Comparability of long-term temporal trends of POPs from co-located active and passive air monitoring networks in Europe

Jiří Kalina[a], Kevin B. White[a], Martin Scheringer[a,b], Petra Přibylová[a], Petr Kukučka[a], Ondřej Audy[a] and Jana Klánová[a]

[a] RECETOX Centre, Faculty of Science, Masaryk University, Kamenice 753/5, 625 00 Brno, Czech Republic
[b] Institute of Biogeochemistry and Pollutant Dynamics, ETH Zürich, 8092 Zürich, Switzerland

Electronic Supplementary Information

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Table S1 – Location, sampling and analytical details for EMEP active monitoring sites.

Site				Sampling				Analytical	
Name	**Country**	**Latt**	**Long**	**Years**	**Duration**		**Instrument**	**Method**	**Lab**
Birkenes	Norway	58.380	8.252	2003–2016	24 h	Weekly	High vol (480 m3/d) Glass fibre filter + 2 PUF	GC-MS	NILU (Norway)
Košetice	Czechia	49.573	15.080	1996–2016	24 h	Weekly	High vol (720 m3/d) Glass fibre filter + 2 PUF	GC-MS/MS	RECETOX (Czechia)
Pallas	Finland	68.000	24.246	1996–2016	7 d	Weekly	High vol (570 m3/d) Glass fibre filter + 3 PUF	HPLC, GC-MS, GC-ECD	IVL (Sweden)
Råö	Sweden	57.394	11.914	2002–2016	7 d	Weekly	High vol (570 m3/d) Glass fibre filter + 3 PUF	HPLC, GC-MS, GC-ECD	IVL (Sweden)
Stórhöfði	Iceland	63.400	-20.283	1995–2016		Bi-Weekly	Low vol (70 m3/d) PUF	GC-MS	
Zeppelin	Norway	78.880	11.883	1993–2016	48 h	Weekly	High vol (480 m3/d) Glass fibre filter + 2 PUF	GC-MS	NILU (Norway)

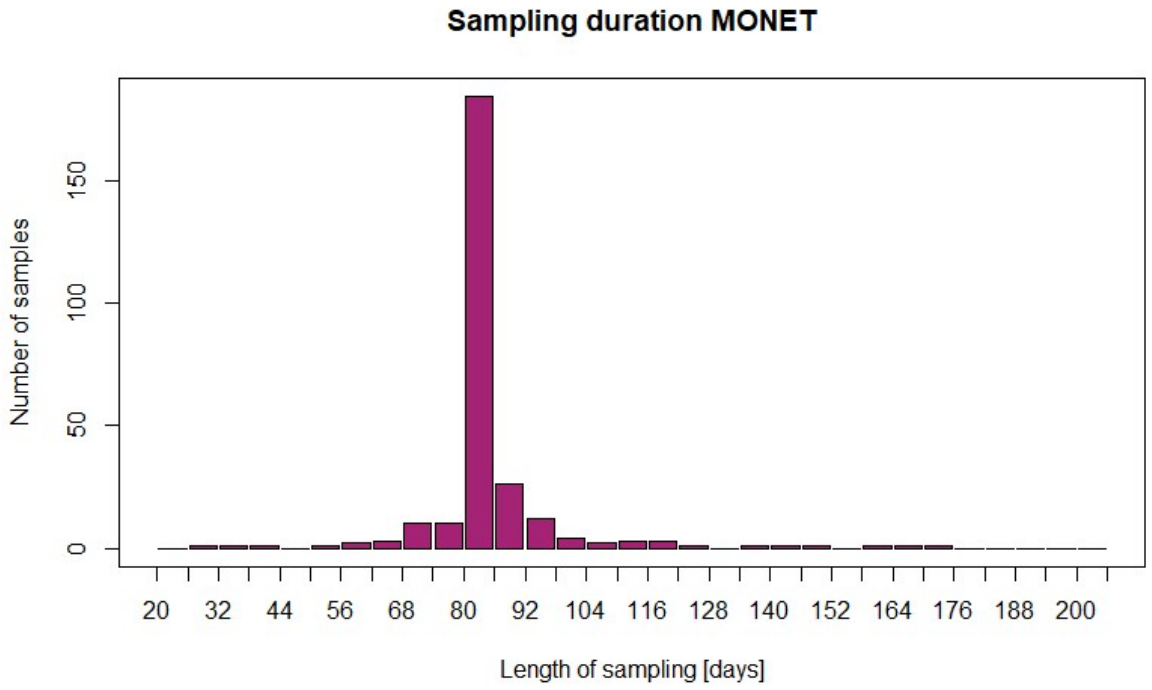
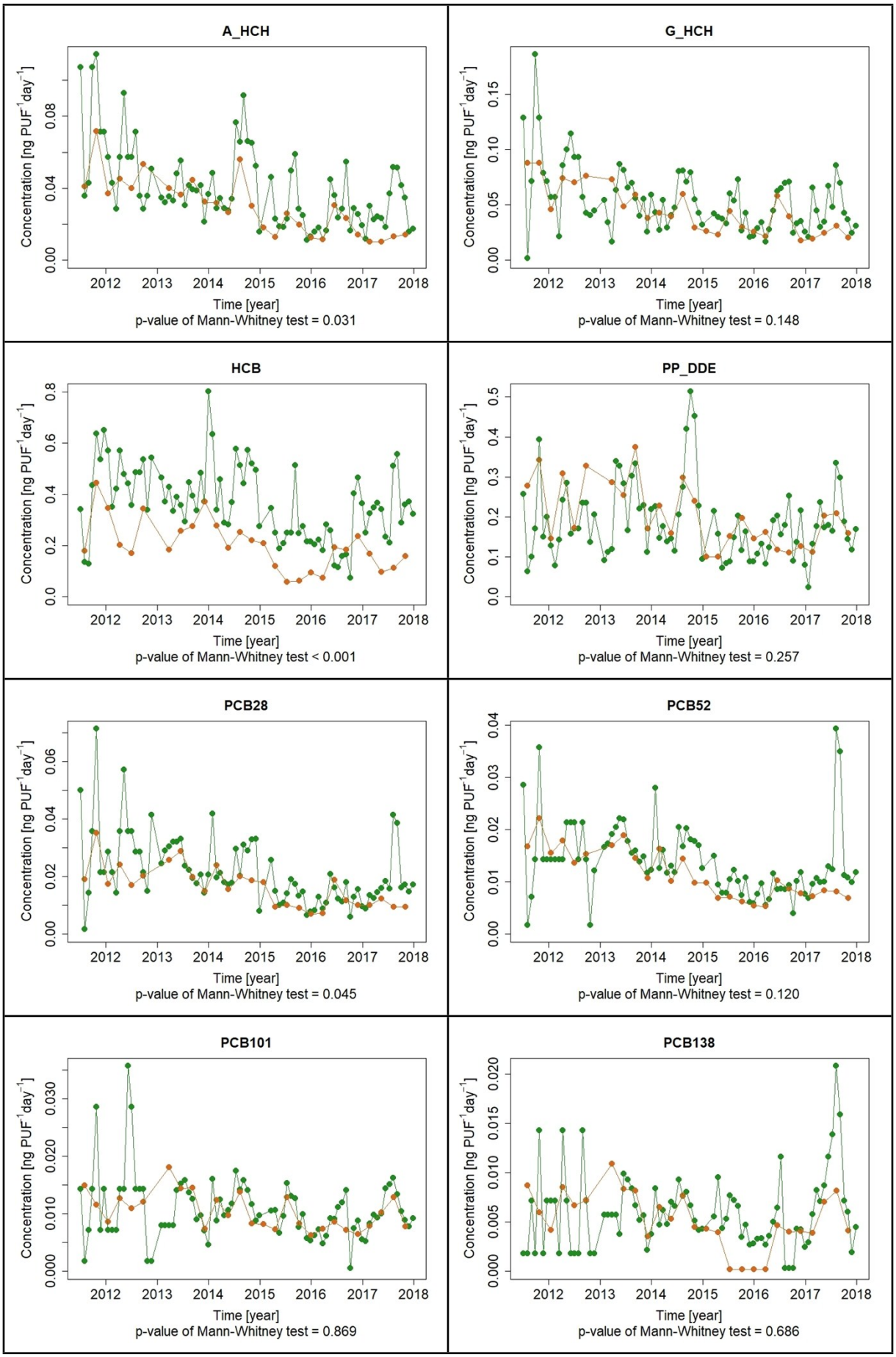


Figure S1 – Sampling duration distribution of passive MONET samples used in this study.


A_HCH
Concentration [ng PUF^{-1}day^{-1}]
0.00
0.04
0.08
2012
2013
2014
2015
2016
2017
2018
Time [year]
p-value of Mann-Whitney test = 0.031
G_HCH
Concentration [ng PUF^{-1}day^{-1}]
0.00
0.05
0.10
0.15
2012
2013
2014
2015
2016
2017
2018
Time [year]
p-value of Mann-Whitney test = 0.148
HCB
Concentration [ng PUF^{-1}day^{-1}]
0.0
0.2
0.4
0.6
0.8
2012
2013
2014
2015
2016
2017
2018
Time [year]
p-value of Mann-Whitney test < 0.001
PP_DDE
Concentration [ng PUF^{-1}day^{-1}]
0.0
0.1
0.2
0.3
0.4
0.5
2012
2013
2014
2015
2016
2017
2018
Time [year]
p-value of Mann-Whitney test = 0.257
PCB28
Concentration [ng PUF^{-1}day^{-1}]
0.00
0.02
0.04
0.06
2012
2013
2014
2015
2016
2017
2018
Time [year]
p-value of Mann-Whitney test = 0.045
PCB52
Concentration [ng PUF^{-1}day^{-1}]
0.00
0.01
0.02
0.03
0.04
2012
2013
2014
2015
2016
2017
2018
Time [year]
p-value of Mann-Whitney test = 0.120
PCB101
Concentration [ng PUF^{-1}day^{-1}]
0.000
0.010
0.020
0.030
2012
2013
2014
2015
2016
2017
2018
Time [year]
p-value of Mann-Whitney test = 0.869
PCB138
Concentration [ng PUF^{-1}day^{-1}]
0.000
0.005
0.010
0.015
0.020
2012
2013
2014
2015
2016
2017
2018
Time [year]
p-value of Mann-Whitney test = 0.686

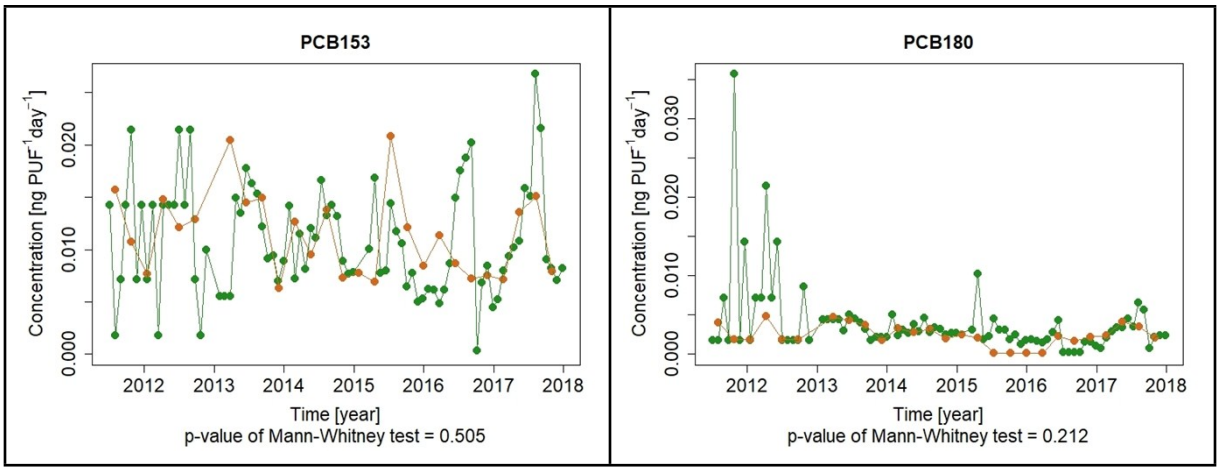


Figure S2 – Comparison of atmospheric concentrations measured by 28-day (green) and 84-day sampling (orange). Ten compounds are available from parallel monitoring at the same site between 2011 and 2017. The Mann-Whitney U test p-value is shown below each graph ($p<0.05$ indicates a significant difference between the two sampling durations).

