

Supplementary material

In search of potential source regions of semi-volatile organic contaminants in air in the Yukon Territory, Canada from 2007 to 2009 using hybrid receptor models

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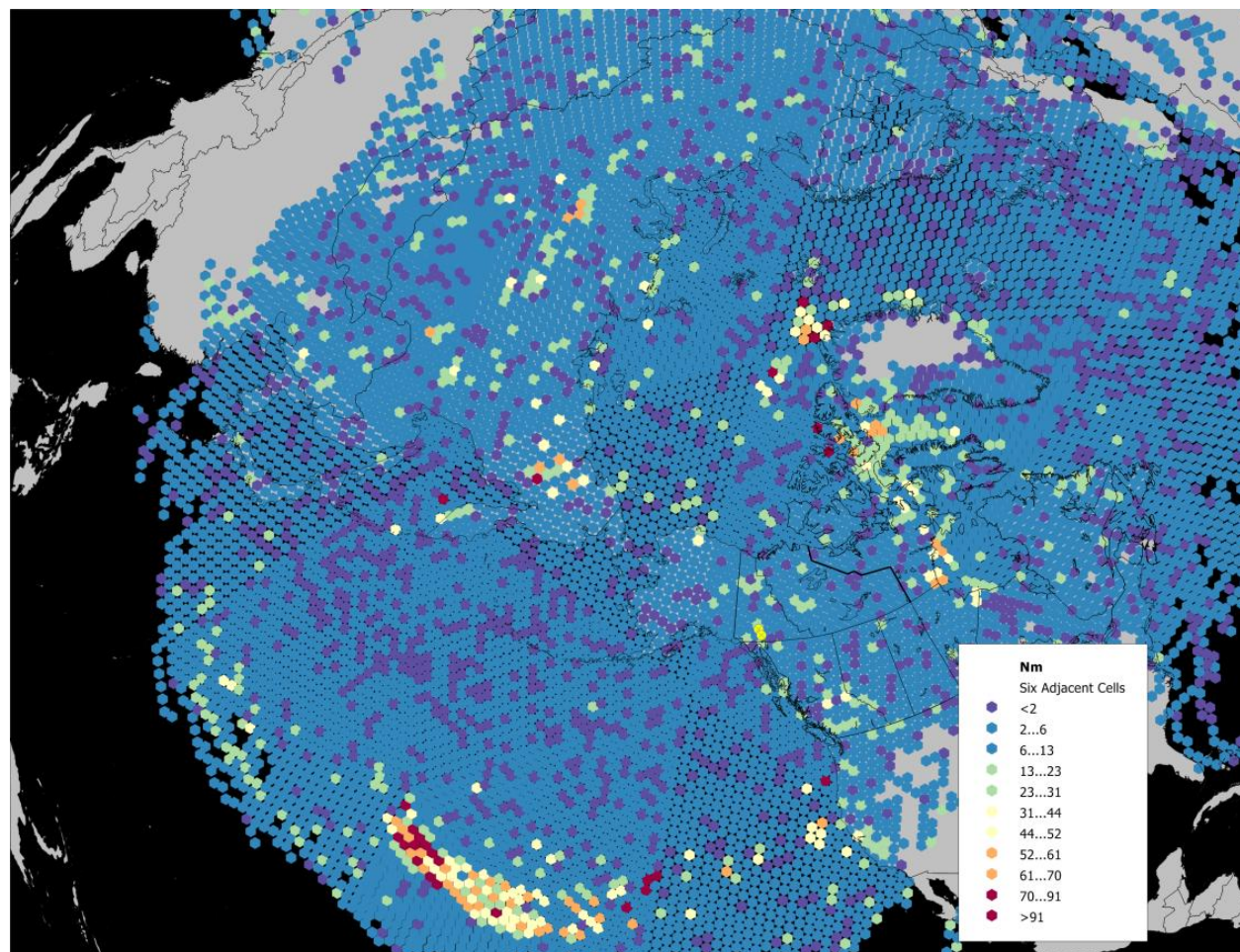


Fig. S1. All endpoint counts for the six cells adjacent to the cell in which LFL lies, divided by six, for trajectories arriving while the air sampler was active. The range is very small and quite 'flat', so that, as a set, there is no single source region to the area suggested, perhaps excepting the belt in the middle of the Pacific Ocean. Note that this is strictly meteorological: there are no measurements or emission estimates associated with these values.

Method detection limits

Method detection limits were defined as in Su et al.^[1] As these are based on field and laboratory blanks they are not actually associated with a volume of air and are presented in Table S1 using an assumed air volume of 12 000 m,^[2] the theoretical average volume of a weekly sample. It is for this reason that compounds that are found near detection limits in the week-long samples can also be found in the day-long samples: the detection limits are independent of sample volume.

Table S1. Method detection limits (MDLs)

Compound	MDL (pg m ⁻³)	Compound	MDL (pg m ⁻³)
1,2,4,5-tetrachlorobenzene	0.62	Trichloroveratrole	0.17
1,2,3,4-tetrachlorobenzene	0.62	Tetrachloroveratrole	0.17
Pentachlorobenzene	0.27	Endrin	0.18
Hexachlorobenzene	0.47	1,3-dichlorobenzene	1.43
α -hexachlorocyclohexane	0.07	1,4-dichlorobenzene	12.20
β -hexachlorocyclohexane	0.11	1,2-dichlorobenzene	2.46
γ -hexachlorocyclohexane	0.05	1,3,5-trichlorobenzene	0.19
Heptachlor	0.27	1,2,4-trichlorobenzene	10.40
Octachlorostyrene	0.03	1,2,3-trichlorobenzene	0.92
Oxychlorane	0.15	Hexachlorobutadiene	0.05
<i>trans</i> -chlordane	0.11	Aldrin	0.20
<i>cis</i> -chlordane	0.10	β -endosulfan	0.29
<i>trans</i> -nonachlor	0.07	BDE17	0.05
<i>cis</i> -nonachlor	0.05	BDE28+33	0.04
Heptachlor epoxide	0.15	BDE47	0.21
Dieldrin	0.12	BDE66	0.05
<i>ortho,para'</i> -dichlorodiphenyldichloroethene	0.10	BDE100	0.07
<i>para,para'</i> -dichlorodiphenyldichloroethene	0.42	BDE99	0.25
<i>ortho,para'</i> -dichlorodiphenyldichloroethane	0.19	BDE85	0.10
<i>para,para'</i> -dichlorodiphenyldichloroethane	0.75	BDE154	0.04
<i>ortho,para'</i> -dichlorodiphenyltrichloroethane	0.25	BDE153	0.06
<i>para,para'</i> -dichlorodiphenyltrichloroethane	0.43	HBCD	3.35
Pentachloroanisole	0.11	BDE138	0.05
α -endosulfan	0.07	BDE183	0.04
Methoxychlor	2.64	BDE190	0.08
		BDE209	0.32
		BDE49	0.18

Table S2. Concentrations of OCs (pg m⁻³) measured in week-long samples in this and previous work at LFL

IBP, industrial by-product; F, fungicide; CBP, combustion by-product; TP, transformation product; I, insecticide; M, miticide; R, rodenticide; IS, industrial solvent. Status under the Stockholm Convention: N, not banned; B, banned; L, listed for future banning; R, restricted use. AMDL, above method detection limits. MeanA, mean calculated only from AMDL values, for comparison to values from Su et al.^[1]; MeanB, mean calculated with random value from 0 to 2/3 MDL for all BDLs, for comparison to values in Su et al.^[3]

Compound	Short name	Uses	Status	Fraction AMDL	This work Concentration					Previous studies at same site Concentration			
					MeanA	MeanB	Med	Min	Max	Mean	Med	Min	Max
Chlorobenzenes													
tri	1,2,3-CIBz	BP	N	0.03	1.45 ^A	0.45	1.53	1.28	1.87				
tetra	1,2,3,4-CIBz	BP	N	0.20	2.39 ^A	0.77	1.49	0.35	9.89				
	1,2,4,5-CIBz	BP	N	0.27	1.63 ^A	0.45	1.13	0.49	5.55				
penta	5CIBz	BP	B	1.00	NR ^B	NR	NR	NR	NR				
hexa	6CIBz	F, IBP, CBP	B	1.00	NR ^B	NR	NR	NR	NR	88 ^E	86	71	120
Chloroveratroles													
tri	3CIVER	IBP	N	0.01	n/a ^C	0.08	0.69	n/a ^C	n/a ^C				
tetra	4CIVER	IBP	N	0.98	2.64	2.56	1.95	0.28	8.61	0.92 ^D	0.05	0.03	9.9
Pentachloroanisole	5CIANI	TP	N	0.30	2.47	0.78	0.40	0.13	19.3	4.6 ^D	3.9	0.28	19
Octachlorostyrene	8CISTY	IBP	N	0.92	0.40	0.36	0.42	0.04	0.87	0.27 ^D	0.31	0.04	0.52
Endosulphans													
alpha	α-ENDO	I	L	0.58	2.70	1.16	2.50	0.37	10.2	8.3 ^D	5.8	3.1	18
Hexachloro-cyclohexanes													
alpha	α-HCH	IBP	B	0.98	7.81	7.56	6.01	0.34	22.1	48 ^E	50	17	87
beta	β-HCH	IBP	B	0.14	1.71	0.32	1.18	0.28	4.15	0.19 ^E	0.09	0.02	0.85
gamma	γ-HCH	I,M	R	0.59	0.96	0.58	0.62	0.10	6.90	4.5 ^E	3.9	1.7	10
Chlordanes													
cis	c-CHLOR	I	B	0.60	0.36	0.23	0.30	0.10	0.93	0.50 ^D	0.48	0.23	0.85
trans	t-CHLOR	I	B	0.10	0.31	0.11	0.29	0.20	0.52	0.14 ^D	0.13	0.07	0.24
oxy	OxCHLOR	TP	n/a	0.01	n/a ^C	0.25	0.54	n/a ^C	n/a ^C	0.30 ^D	0.30	0.14	0.47
Nonachlors													
cis	c-NONA	I	B	0.07	0.18	0.04	0.13	0.08	0.36				
trans	t-NONA	I	B	0.59	0.25	0.16	0.22	0.09	0.68	0.36 ^D	0.29	0.19	0.64
Dieldrin	DIEL	I	B	0.50	0.30	0.18	0.29	0.16	0.50	0.31 ^D	0.28	0.11	0.60
Endrin	ENDR	I,R	B	0.02	1.14	0.19	n/a ^F	0.89	1.39				

