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Supplement of

**Polycyclic aromatic hydrocarbons, polychlorinated biphenyls,
and chlorinated pesticides in background air in central Europe –
investigating parameters affecting wet scavenging of polycyclic
aromatic hydrocarbons**

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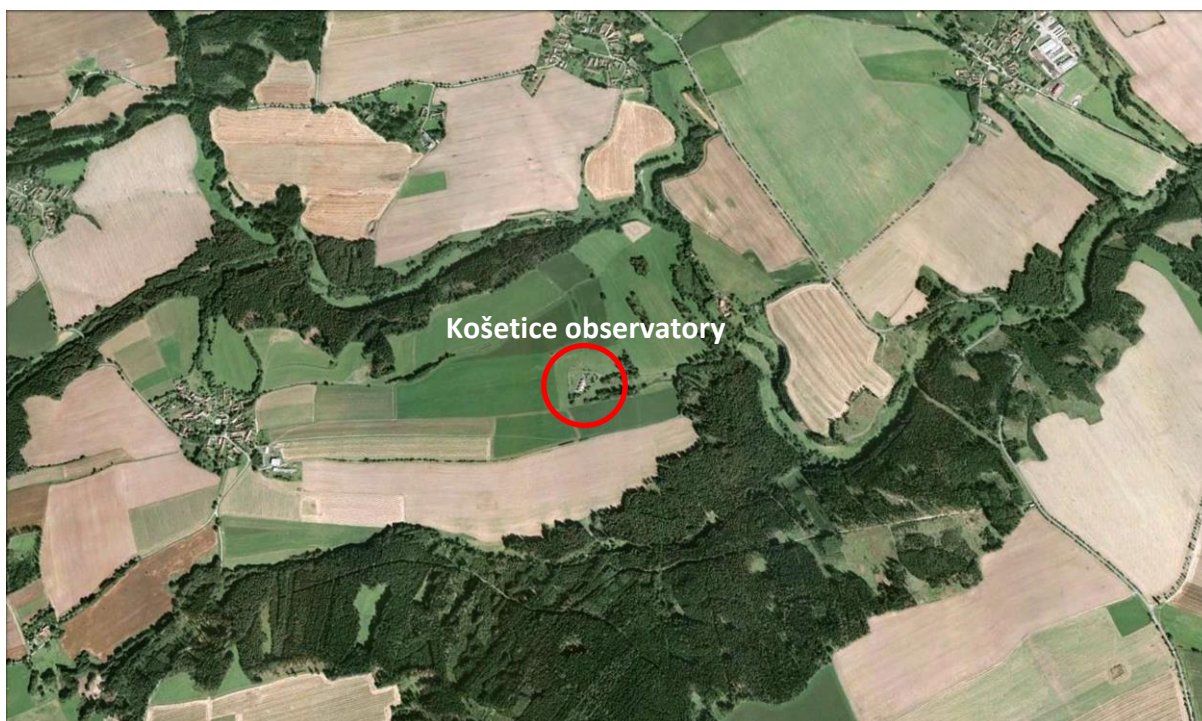


Figure S1. Aerial picture of Košetice observatory (49° 34' N, 15° 05' E, 534 m a.s.l.). The source of the picture is Google Earth 2014.

Table S1. Rainwater and air sample information

	Collection period	Number of samples (<i>n</i>)	^a <i>n</i> considered for calculating scavenging ratios	^b <i>n</i> used for calculating scavenging ratios
Rainwater	2 Dec 2011 – 14 Jan 2014	231	54	32
Air	7 Dec 2011 – 20 Nov 2013	162	54	32

^a air samples collected 6 – 35 hours prior to the onset of rain events; ^b frontal passage was not expected prior to the onset of rain.