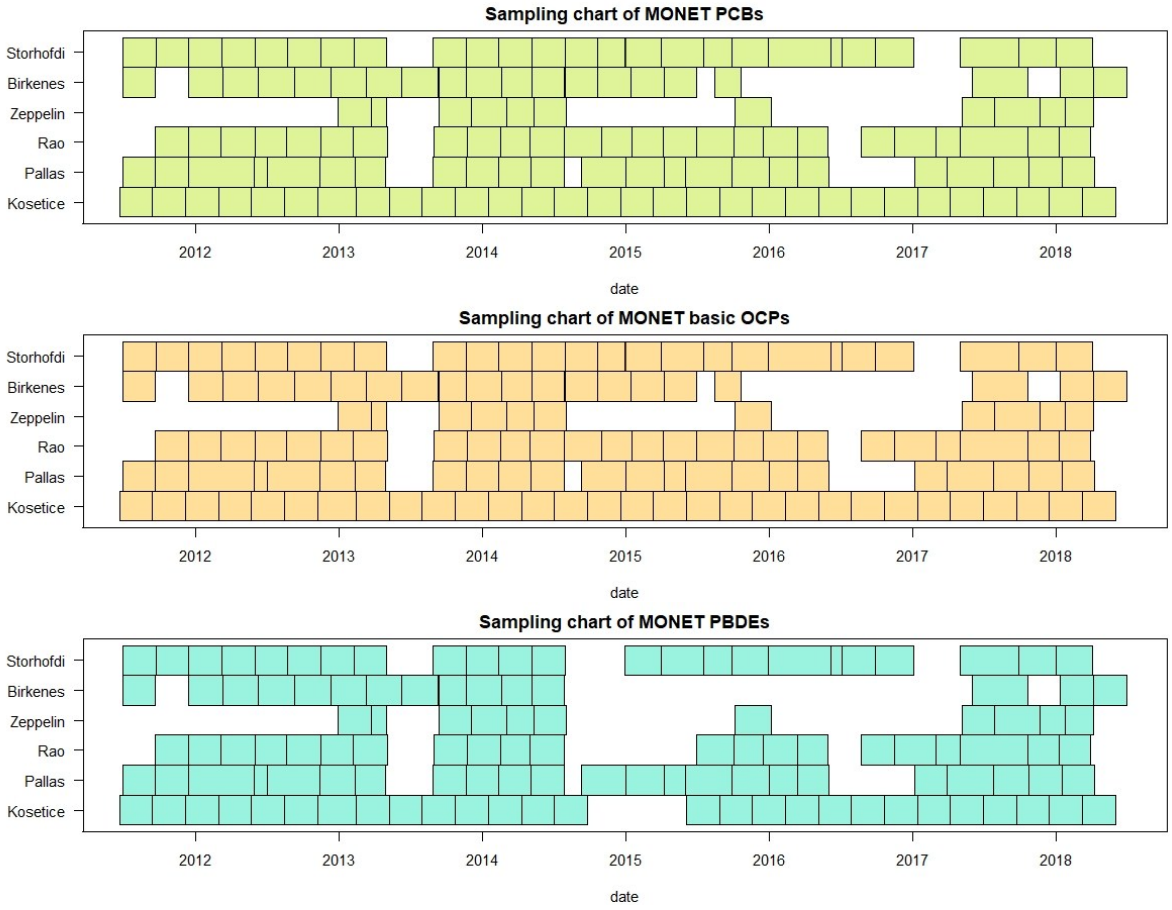
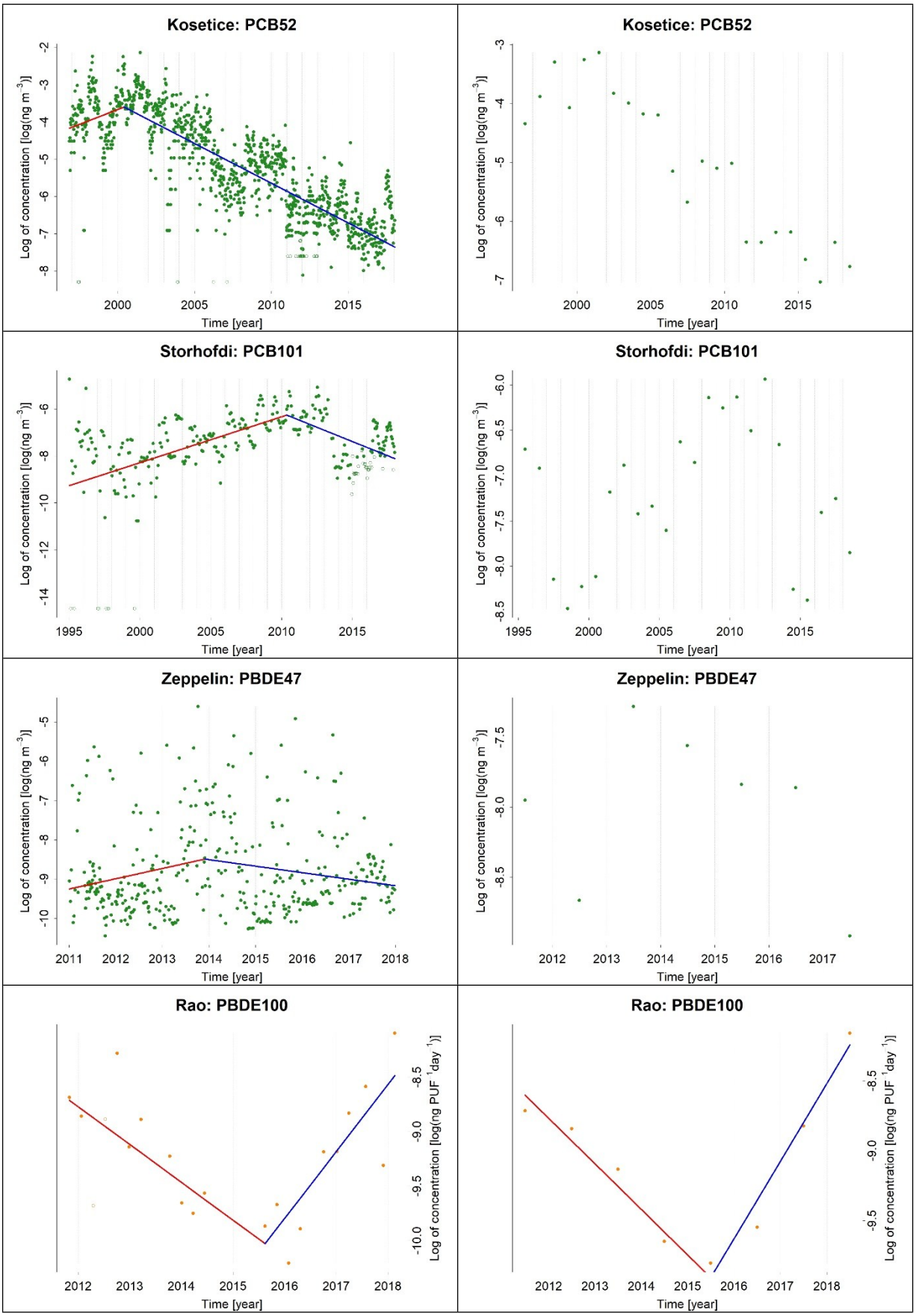


Figure S3 – Compound-specific temporal coverage of MONET site sampling regimes. Each coloured rectangle represents a single sample/value used in the temporal trend calculations.

Table S2 – Percentage (%) of samples/values below instrumental LOQ.

S2a – EMEP	Košetice	Pallas	Råö	Zeppelin	Birkenes	Stórhöfði
PCB 28	1	0	0	11	1	7
PCB 52	3	0	0	12	3	7
PCB 101	8	1	1	11	2	13
PCB 153	5	0	0	17	28	30
PCB 138	16	4	0	15	2	36
PCB 180	24	13	1	29	2	42
α-HCH	1	0	0	2	3	9
ɣ-HCH	3	2	0	2	3	10
HCB	0	14	14	1	1	3
p,p'-DDE	0	1	0	20	4	35
PBDE 47	*n/a*	13	5	3	2	62
PBDE 99	*n/a*	42	20	8	3	90
PBDE 100	*n/a*	94	48	29	18	90
PBDE 153	*n/a*	100	40	34	28	30
PBDE 154	*n/a*	100	48	36	29	*n/a*
PBDE 209	*n/a*	100	44	4	13	*n/a*

S2a – MONET	Košetice	Pallas	Råö	Zeppelin	Birkenes	Stórhöfði
PCB 28	0	0	0	9	5	0
PCB 52	0	0	0	0	10	0
PCB 101	0	35	0	18	45	0
PCB 153	0	57	0	18	45	0
PCB 138	13	91	4	27	45	0
PCB 180	27	96	16	36	50	23
α-HCH	0	0	0	0	0	0
ɣ-HCH	0	4	0	9	0	0
HCB	0	0	0	0	0	0
p,p'-DDE	0	0	0	0	0	0
PBDE 47	0	0	0	0	0	0
PBDE 99	0	0	14	0	0	0
PBDE 100	0	27	10	0	20	0
PBDE 153	70	80	62	91	73	42
PBDE 154	48	80	19	55	53	33
PBDE 209	15	0	14	9	7	33


Kosetice: PCB52
Log of concentration [log(ng m-3)]
Time [year]
Kosetice: PCB52
Log of concentration [log(ng m-3)]
Time [year]
Storhofdi: PCB101
Log of concentration [log(ng m-3)]
Time [year]
Storhofdi: PCB101
Log of concentration [log(ng m-3)]
Time [year]
Zeppelin: PBDE47
Log of concentration [log(ng m-3)]
Time [year]
Zeppelin: PBDE47
Log of concentration [log(ng m-3)]
Time [year]
Rao: PBDE100
Log of concentration [log(ng PUF-1day-1)]
Time [year]
Rao: PBDE100
Log of concentration [log(ng PUF-1day-1)]
Time [year]

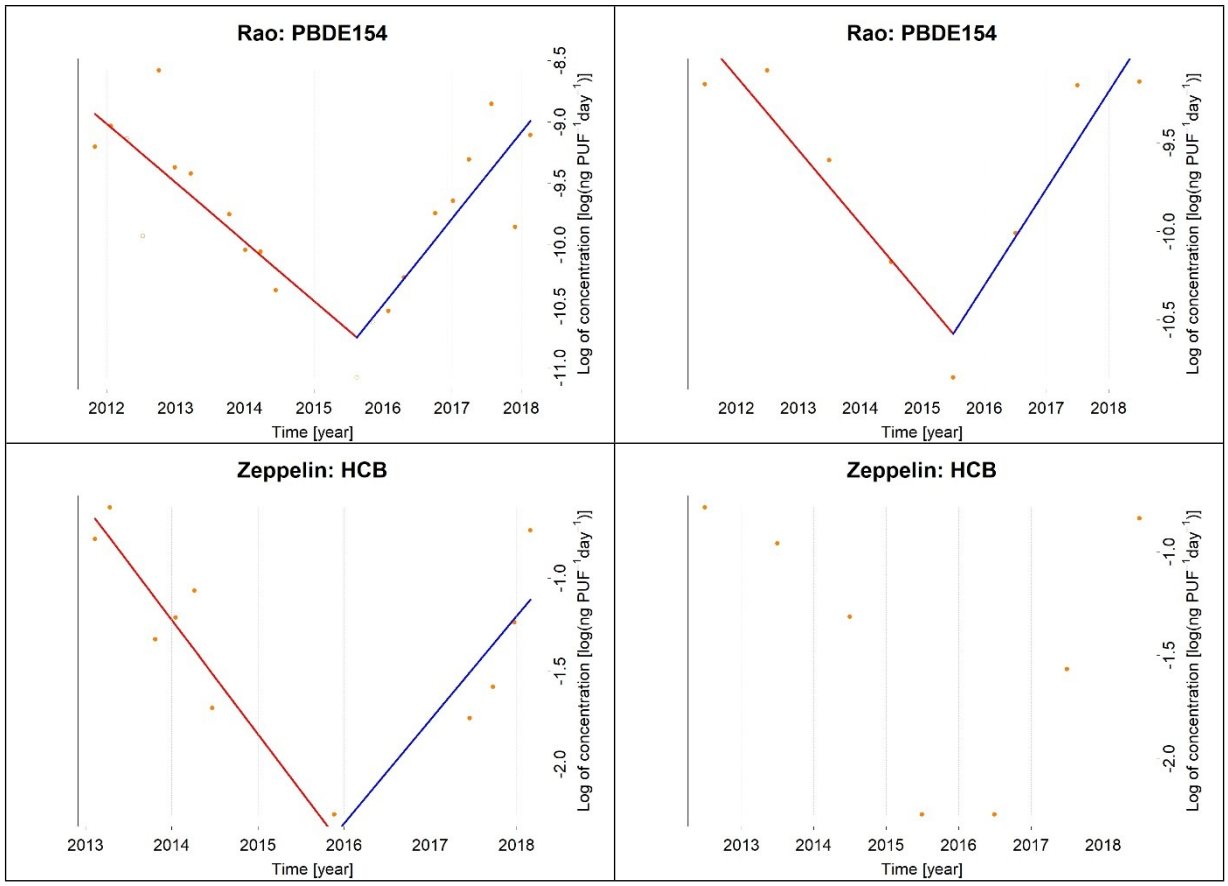


Figure S4 – Identified break points in primary data (left column) or annual averages (right column). The first three rows (green dots) are active EMEP data, the last three rows (orange dots) are passive MONET data.

Table S3 – Summary of compound-specific sampling rates for selected compounds (m^3/PUF/day) based on comparison of measured active and passive concentrations.

	PCB 28	PCB 153	α-HCH	γ-HCH	HCB	p,p'-DDE	PBDE 47	PBDE 99
Birkenes	4.1	5.3	2.8	4.0	1.7	5.7	3.1	5.4
Košetice	6.6	10.1	5.5	6.2	3.7	14.0	*n/a*	*n/a*
Pallas	4.8	7.4	5.0	6.9	6.5	9.7	3.3	5.7
Råö	9.4	10.5	8.6	11.1	10.3	23.3	6.2	5.4
Stórhöfði	8.6	53.6	56.7	39.2	70.1	262.2	3.1	14.4
Zeppelin	3.8	25.9	4.9	8.8	3.7	39.0	1.8	11.7

Table S4 – Rate constants of temporal trends (percent annual change, %/year). Asterisks denote statistical significance (p-values) of Kendall correlations (listed in Tables S5 & S6). *: 0.05–0.01; **: 0.01–0.001; ***: <0.001.

S4a – PCBs	**PCB 28**	**PCB 52**	**PCB 101**	**PCB 153**	**PCB 138**	**PCB 180**
Košetice_ACT	-13***	-17***	-17***	-18***	-20***	-19***
Košetice_PAS	-13***	-14***	-7**	-4	-8**	-4
Pallas_ACT	-4***	-3***	-4***	-5***	-6***	-8***
Pallas_PAS	-16**	-14**	-9	-13*	-24**	-30***
Råö_ACT	-6***	-4***	-4***	-3***	-4***	-5***
Råö_PAS	-11	-13**	-4	-4	-1	-7
Zeppelin_ACT	-7***	-6***	-5***	-7***	-9***	-12***
Zeppelin_PAS	-14	-12	-6	-8	9	-9
Birkenes_ACT	-8***	-6***	-6***	-6***	-9***	-11***
Birkenes_PAS	-9*	-8**	-2	-3	-17*	-28**
Stórhöfði_ACT	-2***	3**	-1	-1	1	2**
Stórhöfði_PAS	-20*	-18*	-12	-10	-9*	-11

S4b – OCPs	**α-HCH**	**ɣ-HCH**	**HCB**	**p,p'-DDE**
Košetice_ACT	-10***	-11***	-3***	-4***
Košetice_PAS	-20***	-19***	-13**	-9**
Pallas_ACT	-9***	-12***	1	-2*
Pallas_PAS	-22**	-25***	-10*	-12*
Råö_ACT	-10***	-11***	1	-3**
Råö_PAS	-19***	-18**	-8*	-8
Zeppelin_ACT	-12***	-14***	1***	-9***
Zeppelin_PAS	-3	-15	-9	5
Birkenes_ACT	-8***	-10***	-1*	1
Birkenes_PAS	-14**	-13*	-13**	3
Stórhöfði_ACT	-11***	-9***	-1	0
Stórhöfði_PAS	-22***	-26**	-11*	-16

S4c – PBDEs	**PBDE 47**	**PBDE 99**	**PBDE 100**	**PBDE 153**	**PBDE 154**	**PBDE 209**
Košetice_ACT	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*
Košetice_PAS	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*
Pallas_ACT	-20**	-16	22***	16*	16*	0
Pallas_PAS	-11	-9	2	-14	-10	43
Råö_ACT	-6	-13*	17***	0	16***	0
Råö_PAS	-2	-7	102**	-19*	159**	7
Zeppelin_ACT	2	-7***	3**	-8***	-13***	19***
Zeppelin_PAS	-2	-3	-1	-6	6	13
Birkenes_ACT	-1	-14***	1*	-4**	-2**	9***
Birkenes_PAS	-13	-21	-9	-10	-10	39
Stórhöfði_ACT	-20***	25***	25***	n/a	n/a	n/a
Stórhöfði_PAS	44	88	91*	n/a	n/a	n/a

Table S5 – Halving times of temporal trends (percent annual change, %/year). Asterisks denote statistical significance (p-values) of Kendall correlations (listed in Tables S5 & S6). *: 0.05–0.01; **: 0.01–0.001; ***: <0.001.