Hexachlorobutadiene	6ClBD	IS	N	0.89	0.54	0.49	0.24	0.09	6.85				
Heptachlors epoxide	HEPOx	TP	n/a	0.13	1.25	0.34	0.92	0.42	3.72	0.53 ^D	0.53	0.21	0.82
Chlorodiphenyl- chloroethane/enes													
<i>ortho,para</i> -di,tri-ane	<i>o,p'</i> -DDT	I	R	0.04	0.46	0.19	0.49	0.27	0.60	0.21 ^D	0.18	0.06	0.45
<i>para,para</i> -di,tri-ane	<i>p,p'</i> -DDT	I	R	0.02	0.76	0.20	n/a ^F	0.69	0.83	0.19 ^D	0.16	0.03	0.60
<i>ortho,para</i> -di,di-ane	<i>o,p'</i> -DDD	I	N	0.01	n/a ^C	0.10	1.01	n/a ^C	n/a ^C				
<i>para,para</i> -di,di-ane	<i>p,p'</i> -DDD	I	N	0.01	n/a ^C	0.36	1.76	n/a ^C	n/a ^C				
<i>para,para</i> -di,di-ene	<i>p,p'</i> -DDE	TP	n/a	0.18	0.67	0.31	0.67	0.34	0.96	0.30 ^D	0.29	0.06	0.69
methoxychlor	MeOx	I	N	0.02	8.55	1.49	n/a ^F	4.50	12.6				

^ACompound quantitation suspect due to possible breakthrough.

^BNR, not reported due to breakthrough.

^CCompound detected in only one sample; only median reported.

^DValues taken from Su et al.^[3]

^EValues taken from Su et al.^[1]

^FCompound detected in only 2 samples; median not reported.

Table S3. Concentrations of polycyclic aromatic hydrocarbons (PAHs) and brominated flame retardants (BFRs) (pg m^{-3}) measured in week-long samples in air at Little Fox Lake

AMDL, above method detection limits. MeanA, mean calculated only from AMDL values, MeanB, mean calculated with random value from 0 to 2/3 MDL for all BDLs. ND, not detected

Compound	Short name	Fraction AMDL	Concentration				
			MeanA	MeanB	Med	Min	Max
PAHs							
Anthracene	ANTH	0.19	14.6	3.69	5.1	2.9	88.3
Benz[a]anthracene	BAAN	0.91	3.8	3.42	2.0	0.5	48.9
Benzo[b]fluoranthene	BBFL	0.94	5.8	5.42	2.6	0.4	45.8
Benzo[e]pyrene	BEPY	0.73	4.3	3.18	2.7	0.5	21.4
Benzo[k]fluoranthene	BKFL	0.72	4.0	2.95	2.3	0.4	27.4
Chrysene	CHRY	0.86	9.6	8.36	5.6	0.9	73.4
Fluorene	FLUR	0.98	77.9	76.3	43.0	6.8	1070
Fluoranthene	FLUT	0.73	48.2	36.4	34.6	13.6	226
Phenanthrene	PHEN	0.99	158	156	116	46.5	1790
Pyrene	PYRE	0.09	90.5	30.3	77.1	68.1	145
Retene	RETE	0.80	195	157	85.8	14.4	6160
Brominated diphenyl ethers							
2,2',4	BDE17	0.12	0.08	0.02	0.06	0.05	0.20
2,4,4'+2,3',4'	BDE28+33	0.65	0.06	0.05	0.05	0.03	0.34
2,2',4,4'	BDE47	0.96	1.42	1.36	0.76	0.09	21.0
2,2',4,5'	BDE49	0.07	0.35	0.11	0.28	0.22	0.75
2,3',4,4'	BDE66	0.39	0.09	0.04	0.08	0.04	0.37
2,2',3,4,4'	BDE85	0.30	0.15	0.07	0.13	0.08	0.43
2,2',4,4',5	BDE99	0.94	1.35	1.28	0.82	0.13	11.6
2,2',4,4',6	BDE100	0.91	0.35	0.31	0.19	0.03	4.39
2,2',3,4,4',5'	BDE138	0.05	0.12	0.02	0.06	0.05	0.36
2,2',4,4',5,5'	BDE153	0.62	0.12	0.08	0.09	0.03	0.51
2,2',4,4',5,6'	BDE154	0.77	0.10	0.08	0.07	0.03	0.39
2,2',3,4,4',5',6	BDE183	0.23	0.08	0.03	0.04	0.02	0.48
2,3,3',4,4',5,6	BDE190	ND	ND	0.02	ND	ND	ND
Hexabromocyclododecane	HBCD	0.03	7.15	1.28	5.85	3.79	11.8

Relations to ambient air temperature

The air concentrations of all detected organochlorine compounds and brominated flame retardants were regressed against the mean air temperature during sample collection as recorded by a meteorological station at the Little Fox Lake sampling site (a few samples have no temperature data due to equipment malfunction). Table S4 lists the fitting parameters for analytes with significant ($P < 0.01$) correlations between the decadic logarithm of air concentration and temperature ($1/T$, K). The relationships between temperature and polycyclic aromatic hydrocarbon concentrations are the subject of previous work.^[2]

The majority of analytes did not have strong correlations with ambient air temperature. In several cases this may at least be in part because of low detection frequencies. Secondary emission, that is, revolatilisation of previously deposited chemical from surface media, such as soil, snow or water bodies, ought to be the dominant source to the atmosphere for those analytes that are no longer produced or used. A strong positive relation between atmospheric concentration and air temperature (negative correlation with inverse air temperature) would suggest an analyte with diffuse secondary sources in the vicinity of the sampling site. This would be confounded by non-temperature related seasonal variations in source strength as would be expected for pesticides still in use, and polycyclic aromatic hydrocarbons from the combustion of heating fuels.

Other than polycyclic aromatic hydrocarbons, most SVOCs are not expected to display a significant negative correlation with temperature, as both vapour pressure and emissions are lower in the cold winter months. Yet the temperature regressions of the five most volatile SVOCs considered here do have positive slopes (Table S3), which is consistent with a higher capacity of the polyurethane foam plugs for these analytes at lower temperatures. This interpretation is bolstered by the strong relationship between inverse temperature and air concentrations of hexachlorobenzene, which is too volatile to be sampled quantitatively, and supports the purported approach towards equilibrium between atmosphere and polyurethane foam plug for the chlorinated benzenes.

Table S4. Significant ($P < 0.01$) regression parameters from fit of the decadic logarithm of organochlorine compound and brominated flame retardant concentration against inverse temperature (K)

Analyte	R^2	Slope
c-CHLOR	0.35	-800
α -ENDO	0.21	-5700
HEPOx ^A	0.59	-9200
6ClBD	0.34	3200
6ClBz	0.22	31000
α -HCH	0.23	-17000
8ClST	0.31	-500
5ClBz	0.56	18000
1,2,3,4-ClBz	0.44	7900
1,2,4,5-ClBz	0.45	5500
4ClVER	0.22	-6000
t-NONA	0.17	-400
BDE28+33	0.27	-600
BDE47	0.27	-1200
BDE49	0.19	-500
BDE66	0.22	-550
BDE85	0.12	-300
BDE99	0.29	-1250
BDE100	0.32	-1250
BDE153	0.16	-600
BDE154	0.35	-1000

^ASignificant only at the $P < 0.05$ level.