Table S2. Sample information and cloud heights for 32 selected rain events

Air sampling start & stop date	^a Air sampling duration (h)	Precipitation duration (h)	^b Cloud heights (m)	
			Base	Top
6 – 7 Jan 12	24	18	780	3713
15 – 16 Jan 12	17	16	1230	2860
6 – 7 Feb 12	15	7	1500	2886
7 – 8 Mar 12	20	3	3000	3529
14 – 14 Apr 12	14	7	1170	4216
2 – 3 Jun 12	22	8	1500	6026
10 – 11 Aug 12	23	2	1290	4713
21 – 22 Sep 12	20	2	2400	5831
30 Sep – 1 Oct 12	15	2	1500	5830
3 – 4 Oct 12	19	4	1050	5299
11 – 12 Oct 12	22	6	300	5248
31 Oct – 1 Nov 12	19	3	2100	4963
27 – 28 Nov 12	28	4	60	4873
25 – 26 Dec 12	26	11	630	NA
4 – 4 Jan 13	6	16	780	4457
15 – 17 Jan 13	35	12	120	3007
28 – 29 Jan 13	23	21	900	4011
2 – 3 Feb 13	22	10	1200	2591
11 – 12 Feb 13	21	1	210	3064
18 – 19 Feb 13	19	24	780	3569
17 – 18 May 13	22	7	1800	5187
23 – 24 May 13	21	10	1110	3743
25 – 26 May 13	16	10	510	3998
31 May – 1 Jun 13	16	21	3900	5075
28 – 29 Jul 13	26	5	1110	6604
3 – 4 Aug 13	22	1	1500	6400
8 – 9 Aug 13	28	7	480	6084
24 – 25 Aug 13	23	18	900	5780
9 – 10 Oct 13	29	8	570	4952
14 – 15 Oct 13	22	8	2400	4470
2 – 3 Nov 13	15	1	3000	4901
19 – 20 Nov 13	16	3	180	4656

^a Duration of air sampling conducted prior to the onset of rain; ^b cloud base and top heights were estimated through ceilometer and radiosonde measurements, respectively; NA: not available.

Table S3. Meteorological parameters, aerosol and rainwater chemical properties, as well as PAH scavenging ratios for 32 selected rain events

Air sampling start & stop date	Precipitation		^b T (°C)	^c Concentrations ($\mu\text{g m}^{-3}$)					^d PM SA ($\mu\text{m}^2\text{cm}^{-3}$)	^e Rain ions (mg L^{-1})	Σ PAHs W_t
	Intensity (mm h^{-1})	^a Type		PM _{2.5}	PM ₁₀	PM Ions	PM EC	PM OC			
6–7 Jan 12	0.5	S/R	1.1	5.9	10.0	2.3	NA	NA	1.5×10^3	<MDL	1.7×10^4
15–16 Jan 12 *	0.3	S/H	-3.9	7.2	10.8	4.1	0.2	2.0	2.3×10^3	<MDL	1.5×10^4
6–7 Feb 12	0.1	S/H	-14.1	43.5	57.8	15	NA	NA	1.2×10^4	<MDL	3.5×10^3
7–8 Mar 12	0.2	S	-0.3	21.4	31.0	12.7	NA	NA	7.0×10^3	<MDL	1.3×10^4
14–14 Apr 12 *	0.1	R	7.1	17.5	26.9	15.6	0.6	3.2	6.3×10^3	8.4	2.3×10^5
2–3 Jun 12 *	1.1	R	16	7.4	14.6	6.6	0.1	1.5	3.5×10^3	2.4	2.3×10^4
10–11 Aug 12 *	0.8	R	15	7.3	12.0	6.3	0.2	1.8	2.8×10^3	2.0	7.4×10^3
21–22 Sep 12 *	0.4	R	9.2	8.8	14.7	6.5	0.3	2.3	3.1×10^3	3.5	9.1×10^3
30 Sep–1 Oct 12	0.4	R	11.5	11.8	15.7	9.0	NA	NA	4.0×10^3	5.0	5.3×10^3
3–4 Oct 12 *	0.7	R	12.5	7.8	10.0	4.7	0.2	2.3	2.7×10^3	<MDL	2.6×10^4
11–12 Oct 12 *	0.5	R	7	15.0	19.4	8.9	0.5	3.1	5.1×10^3	0.8	3.4×10^3
31 Oct–1 Nov 12	0.4	R	4.2	16.0	20.4	10.6	NA	NA	5.6×10^3	2.7	2.6×10^3
27–28 Nov 12 *	0.7	R/H	5.3	12.0	19.2	13.8	1.0	4.1	5.4×10^3	2.7	1.4×10^4
25–26 Dec 12	0.8	R	5.1	7.3	10.0	2.4	NA	NA	2.8×10^3	2.2	5.9×10^3
4–4 Jan 13	1.0	NA	7	4.6	8.4	3.3	NA	NA	NA	2.2	5.6×10^3
15–17 Jan 13 *	0.1	NA	-4.1	29.8	37.5	16.1	1.5	7.5	NA	<MDL	2.7×10^4
28–29 Jan 13	0.8	NA	2.6	9.5	15.6	6.3	NA	NA	NA	4.2	4.2×10^3
2–3 Feb 13 *	0.3	NA	-0.8	6.0	8.0	3.5	0.0	1.1	NA	0.8	1.3×10^4
11–12 Feb 13	5.6	NA	-4	30.2	33.2	15.2	NA	NA	NA	<MDL	2.4×10^3
18–19 Feb 13 *	0.2	NA	-1.5	34.8	40.2	15.2	0.4	3.1	NA	2.7	3.3×10^3
17–18 May 13	0.3	NA	15.9	13.2	26.9	6.0	NA	NA	NA	1.0	1.2×10^4
23–24 May 13	0.2	NA	8.1	10.9	15.8	11.4	NA	NA	NA	15.1	1.6×10^4
25–26 May 13 *	0.3	NA	6.2	11.4	13.1	5.8	0.1	0.6	NA	2.3	5.2×10^3
31 May–1 Jun 13 *	2.4	NA	10.4	5.4	7.0	3.0	0.1	1.6	NA	2.1	4.3×10^3