S5a – PCBs	**PCB 28**	**PCB 52**	**PCB 101**	**PCB 153**	**PCB 138**	**PCB1 80**
Košetice_ACT	5***	3***	4***	3***	3***	3***
Košetice_PAS	5***	5***	9**	15	8**	18
Pallas_ACT	16***	20***	16***	13***	11***	8***
Pallas_PAS	4**	5**	8	5*	3**	2***
Råö_ACT	11***	18***	17***	22***	17***	15***
Råö_PAS	6	5**	16	19	82	10
Zeppelin_ACT	11***	12***	13***	10***	8***	6***
Zeppelin_PAS	4	5	11	8	-8	8
Birkenes_ACT	9***	12***	12***	11***	8***	6***
Birkenes_PAS	7*	9**	31	21	4*	2**
Stórhöfði_ACT	30***	-26**	113	92	-118	-37**
Stórhöfði_PAS	3*	3*	5	7	7*	6

S5b – OCPs	**α-HCH**	**ɣ-HCH**	**HCB**	**p,p'-DDE**
Košetice_ACT	6***	6****	21***	16***
Košetice_PAS	3***	3***	5**	7**
Pallas_ACT	7***	5***	-97	41*
Pallas_PAS	3**	2***	7*	5*
Råö_ACT	7***	6***	-55	36**
Råö_PAS	3***	3**	9*	8
Zeppelin_ACT	6***	5***	-108***	8***
Zeppelin_PAS	20	4	8	-15
Birkenes_ACT	8***	7***	83*	-64
Birkenes_PAS	5**	5*	5**	-27
Stórhöfði_ACT	6***	8***	118	∞
Stórhöfði_PAS	3***	2**	6*	4

S5c – PBDEs	PBDE 47	PBDE 99	PBDE 100	PBDE 153	PBDE 154	PBDE 209
Košetice_ACT	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*
Košetice_PAS	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*
Pallas_ACT	3**	4	-4***	-4*	-5*	∞
Pallas_PAS	6	7	-40	4	7	-2
Råö_ACT	11	5*	-5**	∞	-5***	∞
Råö_PAS	44	9	-1**	3*	-1**	-10
Zeppelin_ACT	-33	10***	-26**	8**	5**	-4***
Zeppelin_PAS	28	27	130	12	-12	-6
Birkenes_ACT	66	5***	-72*	19**	36**	-8***
Birkenes_PAS	5	3	7	7	6	-2
Stórhöfði_ACT	3***	-3***	-3***	*n/a*	*n/a*	*n/a*
Stórhöfði_PAS	-2	-1	-1*	*n/a*	*n/a*	*n/a*

Table S6 – Kendall correlations of individual temporal trends.

S6a – PCBs	PCB 28	PCB 52	PCB 101	PCB 153	PCB 138	PCB 180
Košetice_ACT	-0.577	-0.612	-0.535	-0.617	-0.617	-0.595
Košetice_PAS	-0.490	-0.585	-0.364	-0.217	-0.337	-0.147
Pallas_ACT	-0.289	-0.241	-0.293	-0.364	-0.365	-0.412
Pallas_PAS	-0.455	-0.455	-0.281	-0.325	-0.520	-0.567
Råö_ACT	-0.492	-0.313	-0.275	-0.205	-0.251	-0.300
Råö_PAS	-0.320	-0.367	-0.120	-0.087	-0.017	-0.120
Zeppelin_ACT	-0.364	-0.424	-0.378	-0.293	-0.389	-0.414
Zeppelin_PAS	-0.127	-0.200	-0.055	-0.073	0.164	-0.273
Birkenes_ACT	-0.286	-0.238	-0.187	-0.169	-0.252	-0.297
Birkenes_PAS	-0.358	-0.438	-0.122	-0.154	-0.409	-0.472
Stórhöfði_ACT	-0.134	0.124	-0.021	-0.045	0.032	0.099
Stórhöfði_PAS	-0.307	-0.340	-0.267	-0.253	-0.287	-0.268

S6b – OCPs	α-HCH	ɣ-HCH	HCB	p,p'-DDE
Košetice_ACT	-0.433	-0.396	-0.202	-0.238
Košetice_PAS	-0.641	-0.641	-0.434	-0.338
Pallas_ACT	-0.548	-0.621	0.031	-0.098
Pallas_PAS	-0.455	-0.660	-0.344	-0.296
Råö_ACT	-0.636	-0.588	0.044	-0.129
Råö_PAS	-0.520	-0.433	-0.293	-0.193
Zeppelin_ACT	-0.689	-0.685	0.125	-0.311
Zeppelin_PAS	-0.164	-0.309	-0.200	0.164
Birkenes_ACT	-0.356	-0.235	-0.067	0.021
Birkenes_PAS	-0.474	-0.337	-0.453	0.095
Stórhöfði_ACT	-0.660	-0.561	-0.050	-0.008
Stórhöfði_PAS	-0.507	-0.473	-0.340	-0.260

S6c – PBDEs	PBDE 47	PBDE 99	PBDE 100	PBDE 153	PBDE 154	PBDE 209
Košetice_ACT	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*
Košetice_PAS	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*
Pallas_ACT	-0.287	-0.154	0.655	0.304	0.304	0.545
Pallas_PAS	-0.059	-0.051	0.012	-0.091	-0.083	0.248
Råö_ACT	-0.133	-0.204	0.390	-0.091	0.491	-0.067
Råö_PAS	-0.019	-0.133	0.644	-0.324	0.778	0.067
Zeppelin_ACT	0.034	-0.158	0.111	-0.467	-0.519	0.297
Zeppelin_PAS	-0.018	-0.091	-0.018	-0.091	0.091	0.127
Birkenes_ACT	-0.037	-0.301	0.071	-0.128	-0.114	0.192
Birkenes_PAS	-0.276	-0.352	-0.181	-0.162	-0.181	0.371
Stórhöfði_ACT	-0.321	0.515	0.523	*n/a*	*n/a*	*n/a*
Stórhöfði_PAS	0.249	0.257	0.312	*n/a*	*n/a*	*n/a*

Table S7 – Statistical significance (p-values) of Kendall correlations listed in Table S5.

S7a – PCBs	**PCB 28**	**PCB 52**	**PCB 101**	**PCB 153**	**PCB 138**	**PCB 180**
Košetice_ACT	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Košetice_PAS	<0.001	<0.001	0.005	0.093	0.009	0.260
Pallas_ACT	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Pallas_PAS	0.002	0.002	0.064	0.030	0.001	<0.001
Råö_ACT	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Råö_PAS	0.025	0.010	0.418	0.563	0.907	0.418
Zeppelin_ACT	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Zeppelin_PAS	0.648	0.445	0.879	0.755	0.542	0.283
Birkenes_ACT	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
Birkenes_PAS	0.028	0.007	0.455	0.346	0.012	0.004
Stórhöfði_ACT	<0.001	0.001	0.611	0.230	0.389	0.008
Stórhöfði_PAS	0.033	0.017	0.065	0.080	0.046	0.061

S7b – OCPs	**α-HCH**	**ɣ-HCH**	**HCB**	**p,p'-DDE**
Košetice_ACT	<0.001	<0.001	<0.001	<0.001
Košetice_PAS	<0.001	<0.001	0.001	0.008
Pallas_ACT	<0.001	<0.001	0.668	0.022
Pallas_PAS	0.002	<0.001	0.022	0.050
Råö_ACT	<0.001	<0.001	0.551	0.002
Råö_PAS	<0.001	0.002	0.041	0.185
Zeppelin_ACT	<0.001	<0.001	<0.001	<0.001
Zeppelin_PAS	0.542	0.218	0.445	0.542
Birkenes_ACT	<0.001	<0.001	0.031	0.564
Birkenes_PAS	0.003	0.040	0.005	0.586
Stórhöfði_ACT	<0.001	<0.001	0.174	0.841
Stórhöfði_PAS	<0.001	0.001	0.017	0.072

S7c – PBDEs	PBDE 47	PBDE 99	PBDE 100	PBDE 153	PBDE 154	PBDE 209
Košetice_ACT	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*
Košetice_PAS	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*	*n/a*
Pallas_ACT	0.005	0.131	<0.001	0.018	0.018	<0.001
Pallas_PAS	0.715	0.754	0.958	0.565	0.601	0.125
Råö_ACT	0.137	0.024	<0.001	0.415	<0.001	0.561
Råö_PAS	0.929	0.420	0.009	0.042	0.001	0.698
Zeppelin_ACT	0.316	<0.001	0.002	<0.001	<0.001	<0.001
Zeppelin_PAS	1.000	0.761	1.000	0.761	0.761	0.648
Birkenes_ACT	0.331	<0.001	0.064	0.001	0.003	<0.001
Birkenes_PAS	0.169	0.074	0.379	0.435	0.379	0.059
Stórhöfði_ACT	<0.001	<0.001	<0.001	*n/a*	*n/a*	*n/a*
Stórhöfði_PAS	0.102	0.091	0.038	*n/a*	*n/a*	*n/a*

Table S8 – Literature halving times and rate constants for POPs at the selected sites.

Compound	Site	Period	Halving time (years)	Rate constant (%/year)	Type of sampling	Reference
PCB28	Košetice	1999–2013	4.6	-15.1	active	Wöhrnschimmel et al., 2016
PCB28	Košetice	2003–2013	6.7	-10.3	active	Kalina et al., 2017
PCB28	Košetice	2003–2013	8	-8.7	passive	Kalina et al., 2017
PCB28	Košetice	2003–2015	11.1	-6.2	passive	Kalina et al., 2018
PCB28	Pallas	1996–2005	18	-3.9	active	Hung et al., 2010
PCB28	Pallas	1996–2011	18	-3.9	active	AMAP, 2014
PCB28	Pallas	1996–2011	24.8	-2.8	active	Anttila et al., 2016
PCB28	Pallas	1996–2012	15	-4.6	active	Hung et al., 2016
PCB28	Pallas	1996–2013	15.8	-4.4	active	Wöhrnschimmel et al., 2016
PCB28	Råö	1997–2011	27.7	-2.5	active	Anttila et al., 2016
PCB28	Stórhöfði	1995–2005	19	-3.6	active	Hung et al., 2010
PCB28	Stórhöfði	1995–2011	20	-3.5	active	AMAP, 2014
PCB28	Stórhöfði	1995–2012	24	-2.9	active	Hung et al., 2016
PCB28	Stórhöfði	1995–2013	15.5	-4.5	active	Wöhrnschimmel et al., 2016
PCB28	Zeppelin	1993–2014	8.5	-8.2	active	Wöhrnschimmel et al., 2016
PCB28	Zeppelin	1998–2006	6	-11.6	active	Hung et al., 2010
PCB28	Zeppelin	1998–2012	8.2	-8.5	active	AMAP, 2014
PCB28	Zeppelin	1998–2012	8.2	-8.5	active	Hung et al., 2016
PCB52	Košetice	1999–2013	3.7	-18.7	active	Wöhrnschimmel et al., 2016
PCB52	Košetice	2003–2013	3.1	-22.4	active	Kalina et al., 2017
PCB52	Košetice	2003–2013	3.1	-22.4	passive	Kalina et al., 2017
PCB52	Košetice	2003–2015	4.5	-15.4	passive	Kalina et al., 2018
PCB52	Pallas	1996–2005	47	-1.5	active	Hung et al., 2010
PCB52	Pallas	1996–2011	38	-1.8	active	AMAP, 2014

PCB52	Pallas	1996–2011	43.3	-1.6	active	Anttila et al., 2016
PCB52	Pallas	1996–2012	17	-4.1	active	Hung et al., 2016
PCB52	Pallas	1996–2013	19.1	-3.6	active	Wöhrnschimmel et al., 2016
PCB52	Råö	1994–2011	25.7	-2.7	active	Anttila et al., 2016
PCB52	Stórhöfði	1995–2005	22	-3.2	active	Hung et al., 2010
PCB52	Stórhöfði	1995–2011	-11	6.3	active	AMAP, 2014
PCB52	Stórhöfði	1995–2012	-8.7	8	active	Hung et al., 2016
PCB52	Stórhöfði	1995–2013	-14	5	active	Wöhrnschimmel et al., 2016
PCB52	Zeppelin	1993–2014	10.7	-6.5	active	Wöhrnschimmel et al., 2016
PCB52	Zeppelin	1998–2006	8.3	-8.4	active	Hung et al., 2010
PCB52	Zeppelin	1998–2012	4.4	-15.8	active	AMAP, 2014
PCB52	Zeppelin	1998–2012	4.4	-15.8	active	Hung et al., 2016
PCB101	Košetice	2003–2013	4.3	-16.1	active	Kalina et al., 2017
PCB101	Košetice	2003–2013	3.8	-18.2	passive	Kalina et al., 2017
PCB101	Košetice	2003–2015	63.6	-1.1	passive	Kalina et al., 2018
PCB101	Pallas	1996–2005	11	-6.3	active	Hung et al., 2010
PCB101	Pallas	1996–2011	17	-4.1	active	AMAP, 2014
PCB101	Pallas	1996–2011	19.8	-3.5	active	Anttila et al., 2016
PCB101	Pallas	1996–2012	14	-5	active	Hung et al., 2016
PCB101	Pallas	1996–2013	13.8	-5	active	Wöhrnschimmel et al., 2016
PCB101	Råö	1997–2012	20.4	-3.4	active	Anttila et al., 2016
PCB101	Stórhöfði	1995–2011	-6.1	11.4	active	AMAP, 2014
PCB101	Stórhöfði	1995–2012	-6.2	11.2	active	Hung et al., 2016
PCB101	Stórhöfði	1995–2013	-7.5	9.2	active	Wöhrnschimmel et al., 2016
PCB101	Zeppelin	1993–2014	10.3	-6.7	active	Wöhrnschimmel et al., 2016
PCB101	Zeppelin	1998–2006	9.6	-7.2	active	Hung et al., 2010
PCB101	Zeppelin	1998–2012	11	-6.3	active	AMAP, 2014

