Supplement of

Snow scavenging and phase partitioning of nitrated and oxygenated aromatic hydrocarbons in polluted and remote environments in central Europe and the European Arctic

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Table S1. Overview of concentrations of NMAH sub-classes (ng L^{-1}) in precipitation

	NPs[A]	NCs[B]	NSAs	Total NMAHs	
Urban, western central Europe (Switzerland), rain	1659.8-3041.4^{TOT}	N.A.	N.A.	1659.8-3041.4^{TOT}	Leuenberger et al., 1988
Urban, central Europe (Germany), rain	<300-20180^{TOT} (4-NP+2-NP)	N.A.	N.A.	<300-20180^{TOT}	Levsen et al., 1991
Remote, central Europe (Germany), rain	6300-25700^{TOT} (4-NP, 3-M-4-NP, DNPs)	N.A.	N.A.	6300-25700^{TOT}	Herterich and Herrmann, 1990
Urban, rural and forest, central Europe (Germany), rain	> 1000*** (4-NP), 200-800*** (other NPs)	N.A.	N.A.	N.A.	Schüssler and Nitschke, 2001
Urban background, northern Europe (Denmark), rain	1000-11900^{TOT} (4NP)	N.A.	N.A.	3000-18000^{TOT} (NPs + DNOC)	Bossi et al., 2002
Urban background and rural, northern Europe (Denmark), rain	Rural: 4421$^{TOT\ ***}$, Urban: 7197$^{TOT\ ***}$ (NPs, DNPs)	N.A.	N.A.	Rural: 4421^{TOT***}, Urban: 7197^{TOT***}	Asman et al., 2005
Urban and rural, western Europe (France), rain	Urban: 94300-102700^{TOT}, Rural:71200-76800^{TOT}	N.A.	N.A.	Urban: 94300-102700^{TOT} Rural:71200-76800^{TOT}	Schummer at al., 2009
Urban, southern USA (Texas), rain	200-4890 (4-NP), 0-690 (2-NP), 40-3310 (2,4-DNP), 0-2260 (3-M-4-NP)	N.A.	N.A.	Average total: 3190	Ganranoo et al., 2010
Urban, central Europe (Germany)	2660 – 4400^{TOT}	N.A.	N.A.	2660 – 4400^{TOT}	Alber et al., 1989
Antarctica	5-15^{D} (4-NP)	N.A.	N.A.	5-15^{D} (4-NP)	Vanni et al., 2001
Urban, central Europe (Germany, Austria, Czech Republic) 2015-17	19.5-206.2^{P} 295.0-2969.2^{D}	<0.56**-54.1^{P} 19.5-894.9^{D}	<0.56**-16.4^{P} 13.0-388.4^{D}	19.5-276.6^{P} 291.5-4252.6^{D}	This study
Rural, central Europe(Germany, Austria, Czech Republic) 2015-16	4.6-51.5^{P} 553.0-1264.5^{D}	<0.5**-1.7^{P} 19.3-160.4^{D}	<0.5*-<0.9^{P**} 7.7-383.2^{D}	4.6-51.5^{P} 580.9-1345.3^{D}	This study
Remote, European Arctic 2015	3.5^{P} 361.5^{D}	<0.67^{P*} 3.5^{D}	<1.67^{P**} 22.0^{D}	3.5^{P} 387.0^{D}	This study