Table S3 continued

Air sampling start & stop date	Precipitation		^b T (°C)	^c Concentrations ($\mu\text{g m}^{-3}$)					^d PM SA ($\mu\text{m}^2\text{cm}^{-3}$)	^e Rain ions (mg L^{-1})	Σ PAHs W_t
	Intensity (mm h^{-1})	^a Type		PM _{2.5}	PM ₁₀	PM Ions	PM EC	PM OC			
28–29 Jul 13	6.9	NA	21.1	23.2	28.0	9.5	NA	NA	NA	2.1	5.8×10^3
3–4 Aug 13	28.5	NA	20.2	16.7	23.6	6.9	NA	NA	NA	2.5	5.4×10^3
8–9 Aug 13 *	1.5	NA	16	19.7	31.6	13.5	0.1	4.3	NA	4.8	8.9×10^3
24–25 Aug 13 *	1.6	NA	14.1	20.3	24.0	9.4	0.4	4.6	NA	1.4	1.1×10^4
9–10 Oct 13	0.6	NA	10.7	30.6	37.7	21.4	NA	NA	NA	8.9	1.1×10^4
14–15 Oct 13	0.7	NA	8.9	11.3	16.3	6.6	NA	NA	NA	2.3	8.7×10^3
2–3 Nov 13	0.2	NA	9.9	9.1	10.3	5.7	NA	NA	NA	<MDL	4.7×10^3
19–20 Nov 13	0.6	NA	3.6	28.8	33.4	17.6	NA	NA	NA	4.1	1.9×10^4

^a precipitation type, S: snow, R: rain, H: haze; ^b mean near-ground temperature measured during air sampling; ^c concentrations of particulate matter (PM) with diameter $<2.5 \mu\text{m}$ (PM_{2.5}) and $<10 \mu\text{m}$ (PM₁₀), PM ionic species (i.e. the sum of SO_4^{2-} , NO_3^- , NH_4^+), PM elemental carbon (EC) and organic carbon (OC); ^d mean PM surface area concentrations; ^e the sum of the concentrations of SO_4^{2-} , NO_3^- , and Cl^- in rainwater; asterisk in the first column indicate events for which correlation analysis with environmental parameters were performed; NA: not available.