PCB101	Zeppelin	1998–2012	11	-6.3	active	Hung et al., 2016
PCB138	Košetice	2003–2013	3.1	-22.4	active	Kalina et al., 2017
PCB138	Košetice	2003–2013	2.6	-26.7	passive	Kalina et al., 2017
PCB138	Košetice	2003–2015	8.8	-7.9	passive	Kalina et al., 2018
PCB138	Pallas	1996–2005	5.7	-12.2	active	Hung et al., 2010
PCB138	Pallas	1996–2011	8.9	-7.8	active	AMAP, 2014
PCB138	Pallas	1996–2012	10	-6.9	active	Hung et al., 2016
PCB138	Pallas	1996–2013	9.4	-7.4	active	Wöhrnschimmel et al., 2016
PCB138	Pallas	1997–2011	18.2	-3.8	active	Anttila et al., 2016
PCB138	Råö	1997–2013	21	-3.3	active	Anttila et al., 2016
PCB138	Stórhöfði	1995–2005	5.5	-12.6	active	Hung et al., 2010
PCB138	Stórhöfði	1995–2011	12	-5.8	active	AMAP, 2014
PCB138	Stórhöfði	1995–2012	15	-4.6	active	Hung et al., 2016
PCB138	Stórhöfði	1995–2013	7.4	-9.4	active	Wöhrnschimmel et al., 2016
PCB138	Zeppelin	1993–2014	5.7	-12.2	active	Wöhrnschimmel et al., 2016
PCB138	Zeppelin	1998–2006	3.6	-19.3	active	Hung et al., 2010
PCB138	Zeppelin	1998–2012	5.1	-13.6	active	AMAP, 2014
PCB138	Zeppelin	1998–2012	5.1	-13.6	active	Hung et al., 2016
PCB153	Košetice	2003–2013	4.4	-15.8	active	Kalina et al., 2017
PCB153	Košetice	2003–2013	3.6	-19.3	passive	Kalina et al., 2017
PCB153	Košetice	2003–2015	9.8	-7.1	passive	Kalina et al., 2018
PCB153	Pallas	1996–2005	7.7	-9	active	Hung et al., 2010
PCB153	Pallas	1996–2011	9.8	-7.1	active	AMAP, 2014
PCB153	Pallas	1996–2012	11	-6.3	active	Hung et al., 2016
PCB153	Pallas	1996–2013	10.5	-6.6	active	Wöhrnschimmel et al., 2016
PCB153	Pallas	1997–2011	19.3	-3.6	active	Anttila et al., 2016
PCB153	Råö	1994–2012	23.1	-3	active	Anttila et al., 2016
PCB153	Stórhöfði	1995–2005	6.5	-10.7	active	Hung et al., 2010

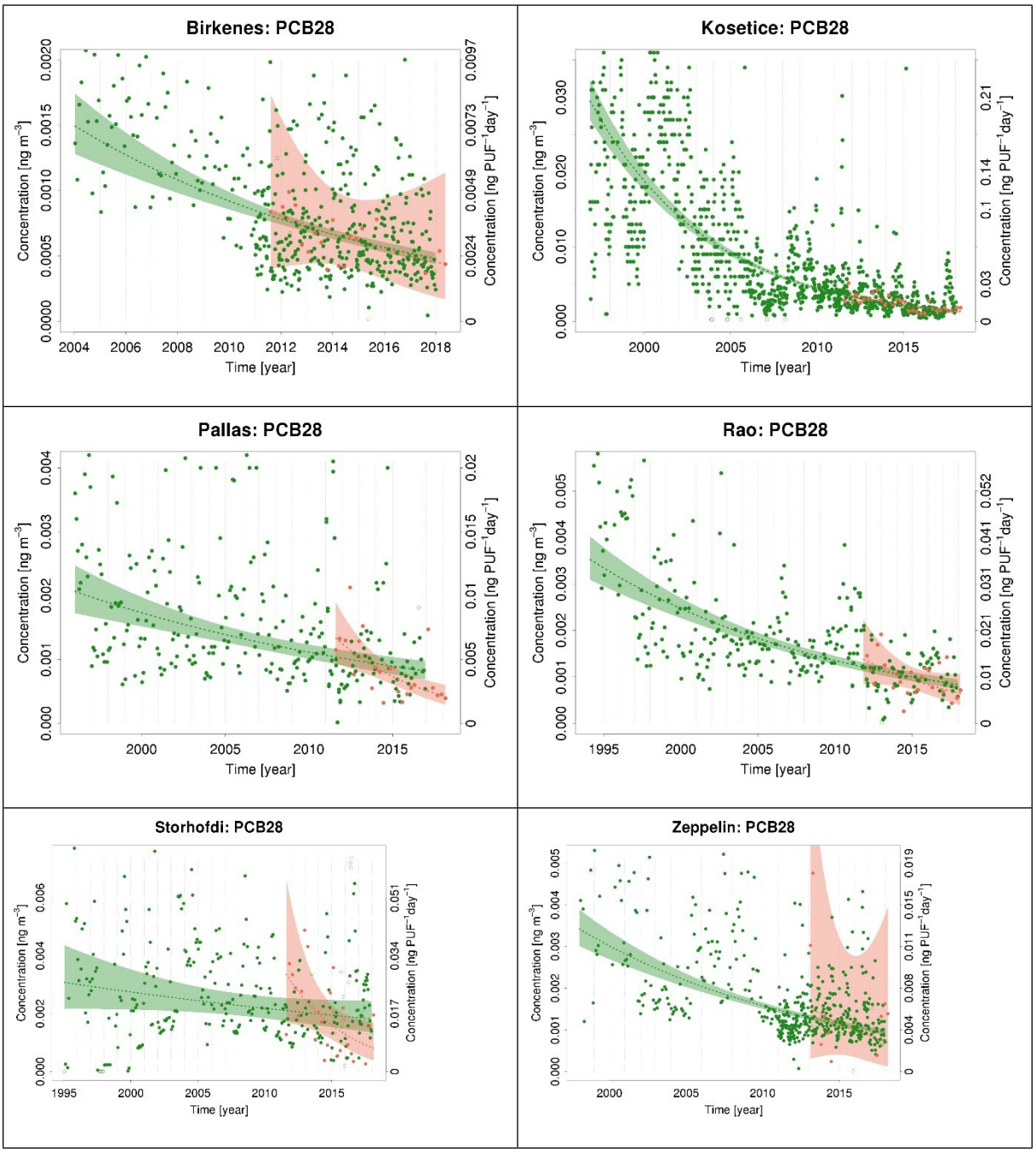
PCB153	Stórhöfði	1995–2011	19	-3.6	active	AMAP, 2014
PCB153	Stórhöfði	1995–2012	27	-2.6	active	Hung et al., 2016
PCB153	Stórhöfði	1995–2013	14.6	-4.7	active	Wöhrnschimmel et al., 2016
PCB153	Zeppelin	1993–2014	6.5	-10.7	active	Wöhrnschimmel et al., 2016
PCB153	Zeppelin	1998–2006	6.3	-11	active	Hung et al., 2010
PCB153	Zeppelin	1998–2012	6.2	-11.2	active	AMAP, 2014
PCB153	Zeppelin	1998–2012	6.2	-11.2	active	Hung et al., 2016
PCB180	Košetice	2003–2013	3.6	-19.3	active	Kalina et al., 2017
PCB180	Košetice	2003–2013	3.4	-20.4	passive	Kalina et al., 2017
PCB180	Košetice	2003–2015	7.7	-9	passive	Kalina et al., 2018
PCB180	Pallas	1996–2005	3.4	-20.4	active	Hung et al., 2010
PCB180	Pallas	1996–2011	5.1	-13.6	active	AMAP, 2014
PCB180	Pallas	1996–2012	5.9	-11.7	active	Hung et al., 2016
PCB180	Pallas	1996–2013	6.1	-11.4	active	Wöhrnschimmel et al., 2016
PCB180	Pallas	1997–2011	12.6	-5.5	active	Anttila et al., 2016
PCB180	Råö	1994–2013	15.8	-4.4	active	Anttila et al., 2016
PCB180	Stórhöfði	1995–2005	5.4	-12.8	active	Hung et al., 2010
PCB180	Stórhöfði	1995–2011	8.4	-8.3	active	AMAP, 2014
PCB180	Stórhöfði	1995–2012	8.4	-8.3	active	Hung et al., 2016
PCB180	Stórhöfði	1995–2013	11.3	-6.1	active	Wöhrnschimmel et al., 2016
PCB180	Zeppelin	1993–2014	4.5	-15.4	active	Wöhrnschimmel et al., 2016
PCB180	Zeppelin	1998–2006	3.9	-17.8	active	Hung et al., 2010
PCB180	Zeppelin	1998–2012	4.4	-15.8	active	AMAP, 2014
PCB180	Zeppelin	1998–2012	4.4	-15.8	active	Hung et al., 2016
A_HCH	Košetice	1999–2013	9.6	-7.2	active	Wöhrnschimmel et al., 2016
A_HCH	Košetice	2003–2013	9.3	-7.5	active	Kalina et al., 2017
A_HCH	Košetice	2003–2013	5.7	-12.2	passive	Kalina et al., 2017

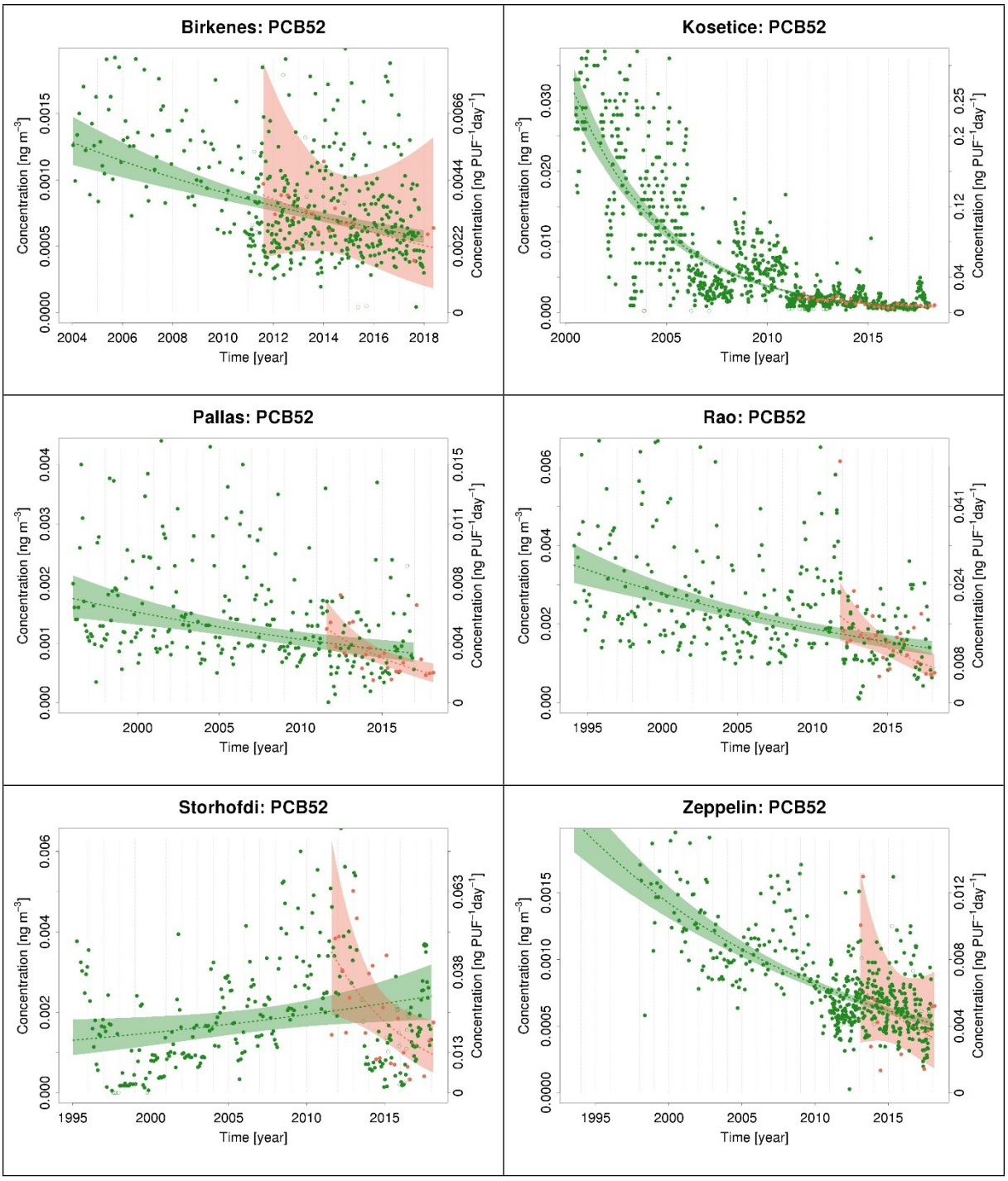
A_HCH	Košetice	2003–2015	11	-6.3	passive	Kalina et al., 2018
A_HCH	Pallas	1996–2005	6.1	-11.4	active	Hung et al., 2010
A_HCH	Pallas	1996–2011	5.7	-12.2	active	AMAP, 2014
A_HCH	Pallas	1996–2012	5.8	-12	active	Hung et al., 2016
A_HCH	Pallas	1996–2013	6.1	-11.4	active	Wöhrnschimmel et al., 2016
A_HCH	Stórhöfði	1995–2005	4.3	-16.1	active	Hung et al., 2010
A_HCH	Stórhöfði	1995–2011	5.6	-12.4	active	AMAP, 2014
A_HCH	Stórhöfði	1995–2012	5.4	-12.8	active	Hung et al., 2016
A_HCH	Stórhöfði	1995–2013	5.4	-12.8	active	Wöhrnschimmel et al., 2016
A_HCH	Zeppelin	1993–2006	4.7	-14.7	active	Hung et al., 2010
A_HCH	Zeppelin	1993–2012	4.9	-14.1	active	AMAP, 2014
A_HCH	Zeppelin	1993–2012	4.9	-14.1	active	Hung et al., 2016
A_HCH	Zeppelin	1993–2014	5.1	-13.6	active	Wöhrnschimmel et al., 2016
G_HCH	Košetice	1999–2013	9.2	-7.5	active	Wöhrnschimmel et al., 2016
G_HCH	Košetice	2003–2013	3	-23.1	active	Kalina et al., 2017
G_HCH	Košetice	2003–2013	4.6	-15.1	passive	Kalina et al., 2017
G_HCH	Košetice	2003–2015	5.2	-13.3	passive	Kalina et al., 2018
G_HCH	Pallas	1996–2005	4.6	-15.1	active	Hung et al., 2010
G_HCH	Pallas	1996–2011	4.2	-16.5	active	AMAP, 2014
G_HCH	Pallas	1996–2012	4.3	-16.1	active	Hung et al., 2016
G_HCH	Pallas	1996–2013	4.6	-15.1	active	Wöhrnschimmel et al., 2016
G_HCH	Stórhöfði	1995–2005	10	-6.9	active	Hung et al., 2010
G_HCH	Stórhöfði	1995–2011	7.7	-9	active	AMAP, 2014
G_HCH	Stórhöfði	1995–2012	7.3	-9.5	active	Hung et al., 2016
G_HCH	Stórhöfði	1995–2013	8	-8.7	active	Wöhrnschimmel et al., 2016
G_HCH	Zeppelin	1993–2006	4.6	-15.1	active	Hung et al., 2010
G_HCH	Zeppelin	1993–2012	7.1	-9.8	active	AMAP, 2014