Table S5. Concentrations ($\mu\text{g m}^{-3}$) in each week-long sample of the SVOCs that were above the method detection limits

Date start	1,2,4,5-CIBz	1,2,3,4-CIBz	5CIBz	6CIBz	α -HCH	β -HCH	γ -HCH	8CISTY	OxCHLOR ^A	t-CHLOR	c-CHLOR
12/08/2007	0.64	0.56	1.02	9.31	0.92		0.21				
19/08/2007		0.78	1.47	17.11	1.47		0.14	0.49			
10/09/2007		0.91	1.98	19.78	10.66		1.18	0.31			0.27
16/09/2007	0.94	1.43	2.82	23.45	0.98		0.11	0.27			
23/09/2007		0.45	1.41	17	0.88		0.11	0.25			
30/09/2007			1.53	18.13	6.79		0.76	0.21			0.26
7/10/2007			1.22	17.98	5.55		0.65				0.16
14/10/2007		1.1	3.08	30.57	0.98		0.1				
22/10/2007		1.13	3.31	36.73	13.83		1.13	0.31			0.46
28/10/2007	0.64	1.5	3.94	35.33	12.94		1.13	0.33		0.21	0.28
4/11/2007		1.04	3.4	34.13	7.17		0.84	0.3			0.25
11/11/2007		0.65	2.05	22.58	4.37		0.52	0.23			0.21
18/11/2007	0.62	1.36	3.34	29.61	1.04		0.37	0.27			0.26
25/11/2007	1.14	4.93	8.85	32.48	5.86		0.78	0.25			0.2
2/12/2007	0.87	2.23	7.91	34.09	4.7		0.67	0.22		0.21	0.23
9/12/2007	1.75	3.48	5.97	32.08	6.42		0.87	0.24			0.2
16/12/2007	1.13	2.7	7.44	26.91	4.16		0.41	0.2			0.18
23/12/2007	1.81	4.27	8.58	31.97	5.37		0.54	0.24			0.25
30/12/2007	1.95	4.46	8.82	32.17	5.53		0.58	0.22		0.2	
6/01/2008	1.79	3.85	7.57	34.61	5.31		0.62	0.29			0.25
13/01/2008	0.83	2.44	5.64	29.21	4.53		0.3				0.35
20/01/2008	4.21	0.35	8.12	25.16	2.67		0.46	0.17			0.11
30/01/2008	3.36	4.89	7.08	21.27	1.96		0.34	0.19			
3/02/2008	5.55	9.89	11.12	24.72	2.18		0.33				
10/02/2008	0.64	1.48	4.24	32.17	5.51		1.05			0.24	0.29
17/02/2008	1.51	3.07	5.78	35.33	5.64		0.8			0.3	0.25
24/02/2008	0.49	1.05	2.93	28.66	4.53		0.7	0.16			0.1
5/03/2008	1.01	2.21	5.02	33.12	6.11		0.82	0.33			0.28
9/03/2008			12.24	45.91	9.33		1.07	0.37			0.38
16/03/2008			6.22	46.13	12.05		1.64	0.28			0.37
23/03/2008			7.9	49.55	10.76		1.39	0.49			0.34
30/03/2008			5.45	42.62	13.84		1.92	0.56			0.5
6/04/2008			4.42	51.27	12.44		1.48	0.59			
13/04/2008			1.81	19.66	13.12		2.7	0.53	0.54		0.13
28/04/2008			0.41	2.51	12.34		1.71	0.04			
4/05/2008			2.48	24.92	15.29		5.37	0.48			0.29
11/05/2008			2.35	27.67	15.33		2.16	0.43			0.36
18/05/2008			1.16	11.9	18.09		6.9	0.58			0.62
25/05/2008			1.97	21.51	11.06	0.67	2.21	0.49			

Date start	1,2,4,5-CIBz	1,2,3,4-CIBz	5CIBz	6CIBz	α -HCH	β -HCH	γ -HCH	8CISTY	OxCHLOR ^A	t-CHLOR	c-CHLOR
1/06/2008			2.22	19.66	0.76		0.14	0.68			
8/06/2008			1.09	14.51	10.26		0.77	0.45			0.14
15/06/2008			1.33	13.86	1.1		0.14	0.51			
22/06/2008			1.44	13.77	8.43	0.3	0.29	0.48			
29/06/2008			1.33	12.3	5.71	0.32	0.3	0.43			
6/07/2008			1.48	16.44	18.46	2.06		0.49			0.73
13/07/2008			2.24	21.74	15.19	1.06		0.45			0.76
20/07/2008			1.67	16.41	16.59	1.08		0.44			0.66
27/07/2008			1.6	15.58	21.57			0.61			0.93
3/08/2008			1.17	16.23	17	1.18		0.55			0.79
10/08/2008			1.5	18.54	22.15	1.65		0.7			0.54
17/08/2008			0.99	15.33	16.6	2.81		0.27			
24/08/2008			1.34	19.49	14.59			0.49			0.35
1/09/2008			1.5	21.68	16.79	4.15	0.38	0.51		0.52	0.21
7/09/2008			1.34	20.94	14.6	2.78		0.33			
28/09/2008			2.59	23.47	16.06	3.87		0.45			0.55
5/10/2008			0.36	3.76	13.48		0.34	0.04			0.5
12/10/2008			3.32	34.85	1.14	0.28	0.22	0.45			
19/10/2008			6.55	42.22	1.2		0.37	0.62			
26/10/2008			6.91	38.55	0.98		0.52	0.32			
2/11/2008			6.32	40.16	1.18			0.4			
9/11/2008			5.21	39.98	0.87			0.32			
16/11/2008			7.87	39.85				0.39			
23/11/2008			4.79	39.64	2			0.42			0.72
30/11/2008			4.74	39.3	9.38			0.46			
7/12/2008			12.2	44.14	10.01			0.59		0.47	
14/12/2008			16.29	42.99				0.45			
21/12/2008			14.55	33.12	4.58			0.32			
28/12/2008			12.1	26.95	0.34			0.09			
20/01/2009			16.68	36.01	4.13			0.24			0.21
25/01/2009			7.19	35.96	6.62			0.33			0.32
1/02/2009			12.75	33.44	5.75			0.26			0.39
8/02/2009			0.99	2.86	6.08						0.3
15/02/2009			12.54	40.86	6.76			0.24			0.52
22/02/2009			7.66	33.35	4.93			0.25			0.27
1/03/2009			7.03	25.09	2.94			0.24			0.2
8/03/2009			5.89	23.46	3.66			0.22			0.22
15/03/2009			11.49	25.06	2.25			0.15			0.19
26/04/2009			1.45	11.29	5.93			0.28			0.22
3/05/2009			2.69	21.72	0.49			0.34			

Date start	1,2,4,5-CIBz	1,2,3,4-CIBz	5CIBz	6CIBz	α -HCH	β -HCH	γ -HCH	8CISTY	OxCHLOR ^A	t-CHLOR	c-CHLOR
10/05/2009			2.47	26.41	0.67				0.46		
17/05/2009			1.44	14.03	0.55				0.49		
24/05/2009			0.95	12.25	1.15				0.66		
1/06/2009			1.44	12.08	8.74		0.55		0.87		
7/06/2009			1.44	14.92	1.01				0.77		
15/06/2009			1.48	18.71	0.8				0.5		
17/07/2009			0.98	11.14	4.65				0.3		
26/07/2009			0.9	9.69	4.53				0.83	0.35	0.5
17/08/2009			0.99	13.83	13.58		0.29		0.62		0.32
23/08/2009			1.3	18.83	16.78		2.01		0.61	0.32	0.57
30/08/2009			1.16	16.19	15.41		0.94		0.51		0.36
6/09/2009			1.35	19	15.19		1.13		0.62		0.45
14/09/2009			2	22.47	15.52		0.61		0.53	0.27	
20/09/2009			2.34	22.92	13.26				0.5		
27/09/2009			1.56	31.62	11.69		0.5		0.48		0.47
4/10/2009			2.47	30.18	9.63				0.5		0.37
11/10/2009			0.62	13.56	8.33		0.39		0.47	0.18	
12/08/2007	0.13				0.7					0.4	
19/08/2007	0.26				0.77					0.55	0.53
10/09/2007	0.37			0.26	0.88					0.37	3.88
16/09/2007	0.15									0.21	
23/09/2007	0.16										
30/09/2007	0.19			0.26	0.68						1.92
7/10/2007	0.21		0.61	0.25						0.16	2.24
14/10/2007	0.18									0.2	0.38
22/10/2007	0.3			0.4	0.96				0.69	0.85	3.38
28/10/2007	0.3				0.61					0.88	4.1
4/11/2007	0.24			0.33	0.61					0.43	3.68
11/11/2007	0.14									0.22	2.67
18/11/2007	0.19			0.28						0.27	3.16
25/11/2007	0.2			0.2						0.32	2.24
2/12/2007	0.14			0.19							1.99
9/12/2007	0.15			0.24	0.68						2.03
16/12/2007	0.14				0.69						1.51
23/12/2007	0.16				0.59						2.13
30/12/2007	0.15			0.26	0.61						1.43
6/01/2008	0.2			0.21							1.66
13/01/2008	0.15									0.58	1.7
20/01/2008	0.1									0.63	1.07
30/01/2008											

