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Supporting Information

Tracing sources of PCDD/Fs in Baltic Sea air by using metals as source markers

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7 pages, 3 tables, 1 figure

Table S1. Location and other characteristics of the pine needle sampling sites as reported in Holt *et al.*[1] and used in this study to identify metal markers for various thermal activities

Source type	Category	Short name in Figure 4	Short name in Holt *et al.*[1]	Country	Sampling site and thermal activity	Co-ordinates
Copper smelter	Industrial	Cu Smelter	SK-KR	Slovakia	Krompachy; near a copper smelter	48°55'23,80"N 20°56'05,80"E
Steel smelter		Steel	CZ-BA	Czech Republic	Bartovice-Ostrava; near steel smelter	49°48'28,55"N 18°20'35,04"E
Secondary steel production with electric arc furnace		ElArc	CZ-HR	Czech Republic	Hrádek-Rokycany; near steelworks with electric arc furnace	49°43'16,2"N 13°38'08,6"E
Brown coal burning		Coal	CZ-TR	Czech Republic	Trmice; near thermal and electric power station powered by brown coal	50°38'45,07"N 13°59'27,93"E
Heavy oil burning		Heavy Oil	CZ-BB	Czech Republic	Bělá pod Bezdězem; near a boiler-house powered using heavy oil (mazut)	50°29'19,00"N 14°50'22,80"E
Traffic emissions	Urban	Traffic	CZ-BR	Czech Republic	Brno; second largest city in the CR; sampling site located near high traffic area	49°10'32,20"N 16°37'20,30"E
Municipal solid waste incineration		MSWI	CZ-PR	Czech Republic	Prague; sampling site located near a MSWI facility	50°05'11,00"N 14°32'12,70"E
Emissions from city		City	SW-UM	Sweden	The city of Umeå; population ~120,000, sampling at a site not heavily impacted by traffic, MSWI, domestic burning and/or industrial emissions	63°49'4.43"N 20°17'7.92"E
Domestic burning		Domestic	SW-LK	Sweden	Lycksele; rural town (population 8,500), known for poor air quality during winter time, highly impacted by domestic burning	64°35'60.00"N 18°40'0.00"E

Table S2. PCA dataset of average metal composition of pine needle samples (*n*=3) collected at sites near thermal sources in Sweden, Czech Republic and Slovakia, expressed as fraction (0-1) of individual metal in relation to total metal level

Source type	Cd	Co	Cr	Cu	Fe	K	Mn	Ni	Pb	Sb	V	Zn	Hg	Se	Tl
Copper smelter	0.025	0.10	0.051	0.21	0.044	0.055	0.027	0.031	0.10	0.089	0.049	0.076	0.093	0.023	0.023
Steel smelter	0.089	0.079	0.053	0.079	0.046	0.085	0.080	0.13	0.068	0.040	0.067	0.056	0.043	0.048	0.037
Secondary steel production using electric arc furnace	0.047	0.045	0.020	0.075	0.017	0.073	0.31	0.018	0.029	0.075	0.017	0.19	0.080	0.009	NaN
Brown coal burning	0.17	0.048	0.041	0.13	0.060	0.11	0.020	0.12	0.047	0.038	0.028	0.13	0.051	0.015	NaN
Heavy oil burning	0.044	0.13	0.027	0.095	0.032	0.069	0.033	0.060	0.055	0.088	0.039	0.059	0.10	0.14	0.027
Traffic emissions	0.11	0.088	0.034	0.065	0.034	0.090	0.080	0.099	0.025	0.070	0.034	0.060	0.15	0.041	0.022
Municipal solid waste incineration	0.063	0.050	0.027	0.17	0.034	0.13	0.11	0.065	0.044	0.045	0.030	0.061	0.13	0.010	0.030
Emissions from city	0.048	0.030	0.031	0.048	0.039	0.13	0.16	0.029	0.016	0.073	0.066	0.12	0.21	NaN	NaN
Domestic burning	0.056	0.076	0.047	0.096	0.054	0.088	0.19	0.11	0.019	0.081	0.052	0.053	NaN	0.084	NaN

NaN: Missing data, i.e. the concentration was reported as <LOD and no value is attributed in the composition dataset to allow replacement with PCA-based best guess

Table S3. Atmospheric concentrations of PCDD/Fs, PCBs, HCB and metals in N-, E-, S- and W-sectors of Aspvreten, and summer to winter percentage increases in the levels of these organic pollutants and metals.

