES
	n	5	5	5	4	5	4	4	4	5	4	4
	R^2	0.83	0.47	0.46		0.17		0.88	0.99	0.89	0.90	0.01
	p value	0.03	0.20	0.21	N/A	0.49	N/A	0.06	0.003	0.02	0.05	0.83
	b	0.23	0.12	0.14		0.22		0.17	0.26	0.59	0.31	0.02
	a	0.14	0.36	0.34		0.15		0.24	0.11	0.01	0.08	0.81
BNT	Scenario II behaviour?			YES	NO	NO	YES	YES	NO	NO	NO	YES
	n			6	6	5	6	5	5	6	5	5
	R^2	no data	no data	0.31			0.83	0.51				0.26
	p value			0.25	N/A	N/A	0.01	0.17	N/A	N/A	N/A	0.24
	b			0.15			0.35	0.27				0.12
	a			0.25			0.05	0.10				0.31
SBD	Scenario II behaviour?		YES	YES	YES	YES	YES	YES	YES	YES	YES	YES
	n		6	6	6	6	6	5	6	6	5	5
	R^2	no data	0.48	0.28	0.66	0.40	0.79	0.80	0.69	0.93	0.77	0.52
	p value		0.13	0.28	0.05	0.18	0.02	0.04	0.04	0.002	0.05	0.17
	b		0.36	0.17	0.29	0.21	0.87	0.49	0.35	0.40	0.16	0.62
	a		0.06	0.23	0.09	0.17	0.001	0.02	0.06	0.03	0.22	0.005
PPT	Scenario II behaviour?		NO	NO	NO	NO	YES	YES	NO	NO	NO	NO
	n		8	7	8	6	6	6	6	6	6	8
	R^2	no data					0.54	0.44				
	p value		N/A	N/A	N/A	N/A	0.10	0.15	N/A	N/A	N/A	N/A
	b						0.21	0.15				
	a						0.16	0.27				
STP	Scenario II behaviour?		NO	YES	NO	NO	YES	NO	YES	YES	YES	NO
	n		6	6	7	7	6	7	7	7	6	6
	R^2	no data		0.88			0.38		0.25	0.36	0.45	
	p value		N/A	0.01	N/A	N/A	0.19	N/A	0.25	0.16	0.14	N/A
	b			0.32			0.28		0.18	0.29	0.21	
	a			0.08			0.10		0.23	0.10	0.16	

Table S4: Regression parameters from scenario II plots (log RSD vs. log CTD) for years showing statistically significant scenario II relationship to 95% ($p < 0.05$).

	Log A (intercept)	$-b$ (slope)	a^1 ($\log A / v^{-b}$)
Alert	No years were significant		
EGH	0.71	0.23	0.14
	0.86	0.26	0.11
	2.01	0.59	0.01
	1.08	0.31	0.08
BNT	1.03	0.35	0.05
SBD	0.96	0.29	0.90
	3.20	0.87	0.001
	1.71	0.49	0.02
	1.24	0.35	0.06
	1.33	0.40	0.03
	0.38	0.16	0.22
STP	1.22	0.32	0.08
PPT	No years were significant		
Mean (\pm SD) for $-b$ and a values		0.39 ± 0.19	0.14 ± 0.25
Literature values ²		0.27 for small t_a 0.99 for large t_a	0.14

¹ Values of a were calculated from the intercept, $\log A$, using the equation $a = \log A / v^{-b}$, where v is the average wind speed of 4 m/s (126,144 km/yr) and $-b$, the slope

² a value, Junge 1974 (1); $-b$ value, Stroebe *et al.* 2006 (4)