Date start	1,2,4,5-CIBz	1,2,3,4-CIBz	5CIBz	6CIBz	α -HCH	β -HCH	γ -HCH	8CISTY	OxCHLOR ^A	t-CHLOR	c-CHLOR
3/02/2008										0.15	0.48
10/02/2008	0.26			0.28	0.61					0.16	3.34
17/02/2008	0.22									0.17	4.66
24/02/2008	0.09									0.31	2.2
5/03/2008	0.33										3.39
9/03/2008	0.23			0.29							
16/03/2008				0.34							
23/03/2008	0.11			0.2	0.63					1.83	
30/03/2008	0.38			0.37						0.15	
6/04/2008	0.19									0.26	
13/04/2008	0.17		0.42	0.16	0.34			0.27		0.13	
28/04/2008											
4/05/2008				0.3							
11/05/2008				0.27							
18/05/2008				0.34							0.48
25/05/2008								0.6			
1/06/2008											
8/06/2008	0.31		3.72	0.44			1.76				2.95
15/06/2008											
22/06/2008	0.12										2.72
29/06/2008	0.34		2.58								10.17
6/07/2008	0.57			0.26							3.6
13/07/2008	0.26			0.32							
20/07/2008	0.68			0.26							3.14
27/07/2008	0.23			0.34							4.45
3/08/2008	0.27			0.26				0.51			4.68
10/08/2008	0.62			0.25							
17/08/2008	0.31			0.24							
24/08/2008	0.22		0.87	0.39							4.45
1/09/2008	0.17		0.83	0.39							3.6
7/09/2008	0.44			0.19							3.45
28/09/2008	0.15			0.45				0.47			7.24
5/10/2008	0.21			0.45							3.27
12/10/2008											
19/10/2008					0.78						0.39
26/10/2008											0.37
2/11/2008										3.88	
9/11/2008										3.64	0.65
16/11/2008											
23/11/2008	0.46		0.98	0.5	0.67						4.93

Date start	1,2,4,5-ClBz	1,2,3,4-ClBz	5ClBz	6ClBz	α -HCH	β -HCH	γ -HCH	8ClSTY	OxCHLOR ^A	t-CHLOR	c-CHLOR
30/11/2008	0.23		0.64	0.44	0.67						3.5
7/12/2008	0.28			0.37							2.77
14/12/2008											
21/12/2008				0.27						19.27	
28/12/2008										1.52	
20/01/2009											
25/01/2009				0.29							
1/02/2009				0.32							
8/02/2009				0.21							
15/02/2009											
22/02/2009				0.2						13.38	
1/03/2009											0.56
8/03/2009										11.87	0.64
15/03/2009										8.86	
26/04/2009											0.67
3/05/2009											
10/05/2009											
17/05/2009											
24/05/2009											
1/06/2009			1.39						0.83		2.19
7/06/2009											
15/06/2009											
17/07/2009		0.12									0.77
26/07/2009	0.33	0.36	1.21			1.01					7.01
17/08/2009	0.18	0.26		0.39							3.04
23/08/2009	0.42	0.2	0.98	0.38							2.96
30/08/2009	0.38										
6/09/2009											
14/09/2009	0.23	0.13	0.8	0.31							2.26
20/09/2009	0.23	0.08		0.38							2.34
27/09/2009	0.16	0.09		0.31							1.92
4/10/2009	0.17			0.27							1.85
11/10/2009				0.27							2.67

Table S5. (Cont.)

Date start	MeOx	3CIVER	4CIVER	ENDR ^A	TCB123	HCBD
12/08/2007			0.45			0.2
19/08/2007			0.52			0.2
10/09/2007			2.72			0.16
16/09/2007			0.46			0.29
23/09/2007			0.43			
30/09/2007			3.72			0.09
7/10/2007			2.34			
14/10/2007			0.41			0.19
22/10/2007			3.37			0.16
28/10/2007			3.38			0.25
4/11/2007			2.47			0.15
11/11/2007			1.57			0.11
18/11/2007			2.16			0.27
25/11/2007			1.75		1.28	1.63
2/12/2007			1.81			0.44
9/12/2007			2.55			1.2
16/12/2007			1.46			0.48
23/12/2007			1.55			0.59
30/12/2007			1.82			0.86
6/01/2008			1.7			0.82
13/01/2008			1.24			0.37
20/01/2008			1.2		1.45	1.49
30/01/2008			1.05			0.83
3/02/2008			1.15		1.87	1.41
10/02/2008			1.38			0.21
17/02/2008			1.89			0.5
24/02/2008			1.34			0.15
5/03/2008			2.05			0.36
9/03/2008			1.76			0.96
16/03/2008			2.33			0.55
23/03/2008			2.37			0.57
30/03/2008			1.36			0.4
6/04/2008			3.04	1.39		0.22
13/04/2008			1.59			0.18
28/04/2008			5.16			
4/05/2008			8.54			0.19
11/05/2008			4.47			0.23
18/05/2008			8.61			0.11
25/05/2008			4.52			0.16
1/06/2008			0.31	0.89		0.27
8/06/2008			5.86			0.09

Date start	MeOx	3CIVER	4CIVER	ENDR ^A	TCB123	HCBd
15/06/2008			0.78			0.11
22/06/2008			3.82			0.12
29/06/2008			6.86			0.21
6/07/2008			6.88			0.19
13/07/2008			4.97			0.23
20/07/2008	12.6		2.44			0.16
27/07/2008			6.46			0.19
3/08/2008			2.89			
10/08/2008			6			0.15
17/08/2008			4.86			
24/08/2008			5.17			0.1
1/09/2008			0.44			0.12
7/09/2008			3.55			
28/09/2008			8.54			0.33
5/10/2008			4.39			
12/10/2008			0.4			0.27
19/10/2008	4.5		0.52			0.69
26/10/2008			0.32			0.56
2/11/2008			0.36			0.8
9/11/2008			0.32			0.49
16/11/2008						0.39
23/11/2008			3.14			0.4
30/11/2008			3.01			0.27
7/12/2008			2.32			1.96
14/12/2008			0.28			1.63
21/12/2008			1.46			2.01
28/12/2008						6.85
20/01/2009			1.85			2.82
25/01/2009			1.76			0.33
1/02/2009			1.65			1.3
8/02/2009			1.41			
15/02/2009			1.76			0.6
22/02/2009			1.07			0.69
1/03/2009			0.8			0.73
8/03/2009			1.01			0.62
15/03/2009		0.69	0.7			1.2
26/04/2009			2.55			0.1
3/05/2009			0.5			0.15
10/05/2009			0.52			0.16
17/05/2009			0.6			0.09
24/05/2009			0.6			

Date start	MeOx	3CIVER	4CIVER	ENDR ^A	TCB123	HCBD
1/06/2009			4.98			0.14
7/06/2009			0.51			0.12
15/06/2009			0.99			0.16
17/07/2009			2.01			0.09
26/07/2009			4.92			0.11
17/08/2009			5.63			
23/08/2009			5.95			0.17
30/08/2009			5.37			0.12
6/09/2009			3.75			0.1
14/09/2009			4.56			0.21
20/09/2009			3.72			0.24
27/09/2009			4.79			0.18
4/10/2009			3.09			0.22
11/10/2009			3.19			

^AMeets a chromatographic acceptance criterion in very few samples

Table S6. Concentrations (pg m^{-3}) in each day-long sample of the SVOCs that were above the method detection limits

Date start	1,2,4,5-CIBz	1,2,3,4-CIBz	5CIBz	6CIBz	α -HCH	β -HCH	γ -HCH	8CISTY	OxCHLOR	t-CHLOR	c-CHLOR
1/01/2007	0.62	0.62	0.27	0.47	0.07	0.11	0.05	0.03	0.15	0.11	0.1
25/08/2007			8.04	52.01	14.31	2.31	1.42	0.81			1.02
26/08/2007			9.09	57.44	13.54	4.02				1.16	0.81
27/08/2007			9.15	56.54	21.94		1.54	0.47			1.02
28/08/2007			9.76	62.08	15.51		1.19	0.76			1.15
29/08/2007		4.87	6.91	45.93	13.75		1.91	0.83			1.34
30/08/2007	7.38	9.28	8.89	47.38	9.98		1.88				1.1
31/08/2007			10.24	46.18	16.97		1.45				
1/09/2007		4.84	7.82	42.49	15.25		2.09	0.4			
2/09/2007		4.32	8.9	46.54	11.04		1.08	0.41		1.05	0.79
3/09/2007		5.51	9.59	47.87	17.31		1.94	0.34			
4/09/2007		5.1	10.1	47.78	16.62		1.13	0.43			
5/09/2007		5.46	10.54	48.35	13.7		1.31	0.41			
6/09/2007		4.02	9.39	45.9	16.39		1.16	0.39			0.68
7/09/2007			5.54	39.65	15.11		1.9	0.36			
14/09/2008			8.85	55.21	14.93			0.62			1.31
15/09/2008			7.33	50.59	16.51	1.72					
16/09/2008			5.93		15.36			0.38	2.1		
17/09/2008			7.55	45.39	19.25			0.6			
18/09/2008			10.57	57.28	17.56	5.86	0.82				2.18
19/09/2008			8.46	54.02	15.47		0.81	0.25			1.24
20/09/2008			13.9		17.51		0.91		1.46		
21/09/2008			14.65	58.5	19.89			0.58			
22/09/2008			12.3	49.58	19.21			0.49			
23/09/2008			12.81	58.9	23.62	3.59		0.61			
24/09/2008			15.84	56.09	17.08			0.63		1.9	
25/09/2008			13.17	51.63	12.99	2.08		0.52			
26/09/2008			14.23	52.93	14.17			0.51			1.24
27/09/2008			12.77	53.04	14.66	6.54					1.21