Table S4. Analyte particulate mass fractions (θ)

Compound	θ				^a r^2
	Min	Max	Mean \pm SD	Median	
ACY	0.00	0.63	0.09 \pm 0.11	0.06	0.03 ($n = 141$)
ACE	0.00	0.12	0.03 \pm 0.03	0.02	0.25 ($n = 144$)
FLN	0.00	0.13	0.01 \pm 0.02	0.01	0.29 ($n = 156$)
ANT	0.01	0.56	0.19 \pm 0.13	0.16	0.35 ($n = 126$)
PHE	0.00	0.53	0.05 \pm 0.08	0.03	0.53 ($n = 154$)
RET	0.01	0.99	0.31 \pm 0.26	0.22	0.75 ($n = 156$)
PYR	0.01	0.96	0.40 \pm 0.22	0.36	0.72 ($n = 150$)
FLT	0.00	0.92	0.28 \pm 0.19	0.24	0.69 ($n = 155$)
BAA	0.05	1.00	0.90 \pm 0.17	0.96	0.26 ($n = 142$)
CHR	0.08	1.00	0.74 \pm 0.21	0.80	0.52 ($n = 162$)
BFN	0.02	1.00	0.59 \pm 0.25	0.61	0.59 ($n = 131$)
BGF	0.04	1.00	0.65 \pm 0.24	0.70	0.69 ($n = 151$)
PER	0.70	1.00	0.94 \pm 0.06	0.96	0.37 ($n = 97$)
CPP	0.74	1.00	0.96 \pm 0.06	0.99	0.39 ($n = 116$)
BBF	0.69	1.00	0.97 \pm 0.06	0.99	0.39 ($n = 152$)
BJF	0.74	1.00	0.95 \pm 0.06	0.99	0.43 ($n = 145$)
TPH	0.09	1.00	0.69 \pm 0.24	0.75	0.73 ($n = 162$)
BKF	0.73	1.00	0.95 \pm 0.07	0.98	0.52 ($n = 140$)
BAP	0.74	1.00	0.96 \pm 0.05	0.99	0.47 ($n = 132$)
DCA	0.72	1.00	0.92 \pm 0.07	0.94	0.34 ($n = 87$)
BEP	0.77	1.00	0.96 \pm 0.05	0.98	0.40 ($n = 141$)
IPY	0.74	1.00	0.97 \pm 0.05	0.99	0.37 ($n = 131$)
DHA	0.74	1.00	0.93 \pm 0.06	0.95	0.33 ($n = 88$)
BPE	0.81	1.00	0.97 \pm 0.05	0.99	0.46 ($n = 146$)
COR	0.67	1.00	0.95 \pm 0.07	0.98	0.16 ($n = 102$)
BNT	0.66	1.00	0.87 \pm 0.09	0.87	0.42 ($n = 75$)
ATT	0.69	1.00	0.91 \pm 0.08	0.94	0.24 ($n = 76$)
PeCB	0.00	0.15	0.02 \pm 0.03	0.01	
HCB	0.00	0.03	0.00 \pm 0.00	0.00	
PCB 28	0.01	0.17	0.06 \pm 0.04	0.04	
PCB 52	0.01	0.21	0.09 \pm 0.04	0.08	
<i>a</i> -HCH	0.00	0.11	0.03 \pm 0.02	0.03	
γ -HCH	0.01	0.23	0.06 \pm 0.04	0.05	
PCB 101	0.02	0.32	0.16 \pm 0.07	0.16	
<i>o,p'</i> -DDE	0.06	0.32	0.18 \pm 0.07	0.18	
<i>o,p'</i> -DDT	0.03	0.33	0.12 \pm 0.07	0.11	
PCB 138	0.05	0.31	0.18 \pm 0.07	0.19	
<i>p,p'</i> -DDE	0.00	0.52	0.04 \pm 0.08	0.02	0.39 ($n = 160$)
PCB 153	0.02	0.33	0.16 \pm 0.08	0.15	
<i>p,p'</i> -DDT	0.03	0.33	0.14 \pm 0.08	0.11	
PCB 118	0.04	0.33	0.18 \pm 0.09	0.19	
PCB 180	0.07	0.33	0.23 \pm 0.07	0.25	
<i>p,p'</i> -DDD	0.10	0.32	0.20 \pm 0.07	0.19	

SD: standard deviation; ^a coefficient of regression between near-ground temperature and Θ . Note that in order to enhance comparison, the analytes are sorted in increasing order according to their octanol-air partitioning coefficients. Θ values are only reported for chlorinated compounds with DF >10% in gas or particulate phase.