	Summer levels per sector[a, b]				Winter levels per sector[a, b]				Summer-to-winter increase (%)[c]			
Sector	**N**	**E**	**S**	**W**	**N**	**E**	**S**	**W**	**N**	**E**	**S**	**W**
2378-TCDD	0.19[d]	0.27[d]	0.26[d]	0.27[d]	0.23[d]	0.22[d]	0.53[d]	0.25[d]	20	-19	110	-4.5
12378-PeCDD	0.28[d]	0.24[d]	0.24[d]	0.23[d]	1.1[d]	0.24[d]	1.8[d]	0.16[d]	280	-0.83	660	-29
123478-HxCDD	0.020[d]	0.022[d]	0.025[d]	0.023[d]	0.10[d]	0.033[d]	0.25[d]	0.019[d]	420	51	930	-16
123678-HxCDD	0.059[d]	0.027[d]	0.026[d]	0.021[d]	0.16	0.085	0.50	0.081	180	220	1900	280
123789-HxCDD	0.044[d]	0.022[d]	0.039[d]	0.020[d]	0.18	0.11	0.43	0.083	300	410	1000	320
1234678-HpCDD	0.025[d]	0.024[d]	0.065[d]	0.033[d]	0.044	0.028	0.12	0.028	80	17	82	-16
OCDD	2.9X10^{-3}	3.4X10^{-3}	0.021	3.5X10^{-3}	0.010	6.1X10^{-3}	0.020	6.1X10^{-3}	230	79	-3.0	75
2378-TCDF	0.039[d]	0.083[d]	0.047[d]	0.032[d]	0.22	0.27	0.97	0.089	470	230	2000	180
12378-PeCDF	5.9X10^{-3} [d]	0.010[d]	6.2X10^{-3} [d]	7.0X10^{-3} [d]	0.051	0.065	0.23	0.010	770	580	3600	45
23478-PeCDF	0.065[d]	0.12[d]	0.078[d]	0.10[d]	0.81	0.98	3.9	0.24	1200	710	4900	130
123478-HxCDF	0.028[d]	0.062[d]	0.024[d]	0.025[d]	0.25	0.36	1.4	0.070	800	480	5800	180
123678-HxCDF	0.022[d]	0.059[d]	0.023[d]	0.021[d]	0.22	0.39	1.5	0.085	910	550	6200	290
123789-HxCDF	0.016[d]	0.022[d]	0.021[d]	0.020[d]	0.014[d]	0.015[d]	0.094[d]	0.054[d]	-15	-32	360	180
234678-HxCDF	0.035[d]	0.073[d]	0.022[d]	0.023[d]	0.20	0.44	1.7	0.11	490	500	7600	780
1234678-HpCDF	0.014	0.036	8.8X10^{-3}	0.011	0.086	0.14	0.49	0.033	500	290	5500	190
1234789-HpCDF	4.6X10^{-3} [d]	6.6X10^{-3} [d]	6.2X10^{-3} [d]	5.9X10^{-3} [d]	0.012[d]	0.014[d]	0.056[d]	2.6X10^{-3} [d]	170	110	810	-56
OCDF	5.0X10^{-4} [d]	7.8X10^{-4} [d]	6.4X10^{-5} [d]	5.9X10^{-4} [d]	2.3X10^{-3} [d]	2.5X10^{-3} [d]	8.9X10^{-3} [d]	3.2X10^{-4} [d]	360	220	1300	-46
PCB28	3.1	5.8	4.9	3.5	2.3	1.7	2.7	2.0	-25	-71	-45	-44
PCB52	2.3	3.7	4.2	2.8	1.6	1.0	1.7	1.2	-29	-72	-60	-58
PCB101	2.2	3.8	5.1	3.5	1.1	0.91	1.4	1.0	-48	-76	-73	-71
PCB138	1.6	2.6	4.0	2.6	0.54[d]	0.29[d]	0.60[d]	0.40[d]	-65	-89	-85	-85
PCB153	1.8	3.1	5.1	3.4	0.61[d]	0.37[d]	0.64[d]	0.63[d]	-65	-88	-87	-81
PCB180	0.50	1.0	1.3	1.3	0.20[d]	0.15[d]	0.24[d]	0.16[d]	-60	-84	-81	-88
PCB118	0.50	0.94	1.2	0.85	0.18[d]	0.10[d]	0.17[d]	0.11[d]	-65	-89	-85	-88