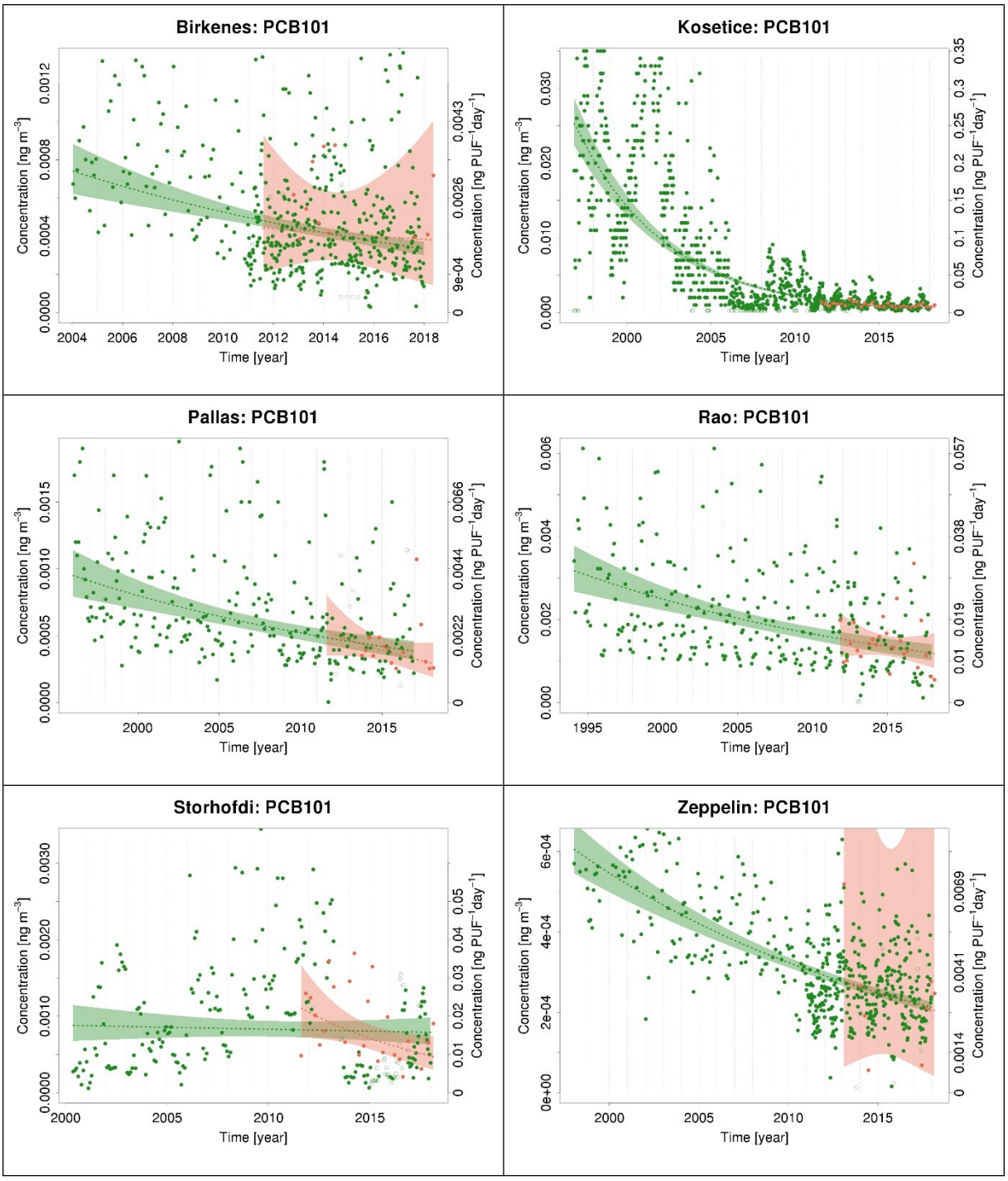
G_HCH	Zeppelin	1993–2012	4.1	-16.9	active	Hung et al., 2016
G_HCH	Zeppelin	1993–2014	4.2	-16.5	active	Wöhrnschimmel et al., 2016
HCB	Košetice	1999–2013	-4	17.3	active	Wöhrnschimmel et al., 2016
HCB	Košetice	2003–2013	12.8	-5.4	active	Kalina et al., 2017
HCB	Košetice	2003–2013	6.3	-11	passive	Kalina et al., 2017
HCB	Košetice	2003–2015	10.7	-6.5	passive	Kalina et al., 2018
HCB	Stórhöfði	1995–2005	6.2	-11.2	active	Hung et al., 2010
HCB	Stórhöfði	1995–2011	20	-3.5	active	AMAP, 2014
HCB	Stórhöfði	1995–2012	19	-3.6	active	Hung et al., 2016
HCB	Stórhöfði	1995–2013	17.8	-3.9	active	Wöhrnschimmel et al., 2016
HCB	Zeppelin	1993–2006	18	-3.9	active	Hung et al., 2010
HCB	Zeppelin	1993–2014	80.4	-0.9	active	Wöhrnschimmel et al., 2016
PP_DDE	Košetice	1999–2013	16.9	-4.1	active	Wöhrnschimmel et al., 2016
PP_DDE	Košetice	2003–2013	67.5	-1	active	Kalina et al., 2017
PP_DDE	Košetice	2003–2013	19.6	-3.5	passive	Kalina et al., 2017
PP_DDE	Košetice	2003–2015	62.1	-1.1	passive	Kalina et al., 2018
PP_DDE	Pallas	1996–2011	43	-1.6	active	AMAP, 2014
PP_DDE	Pallas	1996–2012	56	-1.2	active	Hung et al., 2016
PP_DDE	Pallas	1996–2013	37.7	-1.8	active	Wöhrnschimmel et al., 2016
PP_DDE	Stórhöfði	1995–2011	8.9	-7.8	active	AMAP, 2014
PP_DDE	Stórhöfði	1995–2012	9.8	-7.1	active	Hung et al., 2016
PP_DDE	Stórhöfði	1995–2013	11	-6.3	active	Wöhrnschimmel et al., 2016
PP_DDE	Zeppelin	1993–2006	11	-6.3	active	Hung et al., 2010
PP_DDE	Zeppelin	1993–2012	8	-8.7	active	AMAP, 2014
PP_DDE	Zeppelin	1993–2012	8	-8.7	active	Hung et al., 2016
PP_DDE	Zeppelin	1993–2014	7.8	-8.9	active	Wöhrnschimmel et al., 2016

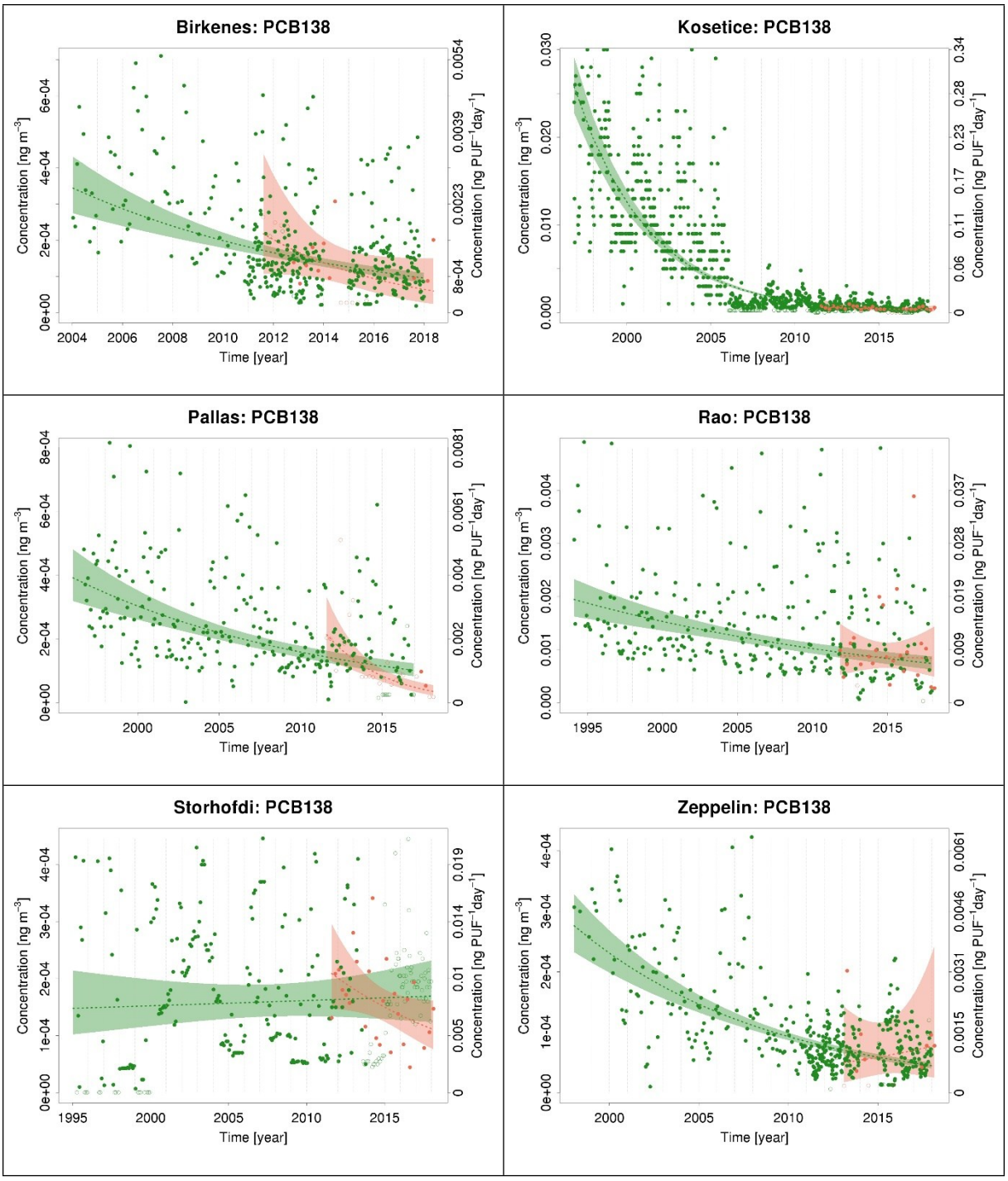
PBDE47	Pallas	2003–2011	4.5	-15.4	active	AMAP, 2014
PBDE47	Pallas	2003–2012	4.4	-15.8	active	Hung et al., 2016
PBDE47	Stórhöfði	2007–2012	3.7	-18.7	active	Hung et al., 2016
PBDE47	Zeppelin	1993–2014	8	-8.7	active	Wöhrnschimmel et al., 2016
PBDE47	Zeppelin	2006–2012	2.6	-26.7	active	AMAP, 2014
PBDE47	Zeppelin	2006–2012	2.6	-26.7	active	Hung et al., 2016
PBDE99	Pallas	2003–2011	2.7	-25.7	active	AMAP, 2014
PBDE99	Pallas	2003–2012	2.9	-23.9	active	Hung et al., 2016
PBDE99	Zeppelin	1993–2014	4.2	-16.5	active	Wöhrnschimmel et al., 2016
PBDE99	Zeppelin	2006–2012	2.6	-26.7	active	AMAP, 2014
PBDE99	Zeppelin	2006–2012	2.6	-26.7	active	Hung et al., 2016
PBDE100	Pallas	2003–2011	4.3	-16.1	active	AMAP, 2014
PBDE100	Pallas	2003–2012	4	-17.3	active	Hung et al., 2016
PBDE100	Zeppelin	2006–2012	4.1	-16.9	active	AMAP, 2014
PBDE100	Zeppelin	2006–2012	4.1	-16.9	active	Hung et al., 2016

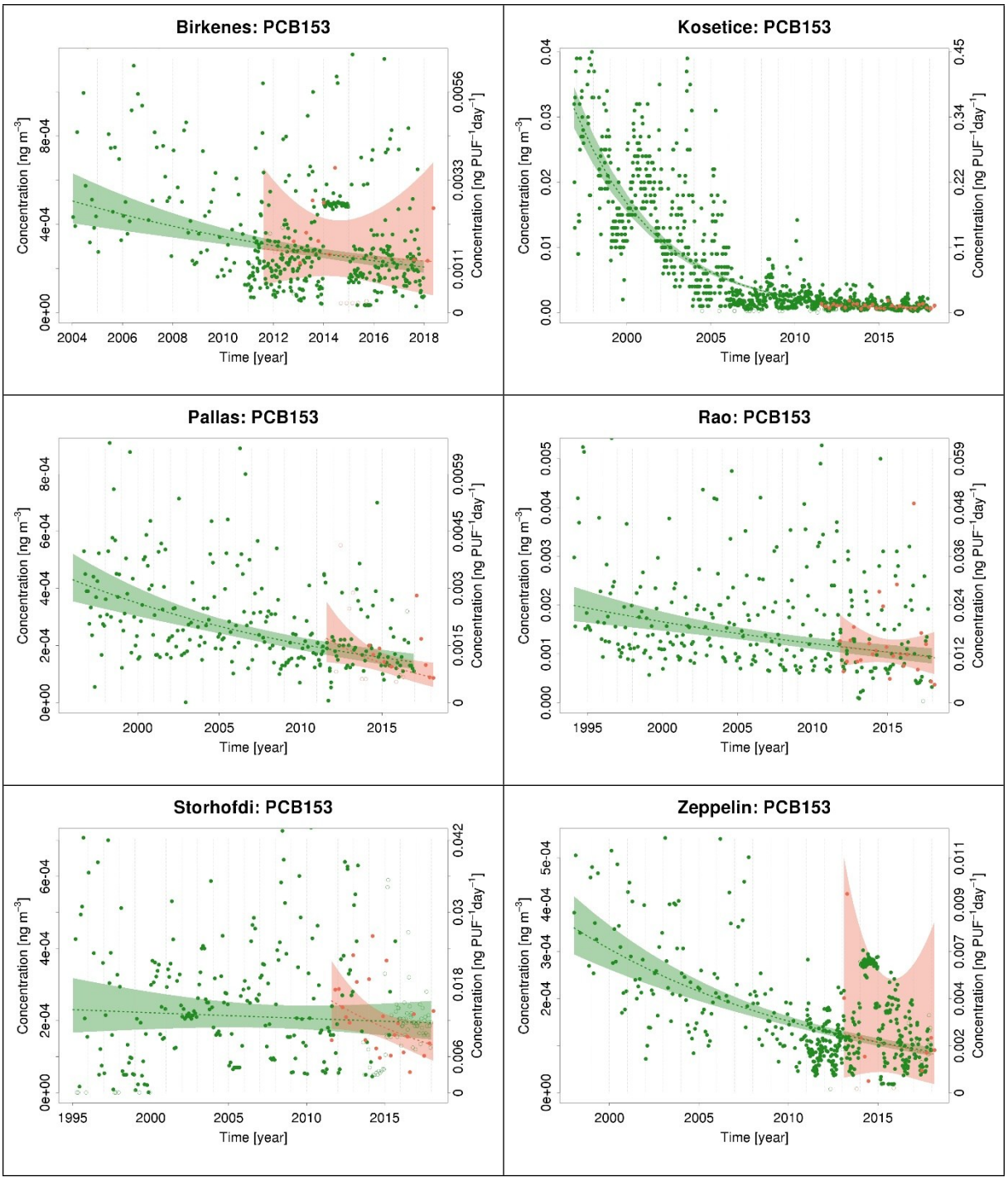
Figure S5 – Plots of exponential active and passive time trends at each site for all remaining compounds (those not included in the manuscript). Individual active (green) and passive (red) sampling measurements are drawn as points, with the calculated exponential trends and their 95% confidence interval shown as lines and shaded areas, respectively.