Table S5. PAH scavenged mass fractions (ε)

	^a Upper-bound ε			^b Lower-bound ε			^c Median
	Min.	Max.	Median	Min.	Max.	Median	
ACY	0	2.2	1.4×10^{-2}	0	1.7	2.3×10^{-3}	5.3×10^{-4}
ACE	0	0.3	8.9×10^{-3}	0	8.1×10^{-2}	1.8×10^{-3}	5.2×10^{-4}
FLN	5.4×10^{-4}	1.5	1.5×10^{-2}	4.3×10^{-4}	0.3	3.6×10^{-3}	3.7×10^{-3}
ANT	0	0.4	1.3×10^{-2}	0	0.1	2.4×10^{-3}	9.4×10^{-3}
PHE	1.3×10^{-3}	0.3	2.6×10^{-2}	5.4×10^{-4}	5.1×10^{-2}	5.5×10^{-3}	1.5×10^{-2}
RET	0	0.4	1.2×10^{-2}	0	0.1	2.8×10^{-3}	NA
PYR	0	1.1	5.2×10^{-2}	0	0.2	1.0×10^{-2}	7.5×10^{-3}
FLT	2.8×10^{-3}	0.9	8.1×10^{-2}	1.1×10^{-3}	0.2	1.3×10^{-2}	9.9×10^{-3}
BAA	0	0.9	1.2×10^{-2}	0	5.7×10^{-2}	1.9×10^{-3}	4.8×10^{-2}
CHR	0	1.8	5.2×10^{-2}	0	0.1	1.2×10^{-2}	1.0×10^{-2}
BFN	0	1.2	3.0×10^{-2}	0	0.2	4.1×10^{-3}	NA
BGF	0	2.4	6.1×10^{-2}	0	0.1	9.1×10^{-3}	NA
PER	0	1.9	0	0	2.7×10^{-2}	0	NA
CPP	0	0.5	3.6×10^{-3}	0	6.8×10^{-2}	4.6×10^{-4}	NA
BBF	0	3.0	2.4×10^{-2}	0	5.0×10^{-2}	3.6×10^{-3}	NA
BJF	0	3.6	2.7×10^{-2}	0	0.2	1.6×10^{-3}	NA
TPH	0	3.2	7.4×10^{-2}	0	0.3	1.4×10^{-2}	NA
BKF	0	2.0	2.9×10^{-2}	0	6.8×10^{-2}	1.9×10^{-3}	NA
BAP	0	1.5	4.1×10^{-3}	0	3.9×10^{-2}	5.2×10^{-4}	NA
DCA	0	2.9	0	0	0.1	0	NA
BEP	0	2.9	4.0×10^{-2}	0	7.5×10^{-2}	6.5×10^{-3}	NA
IPY	0	3.5	1.7×10^{-2}	0	0.3	1.8×10^{-3}	NA
DHA	0	3.1	0	0	3.8×10^{-2}	0	NA
BPE	0	3.6	2.3×10^{-2}	0	0.2	2.5×10^{-3}	NA
COR	0	4.8	2.2×10^{-3}	0	0.2	7.2×10^{-4}	NA
BNT	0	2.8	0	0	0.2	0	NA
ATT	0	3.8	0	0	4.7×10^{-2}	0	NA
Σ PAHs	1.2×10^{-3}	0.8	5.3×10^{-2}	5.2×10^{-4}	0.1	9.7×10^{-3}	NA

^a Upper- and ^b lower-bound ε correspond to cloud base and top height, respectively; ^c median ε values reported by Skrdlíková et al., (2011) for could top heights ($n = 10$); zero value indicates an event in which analyte concentration in rainwater was <LOQ; NA: not available. Analytes are sorted in increasing order based on their octanol-air partition coefficients.