Date start	t-NONA	c-NONA	HEPOx	DIEL	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>p,p'</i> -DDD	5CIANI	α -ENDO	4CIVER	ENDR
1/01/2007	0.07	0.05	0.15	0.12	0.1	0.42	0.75	0.11	0.07	0.17	0.18
25/08/2007			4.87			3.11		1.44	6.64	9.33	
26/08/2007	0.61		3.97			4.15		1.05	6.03	7.08	
27/08/2007	0.47		4.4		0.74	5.28		1.43	7.61	9.09	
28/08/2007			4.41			5.67		1.87	8.92	6.99	
29/08/2007			2.95					1.27	7.36	7.7	
30/08/2007			4.47			2.91		2.05	7.41	9.43	
31/08/2007						3.41			5.39	6.03	
1/09/2007					0.71				4.63	5.62	
2/09/2007	0.64				0.94				5.18	5.66	
3/09/2007	0.57				0.88	3.17			5.34	7.11	
4/09/2007			1.2						5.07	7.42	
5/09/2007					0.98	4.14			3.21	5.28	
6/09/2007									4.02	5.84	
7/09/2007			1.17		0.85	4.67			3.66	4.89	
14/09/2008			1.31						4.2	5.29	
15/09/2008	0.68								4.93	4.53	
16/09/2008	1.53									3.07	
17/09/2008	1.64								2.49	4.54	2.32
18/09/2008	0.88		1.33						6.02	4.85	
19/09/2008	1.58								6.51	5.79	1.45
20/09/2008									4.19	4.56	1.79
21/09/2008	1.72								3.65		
22/09/2008	1.12								1.85	5.91	
23/09/2008	1.15			1.34		2.77			4.06	7.04	
24/09/2008									3.56	5.53	1.31
25/09/2008	1.45						12.82		2.62	3.14	
26/09/2008	1.27						8.27				
27/09/2008											

Table S6. (Cont.)

Date start	1,2-CIBz	1,3,5-CIBz	1,2,4-CIBz	6CIBD	β-ENDO
1/01/2007	2.46	0.19	10.4	0.05	0.29
25/08/2007				1.33	
26/08/2007				1.04	
27/08/2007				1.19	2.34
28/08/2007				1.04	2.53
29/08/2007				0.81	2.41
30/08/2007	22.86		73.47	0.95	
31/08/2007		1.98		0.75	
1/09/2007				0.73	
2/09/2007				0.87	
3/09/2007				1.73	
4/09/2007				1.32	
5/09/2007				1.23	
6/09/2007				0.87	
7/09/2007					
14/09/2008				0.45	
15/09/2008				0.99	
16/09/2008				0.54	
17/09/2008				0.83	
18/09/2008				0.85	
19/09/2008				0.46	
20/09/2008				1.77	
21/09/2008				1.61	
22/09/2008				1.01	
23/09/2008				1.15	
24/09/2008				2.41	
25/09/2008				0.74	
26/09/2008				2.05	
27/09/2008				0.94	

Table S7. Concentrations (pg m⁻³) in each week-long sample of the brominated flame retardants that were above the method detection limits

Date start	BDE17	BDE28/33	BDE47	BDE66	BDE100	BDE99	BDE85	BDE154	BDE153	HBCD	BDE138	BDE183	BDE190	BDE49
12/08/2007	0.2	0.34	21.02	0.37	4.39	11.58	0.43	0.39	0.32		0.05			0.75
19/08/2007	0.12	0.18	10.03	0.16	2	5.38	0.26	0.24	0.26			0.05		0.36
10/09/2007	0.09	0.13	4.75	0.15	0.96	3.41	0.16	0.13	0.12			0.08		
16/09/2007	0.05	0.07	2.88	0.09	0.52	1.61		0.06				0.04		
23/09/2007	0.06	0.08	3.16	0.11	0.57	1.6		0.07	0.08			0.04		
30/09/2007	0.05	0.07	2.56	0.1	0.53	1.96	0.11	0.12	0.15			0.04		
7/10/2007		0.04	0.91		0.2	0.61								
14/10/2007		0.04	1.05	0.04	0.23	0.7		0.03						
22/10/2007		0.05	1.21		0.29	1.2		0.07	0.08			0.05		
28/10/2007		0.04	1.11	0.05	0.27	1.34	0.09	0.11	0.14			0.04		
4/11/2007			0.66		0.17	0.82		0.07	0.08					
11/11/2007			0.36		0.11	0.52		0.05	0.07					
18/11/2007			0.56		0.15	0.66		0.04						
25/11/2007			0.48		0.12	0.63		0.05	0.07			0.04		
2/12/2007			0.73		0.23	1.34	0.1	0.15	0.2			0.04		
9/12/2007		0.03	0.66		0.14	0.52		0.04	0.06					
16/12/2007			0.38		0.09	0.34								
23/12/2007		0.05	0.69		0.12	0.51		0.05	0.06					
30/12/2007		0.04	0.78		0.18	0.8		0.05						
6/01/2008		0.05	0.73		0.12	0.51		0.04						
13/01/2008		0.04	0.69		0.16	0.76		0.05	0.07					
20/01/2008		0.03	0.4		0.08	0.39		0.03				0.02		
30/01/2008		0.07	0.78		0.14	0.63								
3/02/2008		0.04	0.4		0.06	0.34								
10/02/2008		0.05	0.86		0.17	0.77		0.05	0.07			0.06		
17/02/2008		0.05	0.75		0.26	1.4	0.12	0.14	0.21		0.05			
24/02/2008		0.03	0.43		0.09	0.29								
5/03/2008			0.65		0.16	0.44								
9/03/2008			0.48		0.11	0.51		0.04				0.04		

Date start	BDE17	BDE28/33	BDE47	BDE66	BDE100	BDE99	BDE85	BDE154	BDE153	HBCD	BDE138	BDE183	BDE190	BDE49
16/03/2008			0.46		0.11	0.34								
23/03/2008			0.76		0.15	0.73		0.05						
30/03/2008			0.7		0.15	0.72		0.05				0.04		
6/04/2008														
13/04/2008		0.03	0.09		0.03	0.13		0.04	0.03					
28/04/2008		0.05	1.42		0.3	1.14		0.08	0.07					
4/05/2008		0.05	1.21	0.05	0.26	1.12		0.06	0.06			0.06		
11/05/2008		0.04	1.31	0.05	0.28	1		0.07	0.07					
18/05/2008		0.06	1.88	0.07	0.47	2.02	0.12	0.1	0.1					
25/05/2008		0.07	0.98					0.04	0.06					
1/06/2008		0.04				0.27		0.06	0.09	3.79				0.22
8/06/2008	0.05	0.08	2.78	0.1	0.66	2.8	0.14	0.16	0.2	11.81		0.41		0.23
15/06/2008	0.06	0.11			0.09	0.36	0.22	0.26	0.07					0.24
22/06/2008		0.06	2.3	0.09	0.59	2.39	0.13	0.14	0.12	5.85		0.05		0.28
29/06/2008	0.05	0.09	3.37	0.12	0.89	3.64	0.17	0.22	0.2			0.04		
6/07/2008		0.06	1.91		0.46	2.04	0.13	0.12	0.11		0.36			
13/07/2008		0.07	1.93	0.08	0.48	2.27	0.13	0.12	0.13					
20/07/2008		0.04	1.14		0.31	1.34		0.08	0.09					
27/07/2008		0.07	2.33	0.09	0.57	2.67	0.15	0.15	0.15					
3/08/2008		0.06	1.71	0.07	0.45	2.16	0.13	0.13	0.12					
10/08/2008		0.06	1.4			1.99	0.13	0.12	0.11					
17/08/2008		0.07	1.95	0.07	0.41	1.85	0.12	0.11	0.13					
24/08/2008		0.06	1.71	0.09	0.44	1.68	0.1	0.09	0.07					
1/09/2008		0.04	0.89		0.24	0.96		0.07	0.07					
7/09/2008		0.04	0.94		0.23	1.09		0.08	0.08					
28/09/2008		0.06	1.74	0.09	0.45	1.73		0.08	0.07					
5/10/2008			0.36		0.08	0.41								
12/10/2008			0.66		0.19	0.96		0.09	0.09					
19/10/2008			0.63		0.15	0.68		0.04						
26/10/2008			0.78	0.04	0.18	0.74								