Birkenes: PCB52
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Kosetice: PCB52
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PCB52
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PCB52
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Storhofdi: PCB52
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PCB52
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]


Birkenes: PCB101
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Kosetice: PCB101
Pallas: PCB101
Rao: PCB101
Storhofdi: PCB101
Zeppelin: PCB101


Birkenes: PCB138
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Kosetice: PCB138
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PCB138
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PCB138
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Storhofdi: PCB138
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PCB138
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]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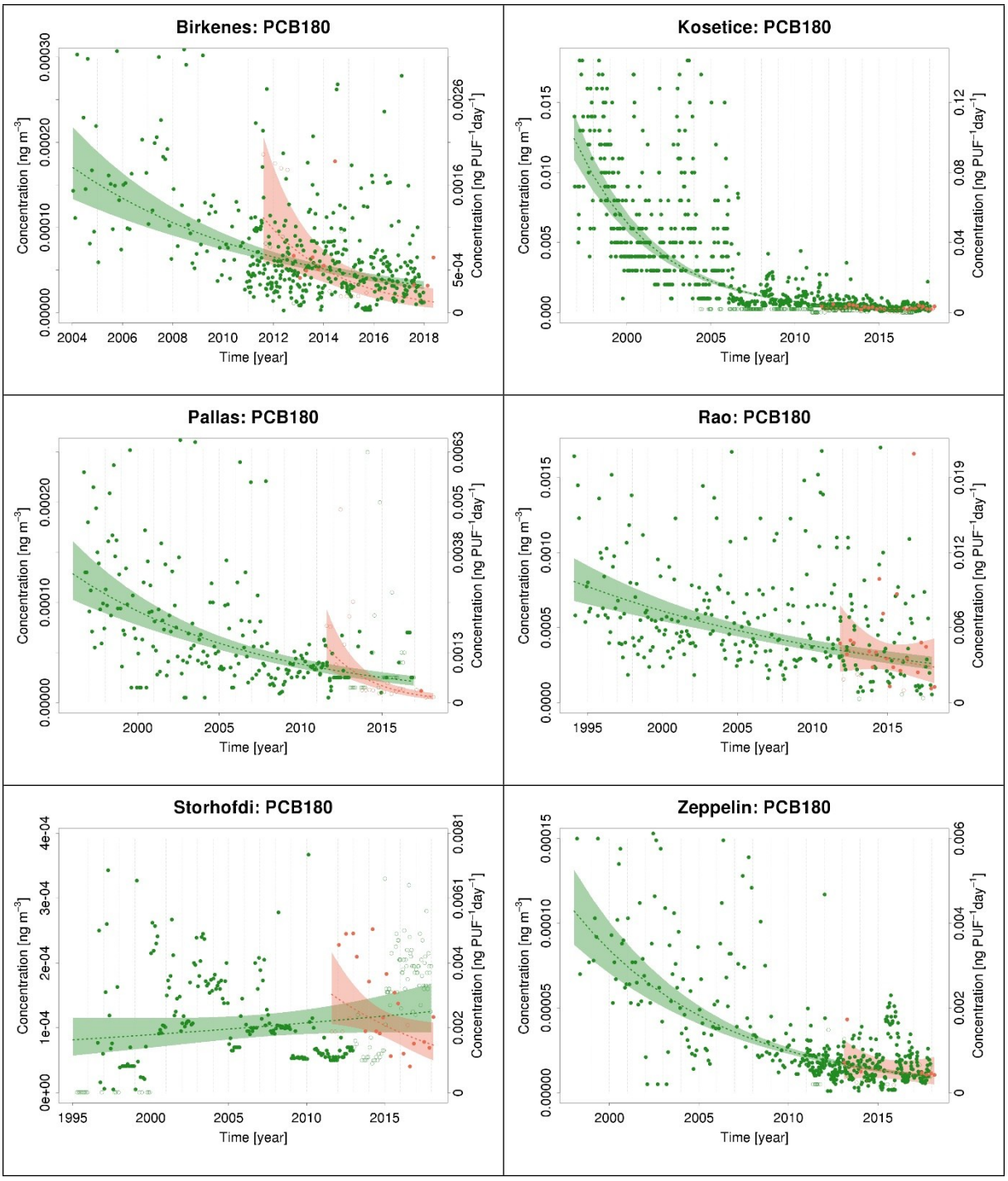
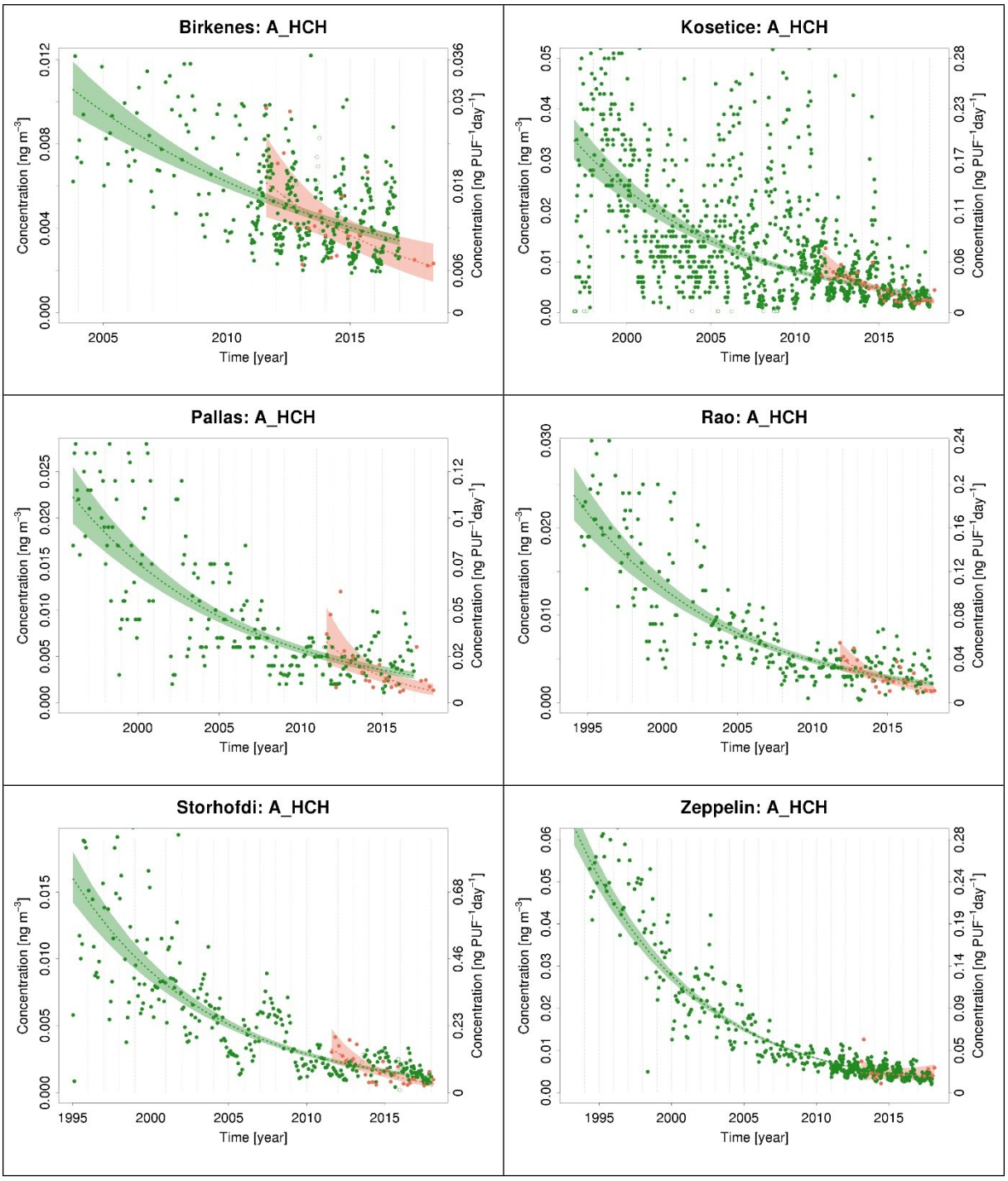
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Kosetice: PCB153
Pallas: PCB153
Rao: PCB153
Storhofdi: PCB153
Zeppelin: PCB153