Table S2. Abraham solute descriptors

Substance	*E*	*S*	*A*	*B*	*V*	*L*	Reference
1,4-Naphthoquinone	1.08	1.13	0	0.62	1.1598	6.341	ABSOLV-EM
9-Fluorenone	1.75	1.51	0	0.36	1.3722	7.568	M. Abraham
Acenaphthenequinone	2.11	1.68	0	0.72	1.29	7.794	M. Abraham
9,10-Anthraquinone	1.41	1.7	0	0.46	1.5288	8.593	ABSOLV-EM
Benzanthrone	2.58	1.56	0	0.49	1.7412	9.798	M. Abraham
2-Nitro-9-fluorenone	2.13	2.01	0	0.48	1.5464	9.127	M. Abraham
Benzo(a)fluorenone	2.58	1.82	0	0.44	1.7412	10.063	M. Abraham
Benzo(b)fluorenone	2.37	1.75	0	0.67	1.7412	9.755	M. Abraham
1,2-Benzanthraquinone	2.5	2.14	0	0.6	1.8978	10.97	M. Abraham
1-Nitronaphthalene	1.6	1.51	0	0.29	1.2596	7.056	M. Abraham
2-Nitronaphthalene	1.6	1.5	0	0.34	1.2596	6.85	ABSOLV-EM
5-Nitroacenaphthene	1.86	1.56	0	0.33	1.4328	7.858	M. Abraham
2-Nitrofluorene	1.85	1.7	0	0.37	1.5307	8.364	M. Abraham
9-Nitroanthracene	2.57	1.88	0	0.38	1.6286	9.712	M. Abraham
9-Nitrophenanthrene	2.57	1.88	0	0.38	1.6286	9.712	M. Abraham
3-Nitrophenanthrene	2.57	1.88	0	0.38	1.6286	9.712	M. Abraham
2-nitrofluoranthene	2.87	2.09	0	0.35	1.7588	10.592	ABSOLV-CAL
3-nitrofluoranthene	2.87	2.09	0	0.35	1.7588	10.592	ABSOLV-CAL
1-Nitropyrene	3.07	2.15	0	0.41	1.7588	11.036	M. Abraham
2-Nitropyrene	3.07	2.15	0	0.41	1.7588	11.036	M. Abraham
7-Nitrobenz(a)anthracene	3.25	2.27	0	0.47	1.9976	12.21	M. Abraham
6-Nitrochrysene	3.29	2.25	0	0.48	1.9976	12.23	M. Abraham
1,3-Dinitropyrene	3.34	2.8	0	0.52	1.933	12.58	M. Abraham
1,6-Dinitropyrene	3.34	2.8	0	0.52	1.933	12.58	M. Abraham
1,8-Dinitropyrene	3.34	2.8	0	0.52	1.933	12.58	M. Abraham
6-Nitrobenzo(a)pyrene	3.91	2.45	0	0.48	2.1278	13.62	M. Abraham
2,4-Dinitrophenol	1.2	1.49	0.09	0.56	1.1235	5.981	ABSOLV-EM
Dinitro-ortho-cresol	1.2	1.59	0.04	0.52	1.2644	6.58	ABSOLV-EM
3-Nitrosalicylic acid	1.08	1.44	0.39	0.36	1.1646	6.253	ABSOLV-CAL
5-Nitrosalicylic acid	1.33	1.23	0.82	0.47	1.1646	6.742	ABSOLV-EM
4-Nitrophenol	1.07	1.72	0.82	0.26	0.9493	5.876	ABSOLV-EM
4-nitroguaiacol	1.04	1.53	0.45	0.57	1.1489	6.029	ABSOLV-CAL
3-Methyl-4-nitrophenol	1.07	1.63	0.8	0.25	1.0902	6.06	ABSOLV-EM
2-Methyl-4-nitrophenol	1.1	1.6	0.78	0.25	1.0902	6.324	ABSOLV-EM
4-Nitrocatechol	1.2	1.65	1.14	0.63	1.008	6.002	ABSOLV-CAL
4-Methyl-5-nitrocatechol	1.22	1.59	1.14	0.63	1.1489	6.474	ABSOLV-CAL
3-Methyl-5-nitrocatechol	1.22	1.59	1.14	0.63	1.1489	6.474	ABSOLV-CAL

Abraham solute descriptors used in multiphase ppLFER gas-particle partitioning model. Values obtained from various sources - ABSOLV-EM (Exact Match): experimental values present in ABSOLV database; ABSOLV-CAL: values estimated by the program using substance molecular structure (ACD/Labs, 2015). The remaining values were obtained through personal communication with Michael H. Abraham, University College London.

Table S3. ppLFER system parameters

System	Unit	*e*	*s*	*a*	*b*	*v*	*l*	*c*	T(K)
DMSO-air [a]	$L_{air}\ L^{-1}_{solvent}$	-0.22	2.90	5.04	0.00	-	0.72	-0.56	298
PU-air [b]	$L_{air}\ kg^{-1}_{PU}$	-	1.69	3.66	0.00	0.36	0.71	-0.15	288
Soot-air [c]	$m^3_{air}\ m^{-2}_{surface}$	-	-	2.70	2.45	-	1.09	-8.47	288
$(NH_4)_2SO_4$ (60% RH) [d]	$m^3_{air}\ m^{-2}_{surface}$	-	-	2.13	5.34	-	0.88	-8.47	288
NaCl (60% RH) [d]	$m^3_{air}\ m^{-2}_{surface}$	-	-	2.86	4.82	-	0.84	-8.47	288
Octanol-water [e]	$L_{water}\ L^{-1}_{octanol}$	-	-1.41	-0.18	-3.45	2.41	0.43	0.34	298