Date start	BDE17	BDE28/33	BDE47	BDE66	BDE100	BDE99	BDE85	BDE154	BDE153	HBCD	BDE138	BDE183	BDE190	BDE49
2/11/2008			0.48											
9/11/2008			0.38		0.19	1.2	0.1	0.12	0.19					
16/11/2008			0.39											
23/11/2008			0.62		0.2	0.72		0.05	0.07				0.05	
30/11/2008			0.41		0.09	0.29								
7/12/2008			0.43		0.11	0.57		0.05	0.07					
14/12/2008														
21/12/2008			0.38		0.11	0.47		0.04	0.05					
28/12/2008		0.03	0.61		0.14	0.76		0.06	0.15		0.06			
20/01/2009		0.05	0.61		0.1	0.48							0.06	
25/01/2009			0.43		0.1	0.47								
1/02/2009			0.62		0.14	0.7								
8/02/2009			0.24			0.23								
15/02/2009			0.76											
22/02/2009			0.37		0.09	0.44								
1/03/2009			0.35		0.09	0.42		0.04	0.05					
8/03/2009			0.41		0.1	0.48		0.04						
15/03/2009			0.44		0.12	0.54			0.15				0.48	
26/04/2009		0.06	0.88	0.06	0.23	0.95		0.06						
3/05/2009			0.55		0.14	0.62		0.04					0.04	
10/05/2009		0.04	0.71	0.05	0.16	0.66								
17/05/2009		0.04	0.56		0.15	0.66		0.04						
24/05/2009		0.05	1.09	0.06	0.28	1.36	0.09	0.08	0.07					
1/06/2009	0.08	0.16	3.58	0.14	0.9	4.44	0.22	0.29	0.25					
7/06/2009		0.07	1.61	0.07	0.38	1.9	0.11	0.13	0.13					
15/06/2009		0.07	1.48	0.08	0.42	2.09	0.13	0.14	0.15					
17/07/2009		0.04	0.73	0.04	0.19	0.94		0.07	0.06					
26/07/2009	0.07	0.12	3.04	0.12	0.84	4	0.18	0.31	0.33					
2/08/2009			0.59		0.16	0.8		0.07	0.07					
17/08/2009	0.08	0.18	3.75	0.22	0.92	3.2	0.13	0.17	0.13					0.35

Date start	BDE17	BDE28/33	BDE47	BDE66	BDE100	BDE99	BDE85	BDE154	BDE153	HBCD	BDE138	BDE183	BDE190	BDE49
23/08/2009		0.05	1.47	0.09	0.51	2.09	0.1	0.11	0.09					
30/08/2009		0.06	1.1	0.07	0.35	1.52		0.09	0.08					
6/09/2009		0.04	0.85	0.05	0.26	1.28	0.08	0.07	0.07					
14/09/2009		0.05	1.58	0.09	0.73	4.22	0.27	0.39	0.51		0.09	0.08		
20/09/2009		0.04	0.73	0.06	0.24	1.05		0.06	0.06					
27/09/2009		0.04	0.77	0.06	0.22	0.96		0.06	0.06					
4/10/2009		0.05	0.94	0.07	0.3	1.22		0.07						
11/10/2009		0.04	0.58	0.05	0.18	0.76		0.04						

Table S8. Concentrations (pg m⁻³) in each day-long sample of the brominated flame retardants that were above the method detection limits

Date start	BDE17	BDE28/33	BDE47	BDE66	BDE100	BDE99	BDE85	BDE154	BDE153	HBCD	BDE138	BDE183	BDE190	BDE49
1/01/2007	0.32	0.25	1.38	0.31	0.45	1.66	0.67	0.26	0.41	22.45	0.36	0.27	0.5	1.21
25/08/2007		0.38	11.29	0.34	2.56	8.66		0.66	0.77					
26/08/2007		0.42	9.06		1.81	5.69		0.31	0.48			0.29		
27/08/2007		0.27	6.08		1.26	4.16		0.32						
28/08/2007		0.32	7.46		1.38	4.11		0.29						
29/08/2007		0.27	9.16		1.64	5.7		0.28						
30/08/2007		0.31	14.62	0.38	2.1	7.24		0.33				0.27		
31/08/2007			9.4		1.78	6.33		0.34				0.52		
1/09/2007			6.02		1.24	4.42		0.27						
2/09/2007			5.65		1.16	4.61								
3/09/2007			6.93		1.38	5.55		0.34	0.41					
4/09/2007			8.38		1.73	5.7		0.33	0.41			0.33		
5/09/2007		0.25	9.45	0.38	1.9	8.48		0.53	0.71					
6/09/2007			5.98		1.56	7.12		0.6	0.74					
7/09/2007		0.27	9.91	0.35	2.02	9.69	0.73	0.53	0.74			0.33		
14/09/2008			3.2		0.7	2.35								
15/09/2008			2.07		0.58	2.23								
16/09/2008			9.43				0.71	0.61	0.78					
17/09/2008			8.99		1.24	4.58								
18/09/2008			3.1		0.89	3.1								
19/09/2008			3.07		0.9	3.04								
20/09/2008			1.94		0.56	1.89								
21/09/2008			3.36		0.79	2.87								
22/09/2008			4.85		0.96	4.31		0.31	0.45					
23/09/2008			7.58		1.55	7.13		0.45	0.55					
24/09/2008			5.11		1.1	5.53		0.35	0.55					
25/09/2008			8.33			7.46		0.37	0.55					
26/09/2008			6.84		1.2	5.74		0.3						
27/09/2008			2.05		0.57	2.18								

Table S9. Concentrations (pg m⁻³) in each week-long sample of the polycyclic aromatic hydrocarbons that were above the method detection limits

Date start	FLUR	PHEN	ANTH	FLUT	PYRE	RETE	BAAN	CHRY	BBFL	BKFL	BEPY
12/08/2007	17.76	154.27	8.87	23.96		27.31		2.04	0.83	0.53	
19/08/2007	14.63	105.6					2.39	1.9	0.82		
10/09/2007	19.58	137.92	6.93	24.44		35.43	1.48	3.96	2.02	1.88	1.23
16/09/2007	13.51	66.56				23.13	1.35	1.29	0.75	0.45	
23/09/2007	10.36	74.11				30.59	1.54	1.51	0.82	0.55	
30/09/2007	23.85	129.59	9.59	29.76		152.62	2.16	5.6	2.7	1.92	1.54
7/10/2007	18.54	64.18				38.45	0.74	1.95	1.05	0.82	0.6
14/10/2007	45.28	131.04	4.45	25.56		118.37	2.06	6.25	4.79	2.86	2.33
22/10/2007	47.83	108.32		24.22		90.77	2.34	6.22	4.03	3.62	2.41
28/10/2007	39.72	94.25	4	23.23		78.13	4.68	6.91	4.02	3.91	2.7
4/11/2007	94.62	196.19	4.64	46.17		157.68	4.92	11.41	7.26	6.29	3.93
11/11/2007	45.33	104.94		27.22		113.55	3.38	9.48	7	6.49	4.66
18/11/2007	57.59	126.15	5.63	27.33		130.33	2.42	9.72	4.75	4.4	3.61
25/11/2007	98.83	146.21		37.58		99.28	3	10.87	7.34	6.17	3.96
2/12/2007	162.57	268.08	15.08	98.99	68.39	223.18	12.8	34.96	26.48	22.57	15.08
9/12/2007	128.06	209.27		54.27		127	3.24	12.65	6.68	6.97	3.99
16/12/2007	138.96	209.17		59.15		120.66	3.7	13.44	9.16	7.87	4.85
23/12/2007	135.65	239.76		73.12		139.79	5.58	14.4	8.42	8	5.07
30/12/2007	135.37	218.04		74.57		163.66	6	18.63	13.08	7.9	5.81
6/01/2008	173.52	300.28	4.45	95.55		204.59	6.23	20.3	10.52	8.17	5.3
13/01/2008	132.7	191.87		70.79		159.44	5.77	21.2	13.25	11.45	6.43
20/01/2008	183.21	192.8	4.98	105.32	68.06	150.66	48.9	42.89	45.83	27.41	21.43
30/01/2008	147.52	141.97		67.08		61.49	5.57	19.93	14.05	11.36	8.3
3/02/2008	186.36	97.28		50.63		28.36	6.16	19.77	20.3	14.03	10.04
10/02/2008	132.76	202.23		56.62		103.99	3.32	11.33	7.16	4.72	3.32
17/02/2008	69.5	147.82		45.45		85.81	3.26	9.2	6.32	4.33	3.22
24/02/2008	65.38	111.01		35.87		51.55	1.76	8.93	5.59	4.43	2.26
5/03/2008	32.98	95		31.17		46.02	1.46	4.38	2.7	2.01	1.28
9/03/2008	59.27	57.1		17.04		43.94	1.62	4.08	4.09	1.48	2