Birkenes: PCB180
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Kosetice: PCB180
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PCB180
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PCB180
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Storhofdi: PCB180
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PCB180
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]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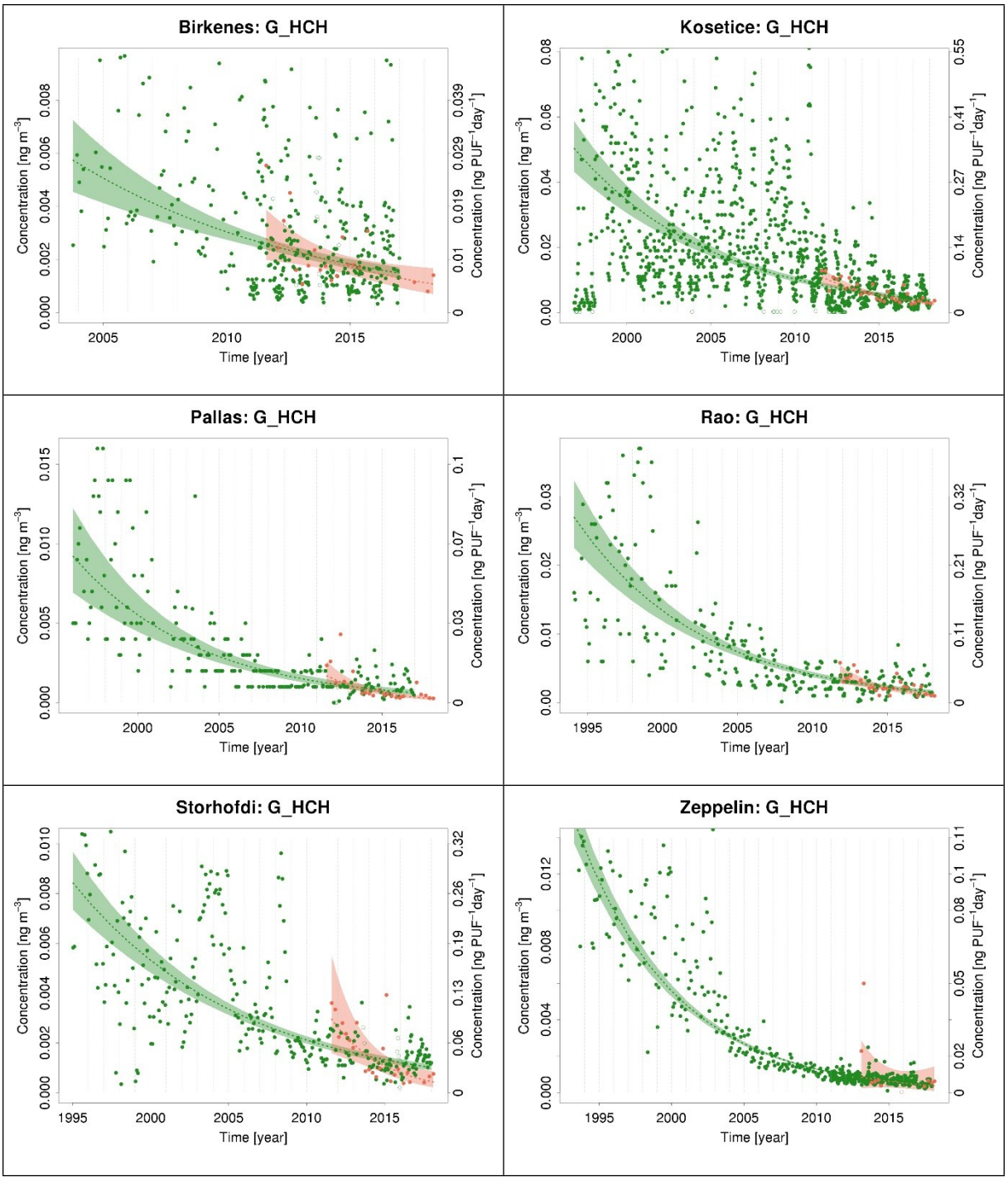
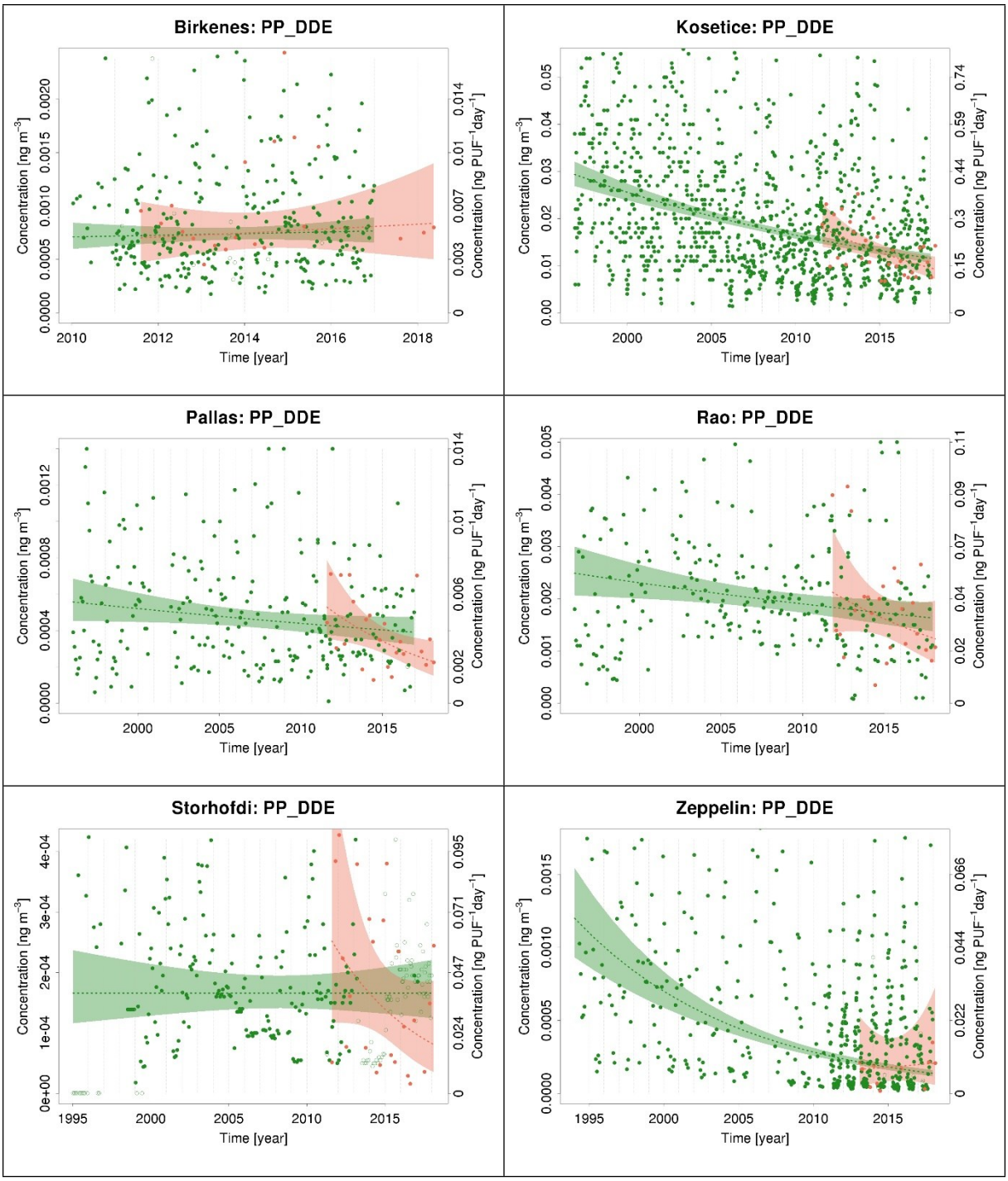
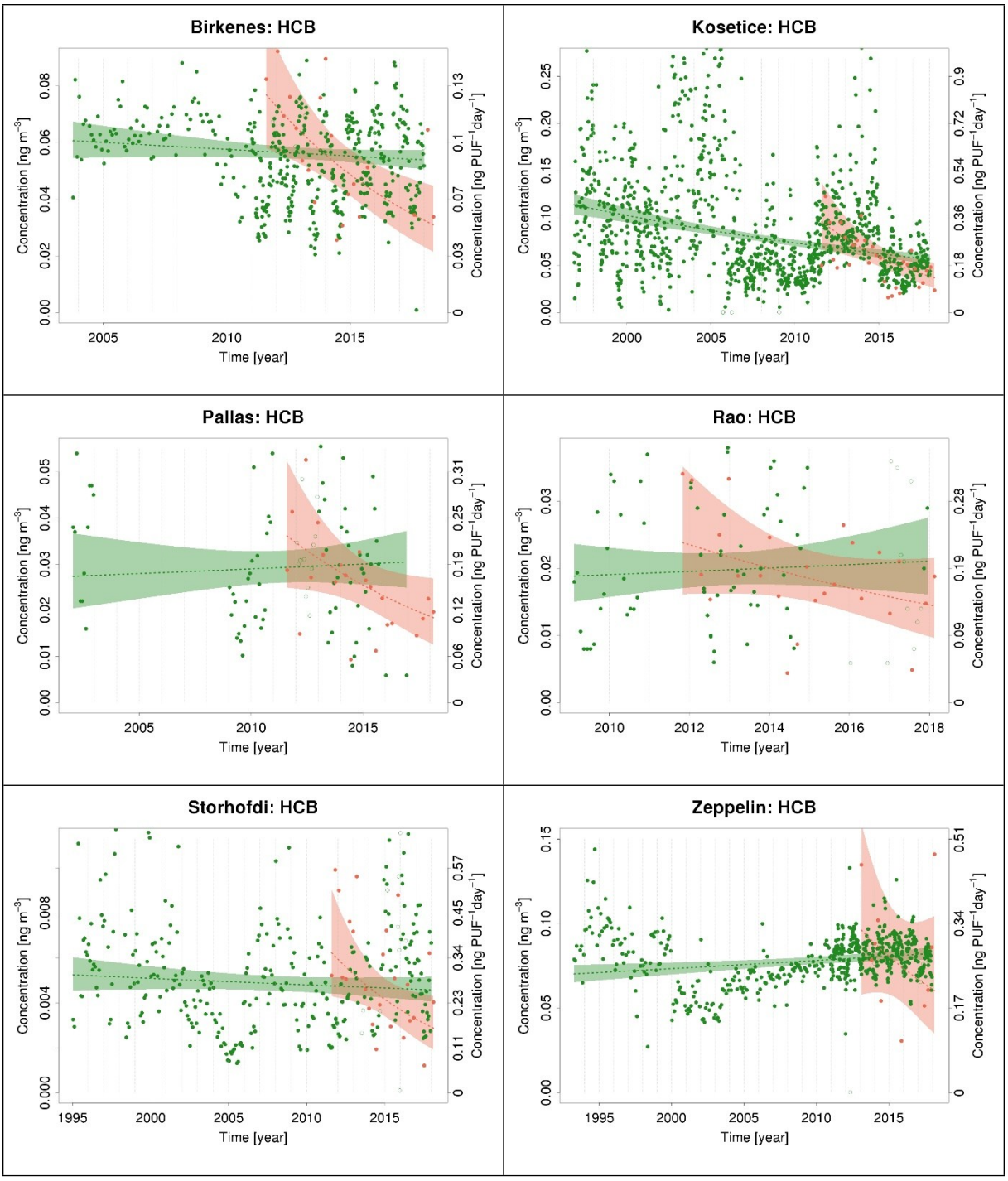
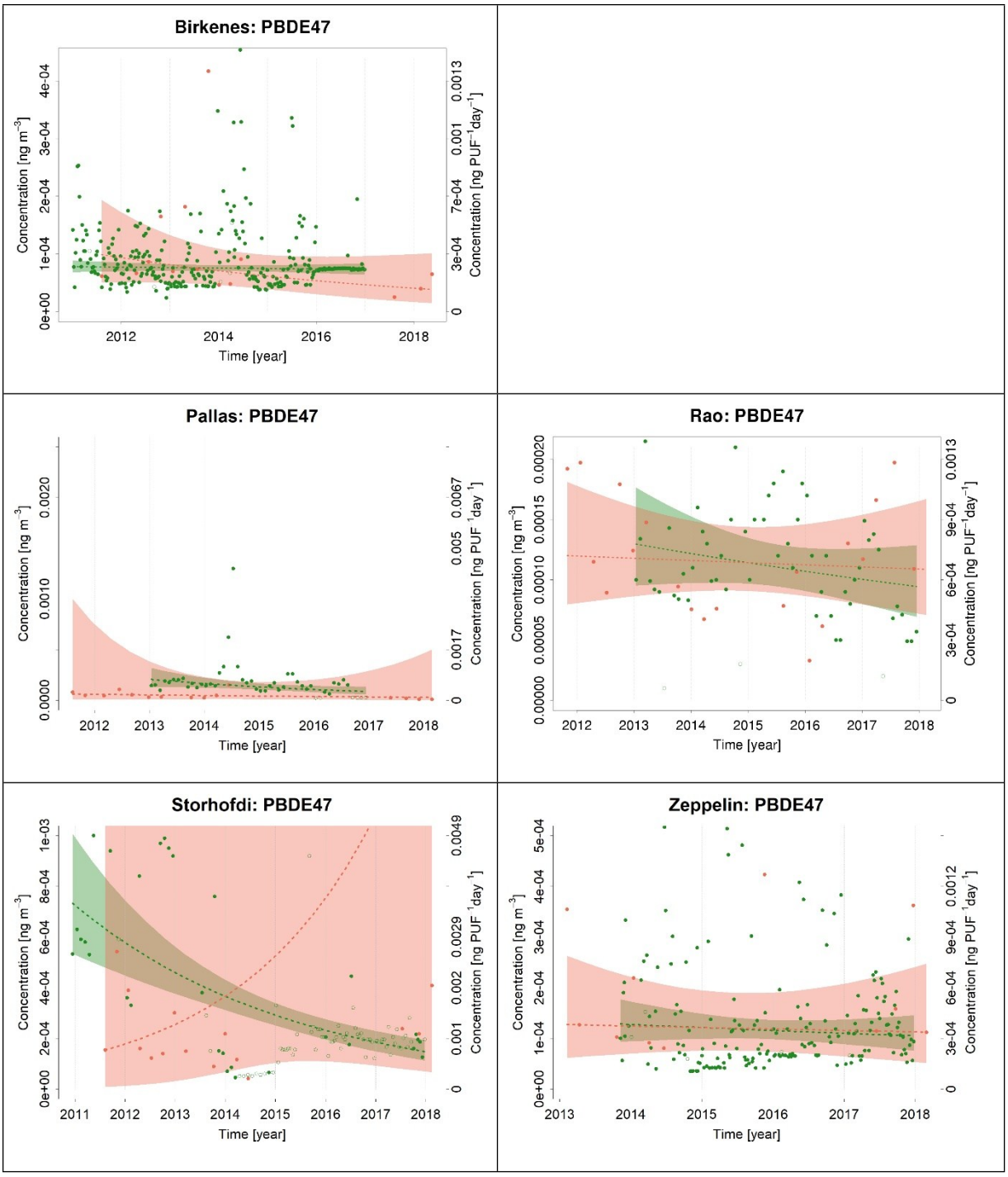
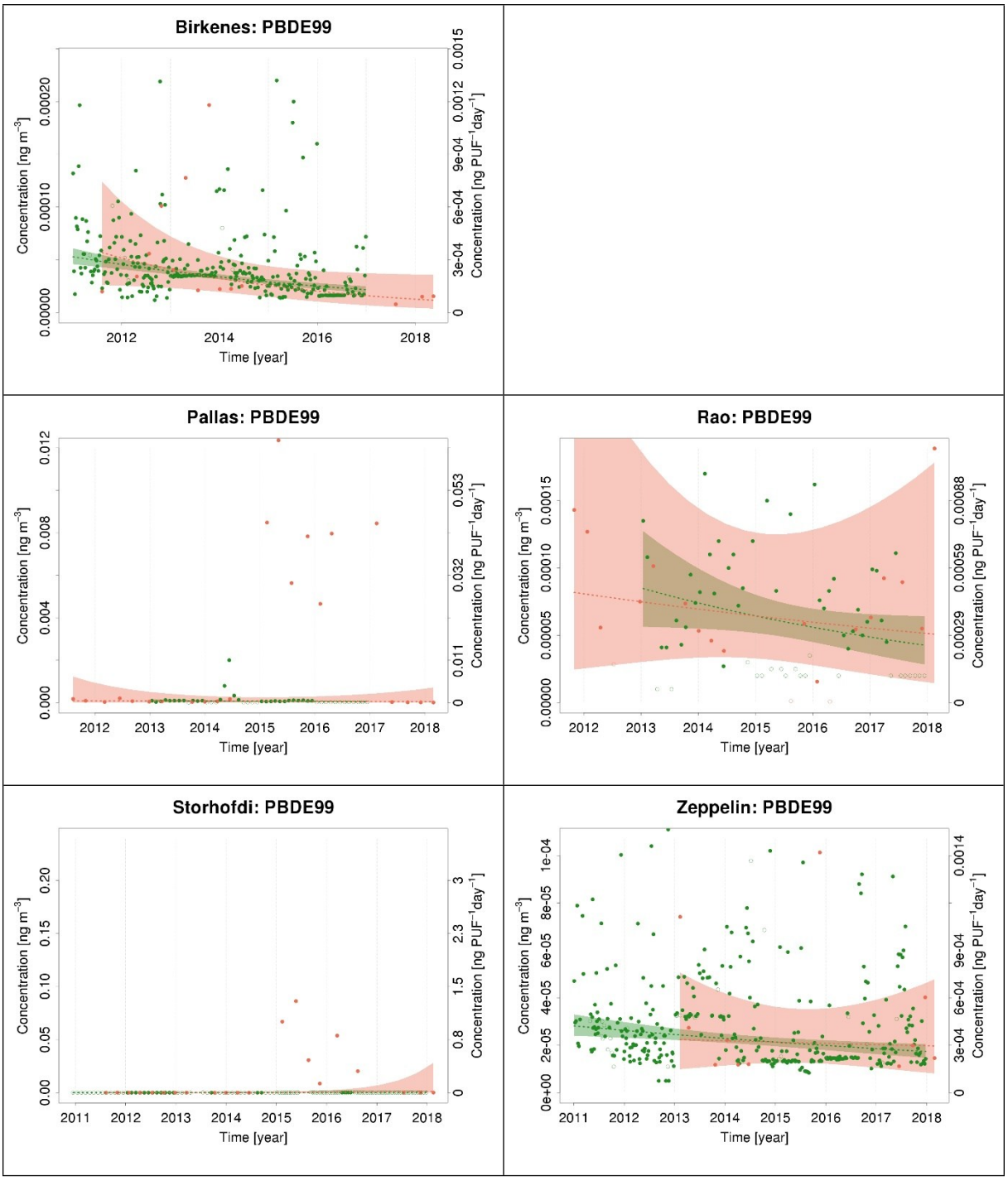
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Kosetice: A_HCH
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: A_HCH
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: A_HCH
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Storhofdi: A_HCH
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: A_HCH
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]