[a] Abraham et al., (2010), [b] Kamprad and Goss, (2007), [c] Roth et al., (2005), [d] Goss et al., (2003); [e] Goss, (2005)

Figure S1. Snowmelt sample separation and extraction assembly

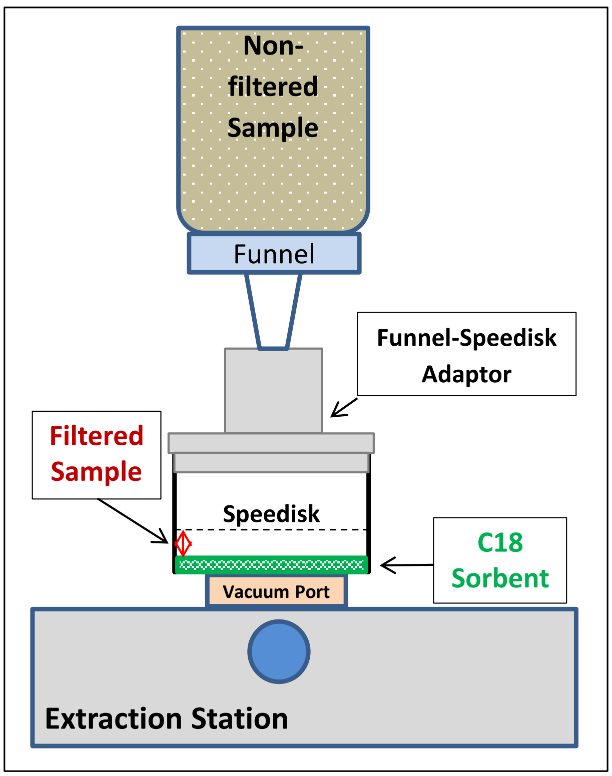
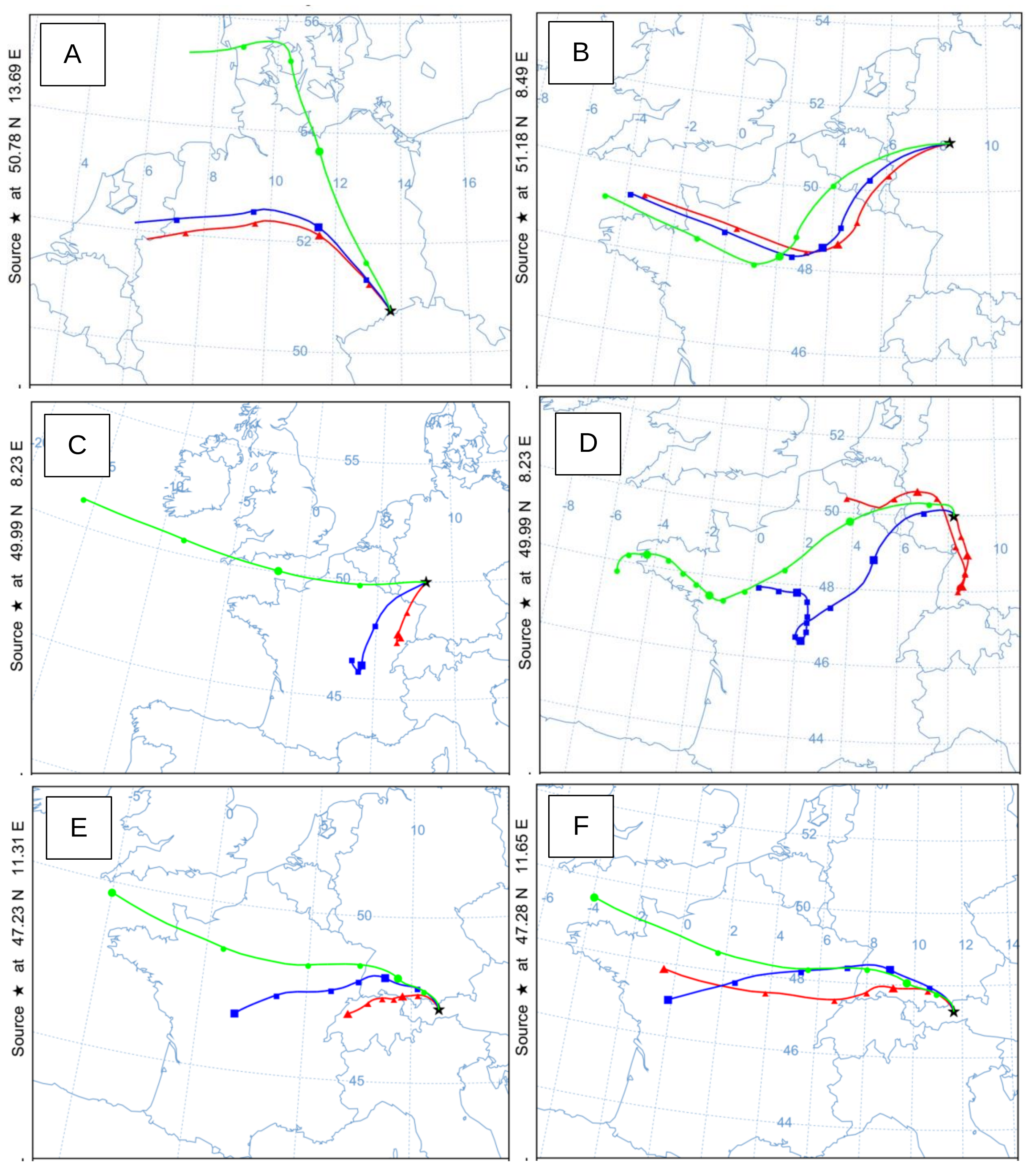
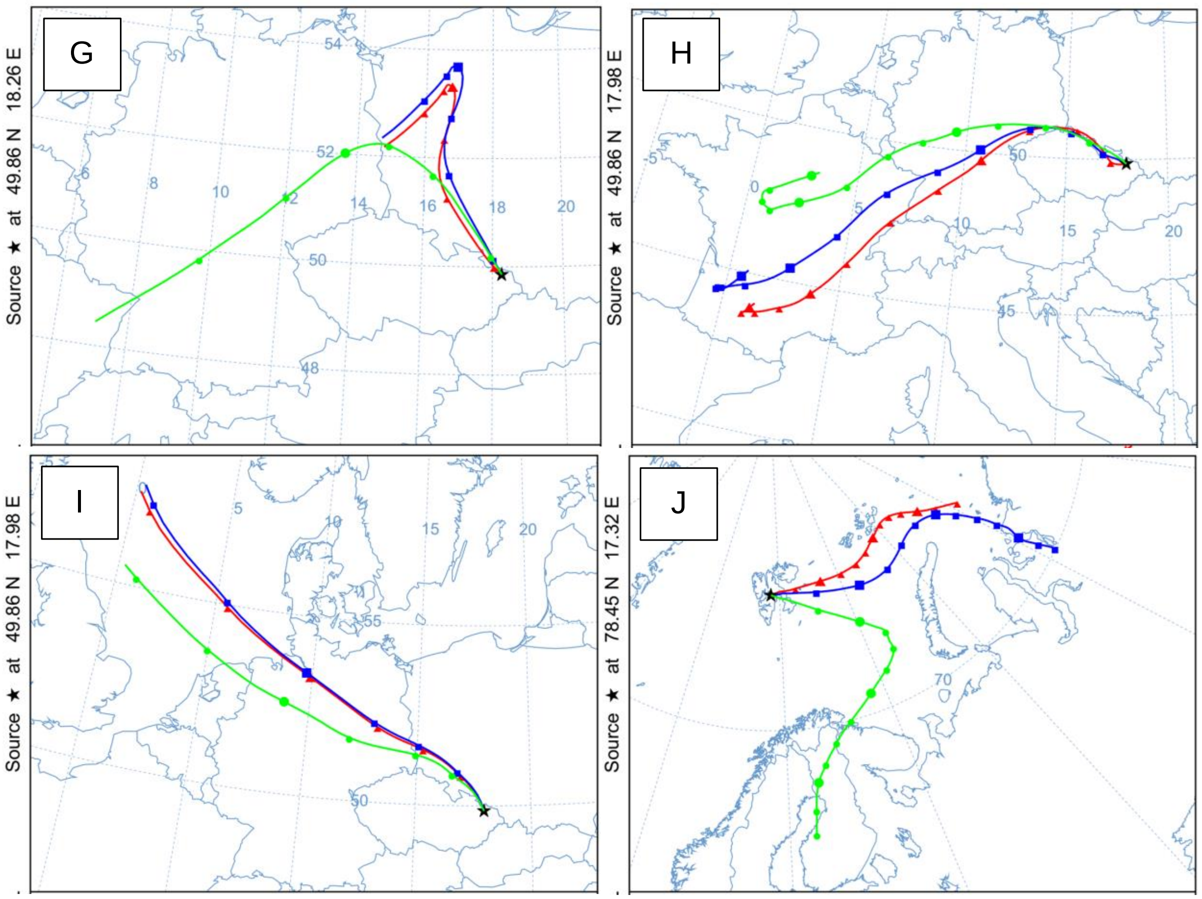