	Summer levels per sector[a, b]				Winter levels per sector[a, b]				Summer-to-winter increase (%)[c]			
∑PCDD	0.62	0.61	0.68	0.60	1.8	0.72	3.7	0.63	190	19	450	6.2
∑PCDF	0.23	0.47	0.24	0.25	1.9	2.7	10	0.69	720	460	4300	180
∑I-PCB	12	21	26	18	6.6	4.5	7.4	5.5	-45	-78	-71	-70
HCB	51	38	37	46	78	71	96	89	53	86	160	96
As	190[d]	180[d]	240[d]	580	220[d]	200[d]	870	170[d]	18	8.0	260	-71
Cd	6.8[d]	77	30	140	32	84	270	14[d]	380	8.8	830	-90
Co	16[d]	24	36	130	41	45	72	13[d]	160	86	100	-90
Cr	450	690	530	700	1300	750	1400	1000	190	9.1	160	49
Cu	230	750	9600	$1.1X10^{4}$	1100	850	1400	590	370	13	-85	-94
Fe	$4.1X10^{4}$	$3.7X10^{4}$	$4.2X10^{4}$	$8.3X10^{4}$	$4.6X10^{4}$	$3.6X10^{4}$	$7.4X10^{4}$	$2.6X10^{4}$	10	-4.5	76	-69
Hg	20[d]	27[d]	27[d]	27[d]	26[d]	29[d]	30[d]	25[d]	27	8.0	9.2	-6.2
K	$4.5X10^{4}$	$7.2X10^{4}$	$1.3X10^{4}$	$1.4X10^{4}$	$7.7X10^{4}$	$1.2X10^{5}$	$1.8X10^{5}$	$7.8X10^{4}$	69	72	31	-44
Mn	1200	1500	1600	3000	1600	1400	3000	1200	38	-5.4	90	-61
Ni	260	490	640	1700	870	1300	1300	400	230	170	110	-77
Pb	330	1300	1200	2100	1200	2800	7700	590	260	110	570	-72
Sb	61[d]	98	120	130	76[d]	98	370	61[d]	24	$-8.0X10^{-3}$	210	-51
Se	68[d]	90	120	130	120	110	380	130	75	20	210	-1.3
Tl	39[d]	45[d]	121	45[d]	43[d]	49[d]	66[d]	42[d]	12	8.0	-45	-6.2
V	200	520	490	1600	490	2700	2500	280	140	410	420	-83
Zn	2200	6400	9200	$1.5X10^{4}$	6700	9400	$2.3X10^{4}$	2900	210	47	150	-81

[a] Concentrations of PCDD/Fs are given in fg WHO-TEQ m^{-3}, PCBs and HCB are given in pg m^{-3}
[b] Concentrations of metals given in pg m^{-3}
[c] Calculated as (Winter level – Summer level) / Summer level * 100
[d] Average sector concentration was calculated by replacing <LOD values with half LOD