Date start	FLUR	PHEN	ANTH	FLUT	PYRE	RETE	BAAN	CHRY	BBFL	BKFL	BEPY
16/03/2008	59.67	63.42		19.8		36.87	1.07	4.59			
23/03/2008	43.08	49.59		16.33		29.5	1.29	3.2	3.02	0.94	2.04
30/03/2008	23.08	46.53		14.45		22.32	0.9	2.53	0.39		
6/04/2008	36.2	50		14.33		48.56	1.41	2.72	1.96	0.79	1.48
13/04/2008	14.77	47.84	2.86	13.57		14.44	1.12	2.14	1.97	0.8	0.66
28/04/2008	26.11					29.41	1.07	2.42	0.48		
4/05/2008	26.15	61.92				43.97	1.08	2.66	1.82	0.88	1.54
11/05/2008	18.91	53.24				26.84	1.01	1.51	0.47		
18/05/2008	15.53	74.99				29.89	0.65	1.37	1.02		1
25/05/2008	8.02	72.52					2.16	1.36	0.95	0.41	
1/06/2008	11.55	63.79				91.71	0.62	1.26	0.45		
8/06/2008	9.96	102.79					0.55	0.89	0.62		0.64
15/06/2008	11.01	105.63		16.14		42.95	3.96		0.4		
22/06/2008		78.12							0.49		
29/06/2008	18	144.59	4.49	23.97		41.59	4.26	1.83	1.16	0.6	
6/07/2008	10.38	66.43					0.63				
13/07/2008	10.39	74.84					0.98		0.4		0.86
20/07/2008	12.23	54.42					1.28		1.14		2.37
27/07/2008		81.17					0.6		0.42		1.91
3/08/2008	6.76	73.7									
10/08/2008	8.99	60.88		17.42			0.82		0.47		
17/08/2008	10.01	109.68				30.02	0.98	1.51			0.69
24/08/2008	9.08	81.06							0.38		
1/09/2008	14.89	102.64				46.01	1.52	1.72	0.83		0.8
7/09/2008	7.47	67.94					1.12		0.45		1
28/09/2008	24.8	93.93		15.86		42	1.01	1.5	0.55		0.54
5/10/2008	47.95	116.46		25.7		81.57	1.69	3.3	1.07	0.63	1.19
12/10/2008	56.63	113.93		24.82		118.01	1.97	2.73	0.95	0.55	0.72
19/10/2008	339.72	687.29	88.29	171.1	141.31	816.35	21.12	53.32	6.22	7.12	19.99
26/10/2008	75.79	148.04		32.62		98.44	2.52	5.08	1.31	0.66	2.07

Date start	FLUR	PHEN	ANTH	FLUT	PYRE	RETE	BAAN	CHRY	BBFL	BKFL	BEPY
2/11/2008	85.44	190.08		42.37		173.15	1.18	8.96	1.85	0.8	1.44
9/11/2008	66.75	148		33.87		112.34	1.44	4.92	2.51	1	1.38
16/11/2008	99.46	204.77		49.91		155.41	1.78	8.53	3.72	1.25	2.52
23/11/2008	89.58	186.43		54.88		144.91	3.31	8.92	2.84	2.43	3.99
30/11/2008	154.97	335.48	4.16	95.63	71.65	254.4	2.66	17.09	5.38	2.08	4.32
7/12/2008	160.74	124.7		46.78		45.98	4.39	12.68	6.76	4.55	7.46
14/12/2008	243.57	180.8		73.96		82.74	8.97	19.14	30.39	6.98	12.95
21/12/2008	185.87	234.98	5.13	91.22		173.7	11.73	19.86	19.49	7.64	12.27
28/12/2008	140.51	68.25		35.26		18	2.59	9.55	14.7	3.56	6.7
20/01/2009	210.09	213.76	5.24	109.18	90.34	183.37	15.46	31.36	41.26	12.62	19.16
25/01/2009	145.59	196.43		76.98		152.19	5.16	13.77	12.9	3.43	5.9
1/02/2009	127.76	143.76		39.8		52.83	1.63	5.2	5.12	0.9	2.63
8/02/2009	137.33	175.18		73.07	82.04	133.09	4.27	15.67	11.94	3.47	5.65
15/02/2009	180.96	398.04		127.6		213.1	5.72	19.6	16.85	4.39	6.63
22/02/2009	87.32	137.86		60.75	77.13	132.2	4.17	12.18	3.71	2.99	5.14
1/03/2009	55.27	91.7		33.48		72.37	2.45	6.72	2.6	2.11	3.48
8/03/2009	65.08	114.83		42.17		112.51	2.63	7.69	2.96	2.33	3.84
15/03/2009	48.58	96.48		32.38		74.83	2.48	6.88	3.08	2.62	4.34
26/04/2009	10.21	85		15.7			0.5	1.89	1.09	0.53	1.21
3/05/2009	10.07	60.76						1.59	0.42	0.46	
10/05/2009	11.9	52.41							0.46		
17/05/2009	9.84	77.71		22.95			1.07	2.27	2.62	0.75	1.81
24/05/2009	7.22	79.4							0.4		
1/06/2009	245.43	550.97		95.08		678.89	4.13	22.63	18.03	3.16	9.23
7/06/2009	12.78	296.38		43.36		393.94	0.58	6.34	3.93	0.39	1.6
15/06/2009	12.23	120.15						1.46			
17/07/2009	10.62	56.41									
26/07/2009	1068.24	1794.28	79.54	226.16	144.75	6156.94	19.81	73.44	21.37	7.05	3.62
17/08/2009	37.74	238.87		24.98		217.27	1.33	1.53	0.62		
23/08/2009	15.77	132.79		19.71		83.79	1.07	1.54	0.63	0.4	

Date start	FLUR	PHEN	ANTH	FLUT	PYRE	RETE	BAAN	CHRY	BBFL	BKFL	BEPY
30/08/2009	16.26	124.61		16.61		63.91	1.23	1.11	0.64		
6/09/2009	15.63	138.81	3.69	19.61		68.64	1.06	2.08	0.92	0.55	1.07
14/09/2009	18.73	124.28		17.74		55.86	1.07	1.82	0.91	0.51	1.02
20/09/2009	16.3	103.46		20.51	70.5	49.76	1.28	2.04	0.78	0.44	
27/09/2009	42.87	117.23		22.98		81.39	1.77	3.04	1.58	0.83	1.34
4/10/2009	49.41	139.43		28.71		97.85	1.27	3.54	1.27	0.46	1.44
11/10/2009	39.9	194.81		47.89		260.3	3.78	10.43	4.72	1.78	3.86

Table S10. Concentrations (pg m^{-3}) in each day-long sample of the polycyclic aromatic hydrocarbons that were above the method detection limits

Date start	FLUR	PHEN	ANTH	FLUT	PYRE	RETE	BAAN	CHRY	BBFL	BKFL	BEPY
1/01/2007	6.71	30.01	0.53	6.15	13.58	21.08	0.33	0.98	0.42	0.43	0.21
25/08/2007	49.04	242.41	16.92	48.67	22.38	44.35	2.05	6.98	3.13	2.07	1.97
26/08/2007	70.33	328.58	22.2	58.99	59.17	79.33	2.6	9.75	4.08	2.68	1.69
27/08/2007	61.22	295.26	16.73	51.31	40.73	57.91	3.1	6.46	3.61	2.85	2.38
28/08/2007	49.83	345.45	19.59	56.21	36.38	46.17	2.22	5.34	3.09	2.08	1.68
29/08/2007	50.72	351.11	23.97	54.41	39.8	43.08	1.78	4.75	2.53	1.61	0.92
30/08/2007	76.2	438.44	45.85	73.63	75.83	135	1.85	5.95	3.43	1.75	1.26
31/08/2007	54.39	319.8	17.68	54.76	58.41	91.46	4.39	7.16	1.92	2.89	3.16
1/09/2007	39.57	280.14	25.57	37.05	60.61	52.66	2.93	4.53	2.2	1.56	0.94
2/09/2007	80.99	421.72	27.98	57	71.31	66.8	2.11	6.37	2.91	2.01	1.39
3/09/2007	82.45	301.94	17.14	47.86	43.75	54.58	3.15	5.96	4.17	2.1	1.43
4/09/2007	59.52	218.26	16.18	39.08	37.55	77.05	2.3	5.86	3.56	2.86	1.8
5/09/2007	39.57	219.49	12.03	34.92	38.97	34.1	2.4	4.46	2.04	1.32	0.95
6/09/2007	62.21	277.48	22.89	42.82	48.96	37.2	2.65	5.79	2.58	1.67	1.23
7/09/2007	49.16	224.76	13.61	39.14	36.07	48.66	2.92	5.83	3.2	2.44	1.51
14/09/2008	37.63	208.27	16.61	115.97	154.82	233.14	8.65	10.22	1.15	1.03	4.63
15/09/2008	42.5	209.73	15.96	56.7	173.5	48.55	10.93	2.66	1.11	0.99	1.99
16/09/2008	58.37	202.97	14.88	55.34	214.62	132.73	8.64	6.31	1.48	1.03	0.95
17/09/2008	40.97	172.12	9.53	25.69	153.55	36.88	3.92	2	1.81	1.01	1.55
18/09/2008	51.08	189.61	4.3	42.88	162.6	72.37	3.18	2.11	2.04	0.97	1.28
19/09/2008	45.65	224.39	15.73	39.3	254.82	71.31	2.31	4.16	1.09	0.98	2.43
20/09/2008	44.85	208.06	3.91	23.26	183.21	34	1.37	2.87	1.09	0.97	0.9
21/09/2008	37.6	176.13	14.47	27.05	252.38	36.46	6.46	2.77	1.4	1	0.92

Table S11. Concentrations (pg m^{-3}) of brominated flame retardants measured in week-long samples