Figure S2. Air mass back trajectories from the sites immediately before sample collection. Tick marks indicate 6-hour intervals; colors indicate different arrival heights: red = 200, blue = 500, green = 1000 m above ground



A: Altenberg (Rr2), B: Winterberg (Rr1), C: Mainz 2015 (Ub1), D: Mainz 2017 (Ub4), E: Götzens (Ub2), F: Kolsassberg (Rr3)

Figure S2 continued



G: Ostrava (Ub3), H: Pustá Polom 1 (Rr4), I: Pustá Polom 2 (Rr5), J: Tempelfjorden (Rm1)

Table S4A. Total N/O-PAH and NMAH concentrations in snow (sum of dissolved and particulate in ng L^{-1})

	Ub1 Mainz	Ub2 Götzens	Ub3 Ostrava	Ub4 Mainz	Rr1 Winterberg
1,4-naphthoquinone	<LOQ	<LOQ	18.57	7.07	<LOQ
9-fluorenone	57.07	40.05	53.87	26.13	44.56
Acenaphthoquinone	144.89	188.79	141.83	781.83	190.83
9,10-anthraquinone	55.15	195.39	281.66	22.29	257.34
Benzanthrone	<LOQ	<LOQ	478.34	3.30	20.05
Benz(a)fluorenone	5.32	7.46	45.91	7.49	16.27
Benz(b)fluorenone	2.86	8.30	36.15	6.26	18.86
1,2-Benzanthraquinone	4.17	11.77	57.82	7.26	17.36
1-nitronaphthalene	0.58	1.34	<LOQ	0.57	1.07
2-nitronaphthalene	<LOQ	0.09	<LOQ	<LOQ	0.32
9-nitroanthracene	0.66	13.58	<LOQ	<LOQ	7.74
2-nitrofluoranthene	<LOQ	2.55	11.58	2.65	0.31
2,4-dinitrophenol	17.48	23.32	73.24	32.09	124.96
Dinitro-ortho-cresol	8.44	8.56	108.31	16.88	26.22
3-nitrosalicylic acid	2.67	14.25	26.38	146.04	6.03
5-nitrosalicylic acid	10.34	40.07	50.77	258.79	11.99
4-nitrophenol	158.07	971.16	44.58	2262.33	759.96
4-nitroguaiacol	<LOQ	<LOQ	155.22	325.46	<LOQ
3-methyl-4-nitrophenol	25.71	82.06	250.55	158.34	147.67
2-methyl-4-nitrophenol	49.28	150.49	28.37	380.24	213.94
4-nitrocatechol	14.85	50.78	48.18	800.43	50.47
4-methyl-5-nitrocatechol	2.40	7.12	7.71	60.18	7.97
3-methyl-5-nitrocatechol	2.24	8.16	5.84	88.41	5.69
∑ OPAHs	269.5	451.8	1114.1	861.6	565.3
∑ NPAHs	1.2	17.6	11.6	3.2	9.4
∑ NPs*	259.0	1235.6	660.3	3175.4	1272.8
∑ NCs	19.5	66.1	61.7	949.0	64.1
∑ NSAs	13.0	54.3	77.2	404.8	18.0

Ostrava sample was analyzed for N/O-PAHs without phase separation; *∑ NPs = 4-NP + ∑M-NPs + 4-NG + 2,4-DNP + DNOC

Table S4A continued.