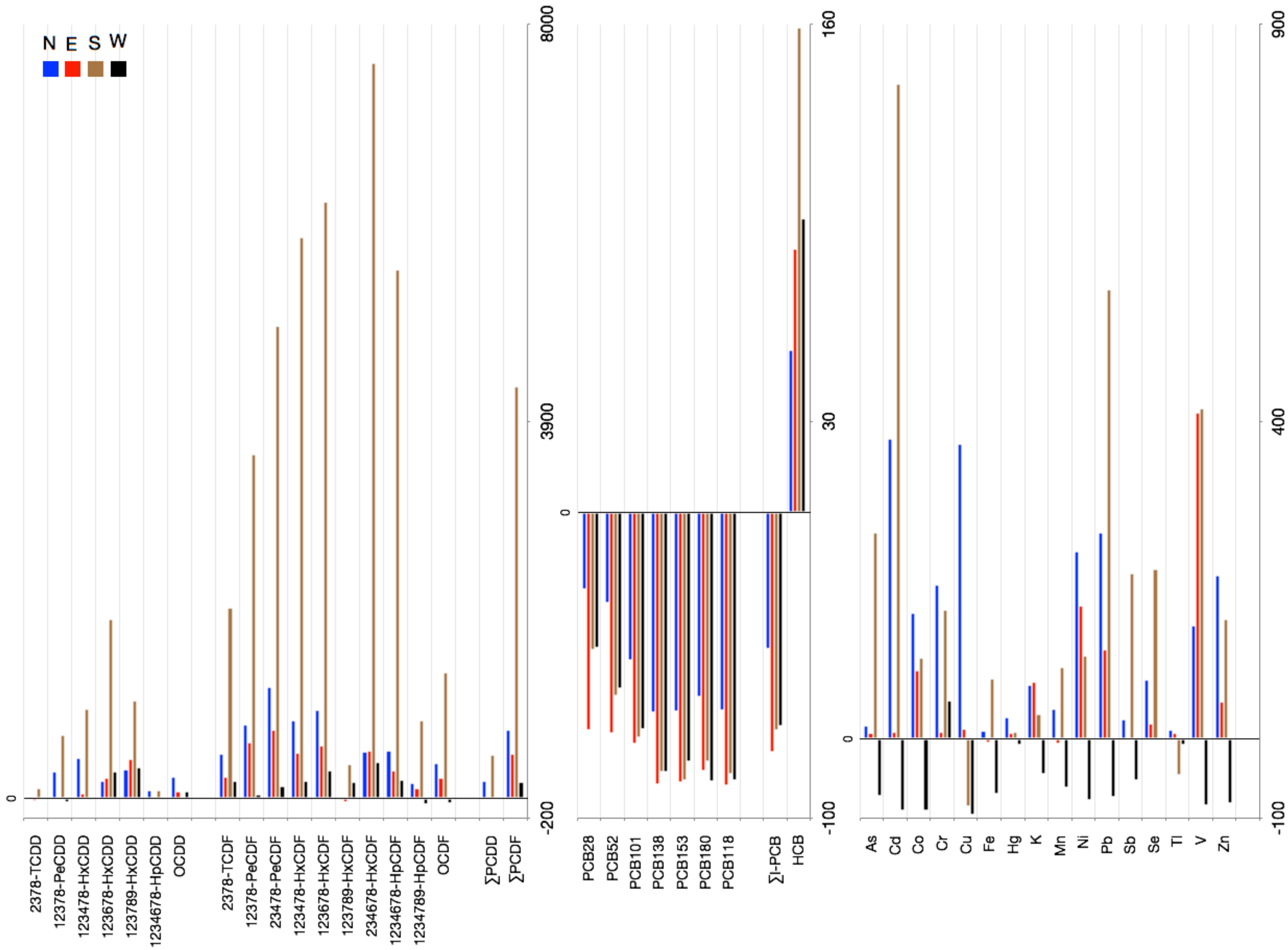

Figure S1. Percentage magnifications of POPs and metals during winter season relative to the preceding summer season in air samples from Aspvreten

REFERENCES:

1. E. Holt, A. Kocan, J. Klanova, A. Assefa and K. Wiberg, *Chemosphere*, 2016, **156**, 30-36.