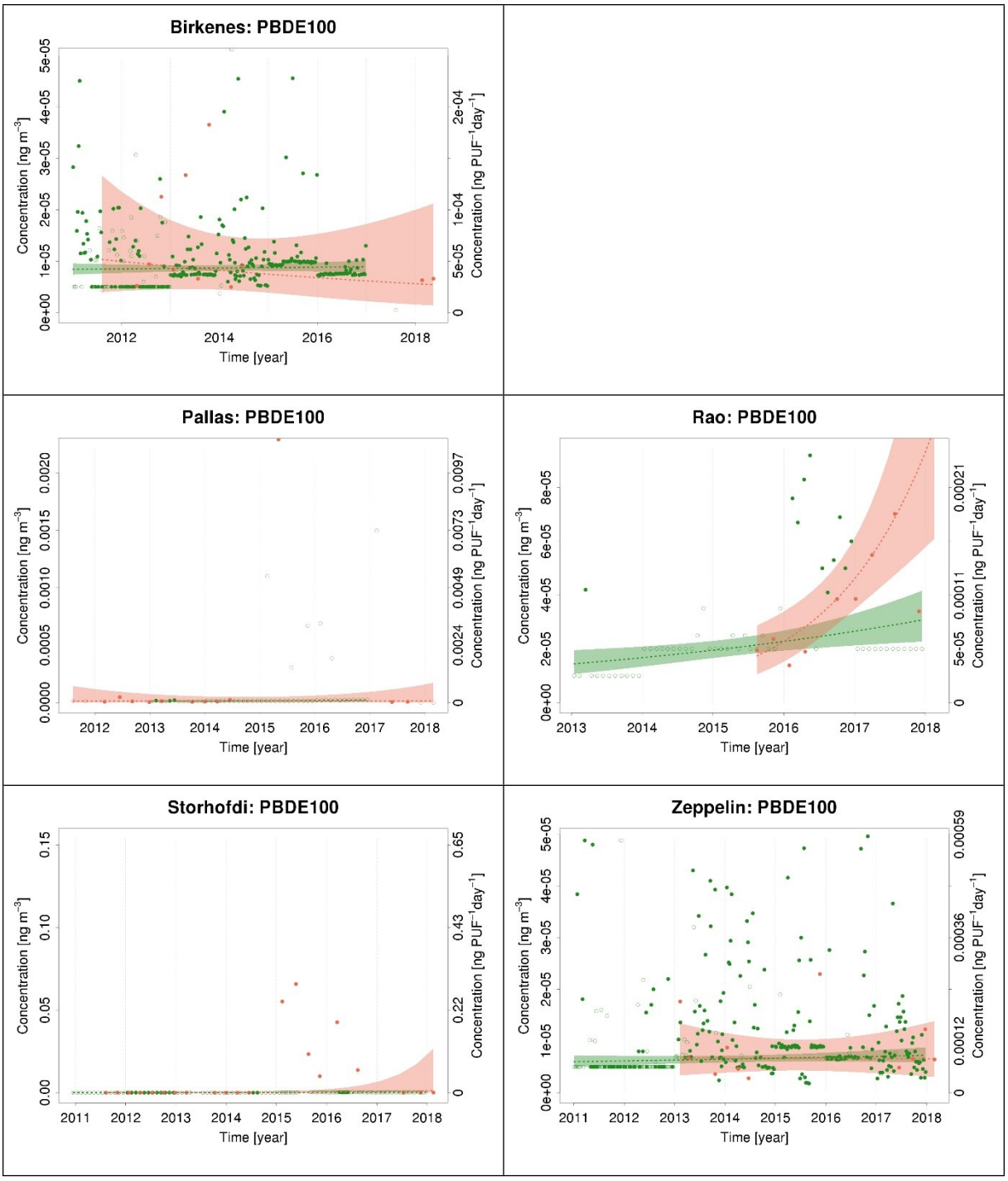

Birkenes: G_HCH
Kosetice: G_HCH
Pallas: G_HCH
Rao: G_HCH
Storhofdi: G_HCH
Zeppelin: G_HCH
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]

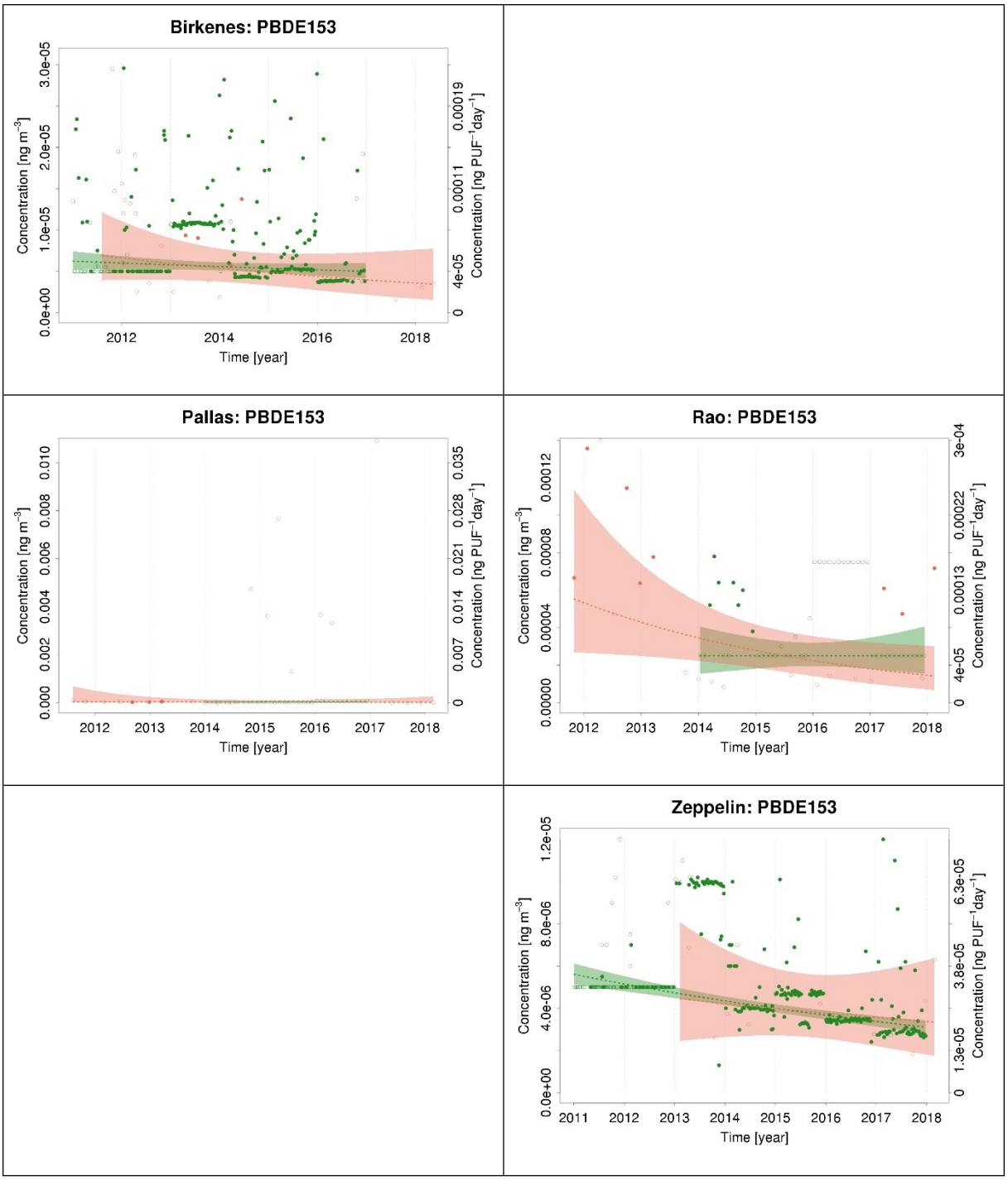

Birkenes: PP_DDE
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Kosetice: PP_DDE
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PP_DDE
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PP_DDE
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Storhofdi: PP_DDE
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PP_DDE
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]

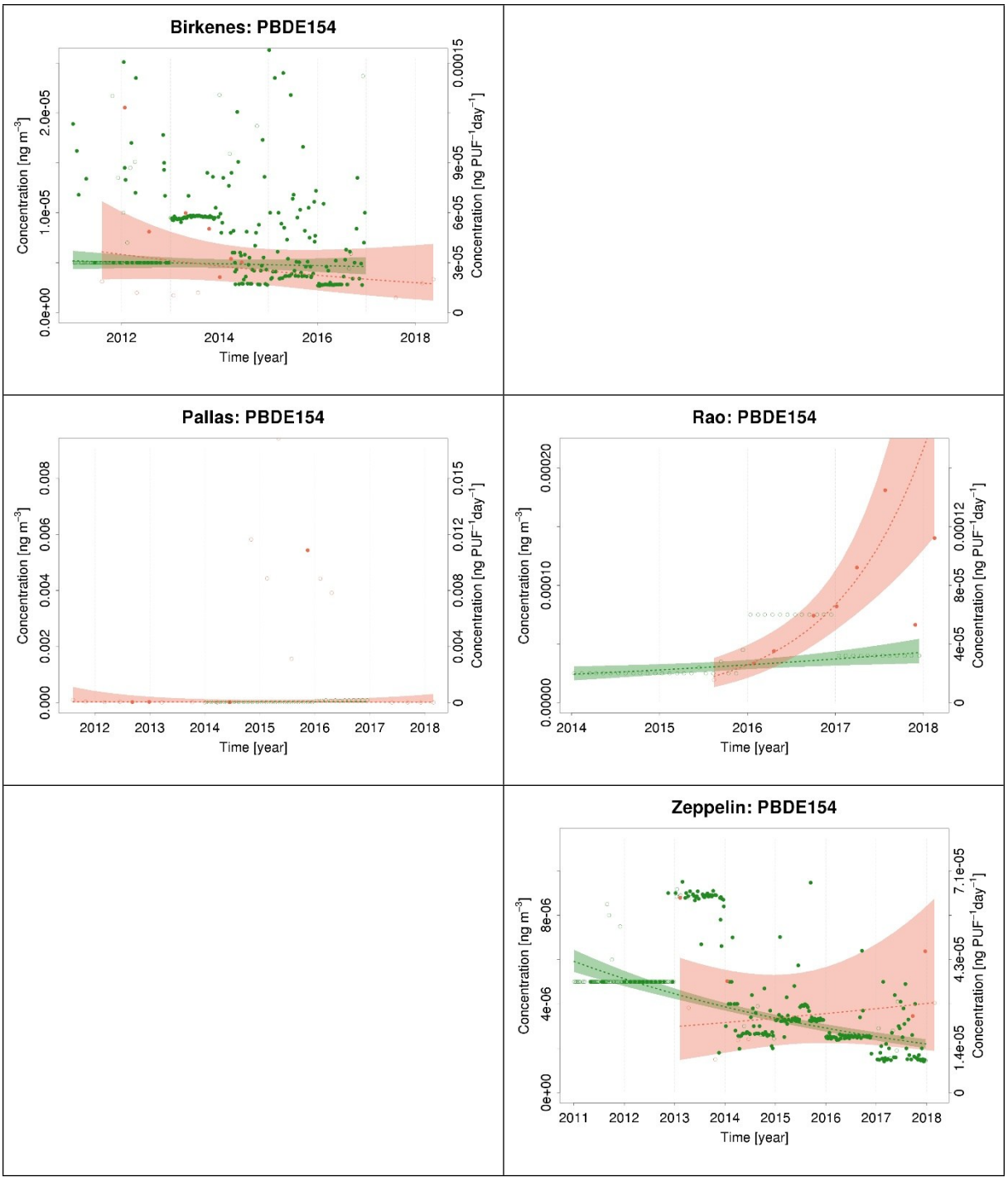

Birkenes: HCB
Kosetice: HCB
Pallas: HCB
Rao: HCB
Storhofdi: HCB
Zeppelin: HCB
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]

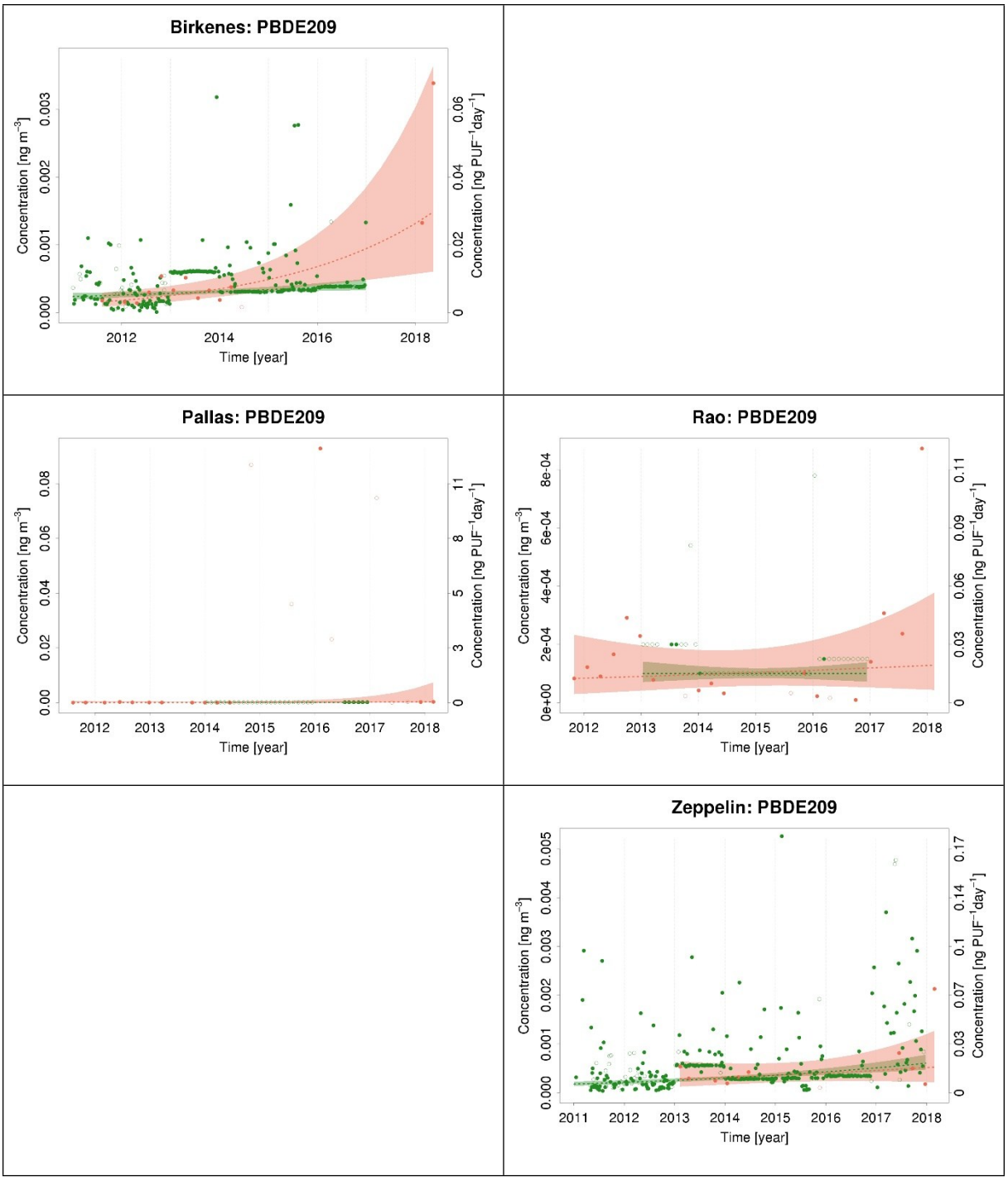

Birkenes: PBDE47
Concentration [ng m-3]
Concentration [ng PUF-1day-1]
Time [year]
Pallas: PBDE47
Concentration [ng m-3]
Concentration [ng PUF-1day-1]
Time [year]
Rao: PBDE47
Concentration [ng m-3]
Concentration [ng PUF-1day-1]
Time [year]
Storhofdi: PBDE47
Concentration [ng m-3]
Concentration [ng PUF-1day-1]
Time [year]
Zeppelin: PBDE47
Concentration [ng m-3]
Concentration [ng PUF-1day-1]
Time [year]


Birkenes: PBDE99
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PBDE99
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PBDE99
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Storhofdi: PBDE99
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PBDE99
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]


Birkenes: PBDE100
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PBDE100
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PBDE100
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Storhofdi: PBDE100
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PBDE100
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]


Birkenes: PBDE153
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PBDE153
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PBDE153
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PBDE153
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]


Birkenes: PBDE154
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PBDE154
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PBDE154
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PBDE154
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]


Birkenes: PBDE209
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Pallas: PBDE209
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Rao: PBDE209
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]
Zeppelin: PBDE209
Concentration [ng m^{-3}]
Concentration [ng PUF^{-1}day^{-1}]
Time [year]