	Rr2 Altenberg	**Rr3** Kolsassberg	**Rr4** Pustà Polom	**Rr5** Pustá Polom	**Rm1** Tempelfjorden
1,4-naphthoquinone	<LOQ	<LOQ	14.85	12.29	<LOQ
9-fluorenone	38.39	55.83	41.94	57.59	2.55
Acenaphthoquinone	152.63	178.38	399.54	609.85	5.71
9,10-anthraquinone	241.29	318.67	234.94	607.67	<LOQ
Benzanthrone	<LOQ	1.99	626.56	758.06	0.66
Benz(a)fluorenone	16.36	27.39	78.85	95.93	0.15
Benz(b)fluorenone	12.38	17.94	60.13	106.51	<LOQ
1,2-Benzanthraquinone	17.67	23.67	86.74	136.51	0.13
1-nitronaphthalene	0.98	0.47	<LOQ	<LOQ	<LOQ
2-nitronaphthalene	0.22	<LOQ	<LOQ	<LOQ	<LOQ
9-nitroanthracene	4.99	13.24	<LOQ	<LOQ	<LOQ
2-nitrofluoranthene	2.64	1.43	19.13	13.78	<LOQ
2,4-dinitrophenol	47.21	24.72	94.31	14.79	13.27
Dinitro-ortho-cresol	14.80	6.51	85.52	12.20	3.61
3-nitrosalicylic acid	1.16	7.00	69.72	34.84	5.69
5-nitrosalicylic acid	6.56	31.22	313.46	105.54	16.31
4-nitrophenol	566.68	564.94	18.35	15.40	310.33
4-nitroguaiacol	18.5	<LOQ	169.40	282.34	14.79
3-methyl-4-nitrophenol	41.28	67.49	218.59	569.09	4.59
2-methyl-4-nitrophenol	33.88	98.81	259.31	71.85	18.39
4-nitrocatechol	19.07	25.57	30.43	67.10	<LOQ
4-methyl-5-nitrocatechol	6.32	4.20	2.27	61.48	<LOQ
3-methyl-5-nitrocatechol	2.35	5.49	2.51	32.84	3.51
∑ OPAHs	478.7	623.9	1543.6	2384.4	9.2
∑ NPAHs	8.8	15.1	19.1	13.8	<LOQ
∑ NPs*	722.4	762.5	845.5	965.7	365.0
∑ NCs	27.7	35.3	35.2	161.4	3.5
∑ NSAs	7.7	38.2	383.2	140.4	22.0

Pustà Polom samples were analyzed for N/O-PAHs without phase separation

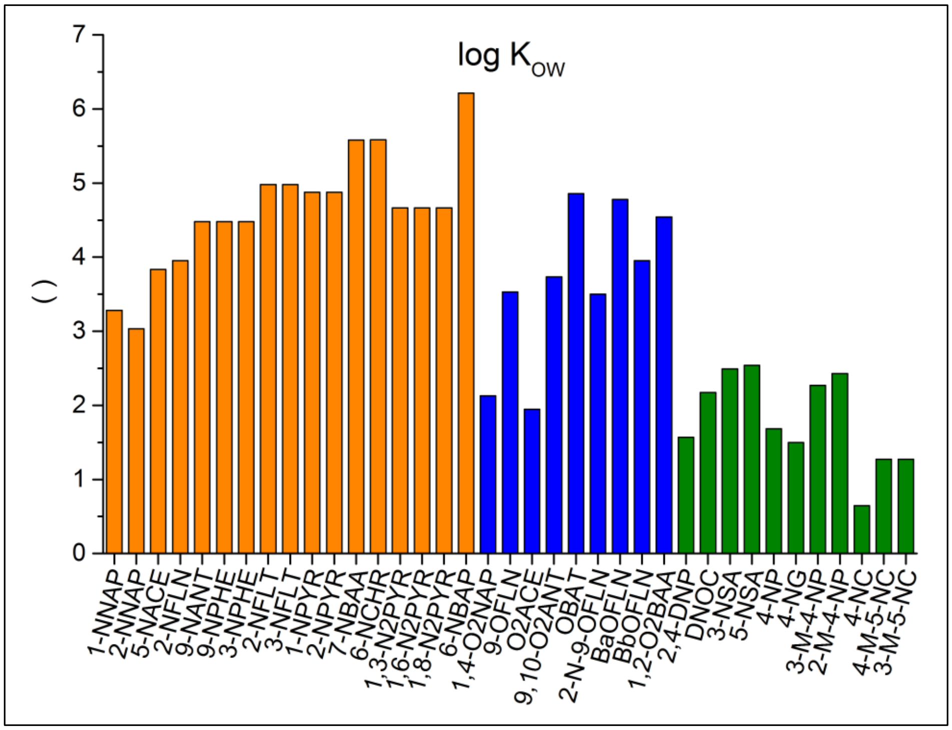
* ∑ NPs = 4-NP + ∑M-NPs + 4-NG + 2,4-DNP + DNOC

Table S4B. Analytes in air gas and particulate phases (ng m^{-3}), and in snow sample from Ostrava (Ub3; ng l^{-1})

	c_g (ng m^{-3})	c_p (ng m^{-3})	$c_{(pd + pp)}$ (ng l^{-1})	W_T	θ 273 K	log K_{OW} 298 K
1,4-naphthoquinone	0.28	0.031	18.57	6.0×10^4	0.001	2.13
9-fluorenone	18.9	4.98	53.87	2.3×10^3	0.12	3.53
Acenaphthoquinone	n.d.	n.d.	141.83	n.a.		
9,10-anthraquinone	1.89	6.31	281.66	3.4×10^4	0.81	3.74
Benzanthrone	n.d.	12.8	478.34	3.7×10^4	0.91	4.86
Benz(a)fluorenone	0.16	6.49	45.91	6.9×10^3	0.99	4.78
Benz(b)fluorenone	n.d.	8.67	36.15	4.2×10^3	0.98	3.95
1,2-benzanthraquinone	n.d.	n.d.	57.82	n.a.		
1-nitronaphthalene	0.23	0.042	n.d.	n.a.		
2-nitronaphthalene	0.21	n.d.	n.d.	n.a.		
5-nitroacenaphthene	0.0002	0.014	n.d.	n.a.		
9-nitroanthracene	0.032	0.42	n.d.	n.a.		
2-nitrofluoranthene	0.0010	0.55	11.58	2.1×10^4	1.00	4.98
2-nitropyrene	0.00013	0.16	n.d.	n.a.		
7-nitrobenz(a)anthracene	n.d.	0.33	n.d.	n.a.		
6-nitrochrysene	n.d.	0.031	n.d.	n.a.		
2,4-dinitrophenol	n.a.	n.d.	73.24	n.a.		
Dinitro-ortho-cresol	n.a.	n.d.	108.31	n.a.		
3-nitrosalicylic acid	n.a.	0.46	26.38	5.7×10^4	0.61	2.49
5-nitrosalicylic acid	n.a.	0.62	50.77	8.2×10^4	1.00	2.54
4-nitrophenol	n.a.	3.34	44.58	1.3×10^4	1.00	1.68
4-nitroguaiacol	n.a.	n.d.	155.22			
3-methyl-4-nitrophenol	n.a.	1.59	250.55	1.6×10^5	1.00	2.27
2-methyl-4-nitrophenol	n.a.	2.14	28.37	1.3×10^4	1.00	2.43
4-nitrocatechol	n.a.	17.26	48.18	2.8×10^3	1.00	0.64
4-methyl-5-nitrocatechol	n.a.	7.09	7.71	1.1×10^3	1.00	1.27
3-methyl-5-nitrocatechol	n.a.	5.26	5.84	1.1×10^3	1.00	1.27

c_g: analyte concentrations in the gas phase, c_p: concentrations in the particulate phase, c_{pd+pp}: sum of the concentrations in snow dissolved and particulate phases; W_T: total scavenging ratios (dimensionless) = c_{pd+pp} (ng m^{-3})/ [$c_g + c_p$ (ng m^{-3})]; W_T was only determined for substances that were detected in both air and snow samples; n.d.: not detected; n.a.: not available; the table only lists substances which were detected in either air or snow samples; NMAH concentrations were not determined in the gas phase; however, except for 2,4-dinitrophenol and dinitro-ortho-cresol, which were not found in the air particulate phase, based on our modeled particulate mass fractions (Figure 3C), we expect the rest of NMAHs to be mainly in the particulate phase at near zero temperatures. Therefore, we do not expect the lack of NMAH measurement in the gas phase to have any significant effect on the calculated W_T values.

Figure S3 Octanol-water partitioning coefficient, K_{ow}, calculated using ppLFER model in Table S3



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