Compound	Short Name	Fraction of detections	Mean	Med	Min	Max
2,2',4-tribromodiphenyl ether	BDE17	0.12	0.08	0.06	0.05	0.20
2,4,4'-tribromodiphenyl ether and 2,3,4'-tribromodiphenyl ether	BDE28+33	0.65	0.06	0.05	0.03	0.34
2,2',4,4'-tetrabromodiphenyl ether	BDE47	0.96	1.42	0.76	0.09	21.02
2,3',4,4'-tetrabromodiphenyl ether	BDE66	0.39	0.09	0.08	0.04	0.37
2,2',4,4',6-pentabromodiphenyl ether	BDE100	0.91	0.35	0.19	0.03	4.39
2,2',4,4',5-pentabromodiphenyl ether	BDE99	0.94	1.35	0.82	0.13	11.58
2,2',3,4,4'-pentabromodiphenyl ether	BDE85	0.30	0.15	0.13	0.08	0.43
2,2',4,4',5,6'-hexabromodiphenyl ether	BDE154	0.77	0.10	0.07	0.03	0.39
2,2',4,4',5,5'-hexabromodiphenyl ether	BDE153	0.62	0.12	0.09	0.03	0.51
hexabromocyclododecane	HBCD	0.03	7.15	5.85	3.79	11.81
2,2',3,4,4',5'-hexabromodiphenyl ether	BDE138	0.05	0.12	0.06	0.05	0.36
2,2',3,4,4',5',6-heptabromodiphenyl ether	BDE183	0.23	0.08	0.04	0.02	0.48
2,3,3',4,4',5,6-heptabromodiphenyl ether	BDE190					
decabromodiphenyl ether	BDE209	0.86	1.03	0.74	0.28	4.84
2,2',4,5'-tetrabromodiphenyl ether	BDE49	0.07	0.35	0.28	0.22	0.75

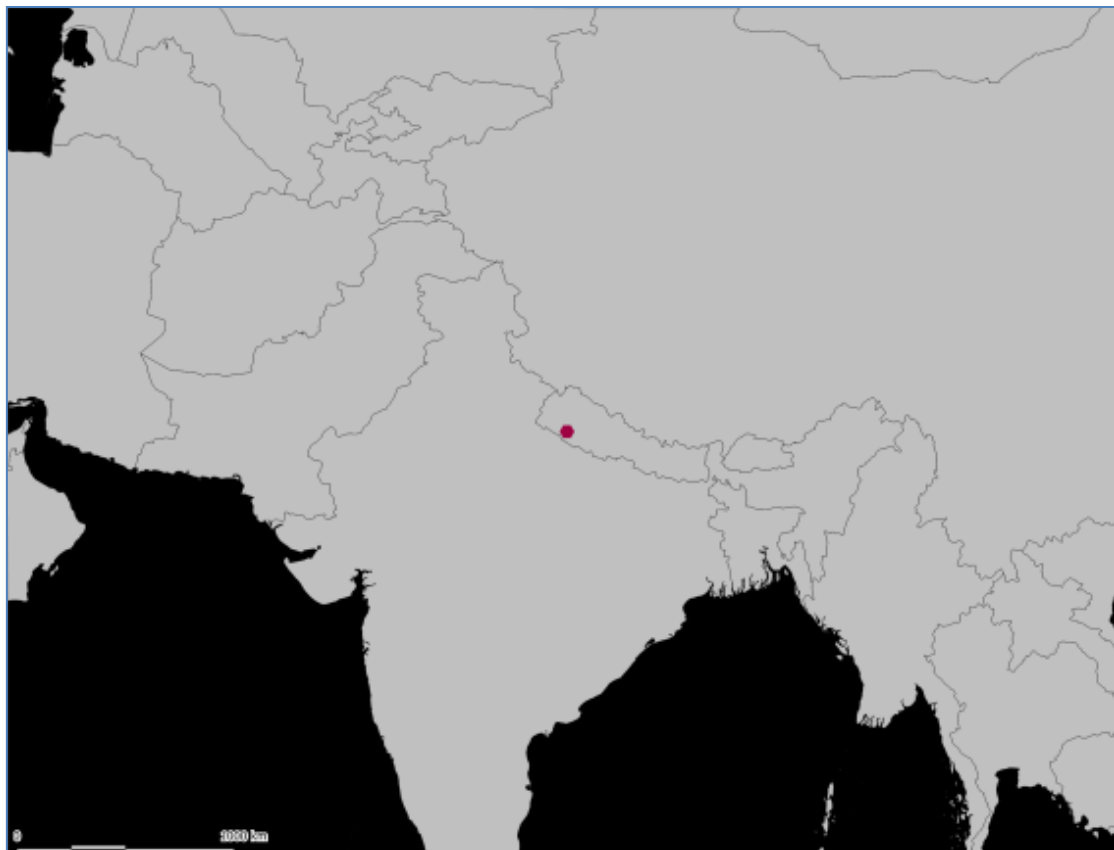


Fig. S2. A single cell in Nepal was highlighted by the technique as being the strongest source of α -endosulfan to Little Fox Lake from the set. The same cell was attributed high CWT values for most compounds.

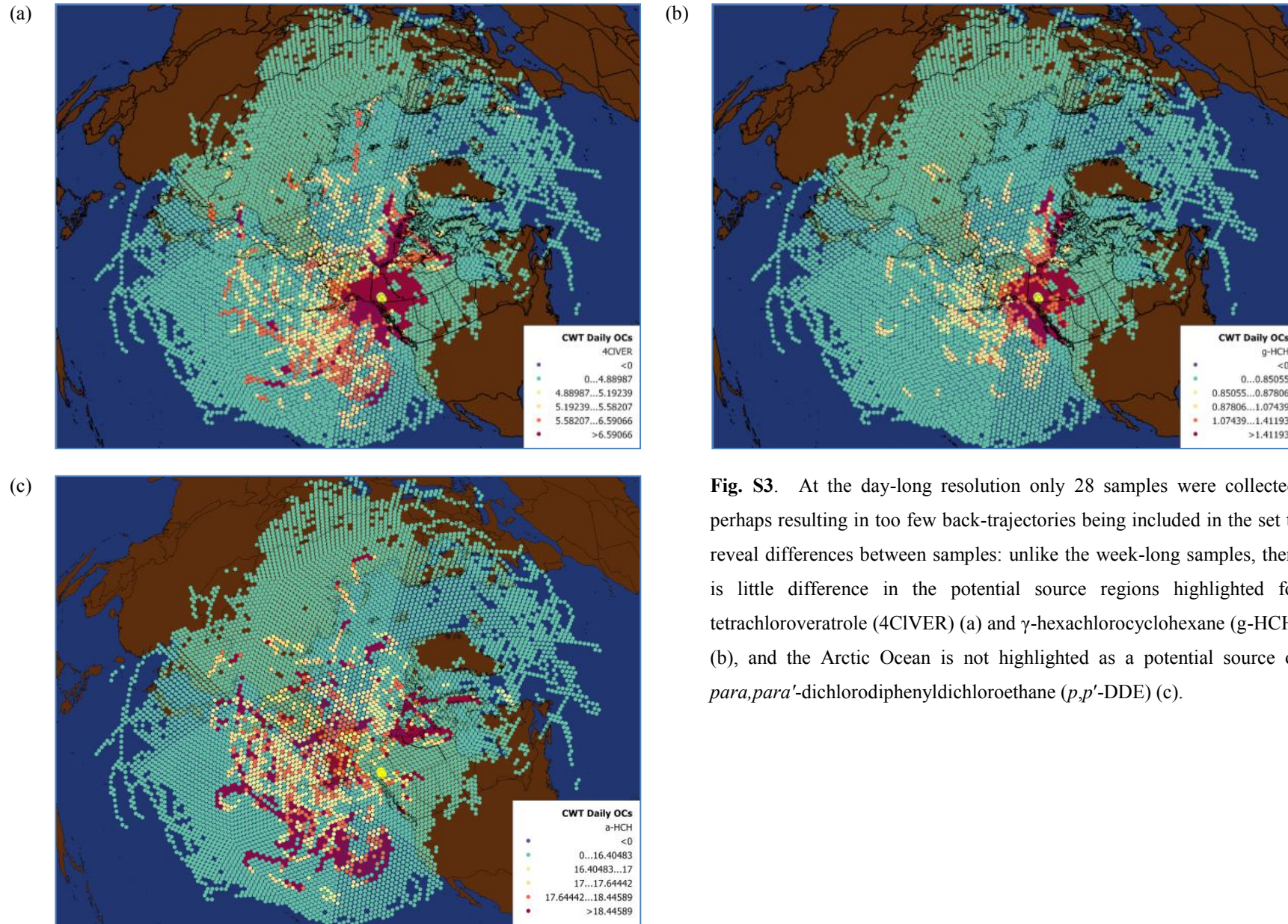


Fig. S3. At the day-long resolution only 28 samples were collected, perhaps resulting in too few back-trajectories being included in the set to reveal differences between samples: unlike the week-long samples, there is little difference in the potential source regions highlighted for tetrachlorooveratrole (4CIVER) (a) and γ -hexachlorocyclohexane (g-HCH) (b), and the Arctic Ocean is not highlighted as a potential source of *para,para'*-dichlorodiphenyldichloroethane (*p,p'*-DDE) (c).

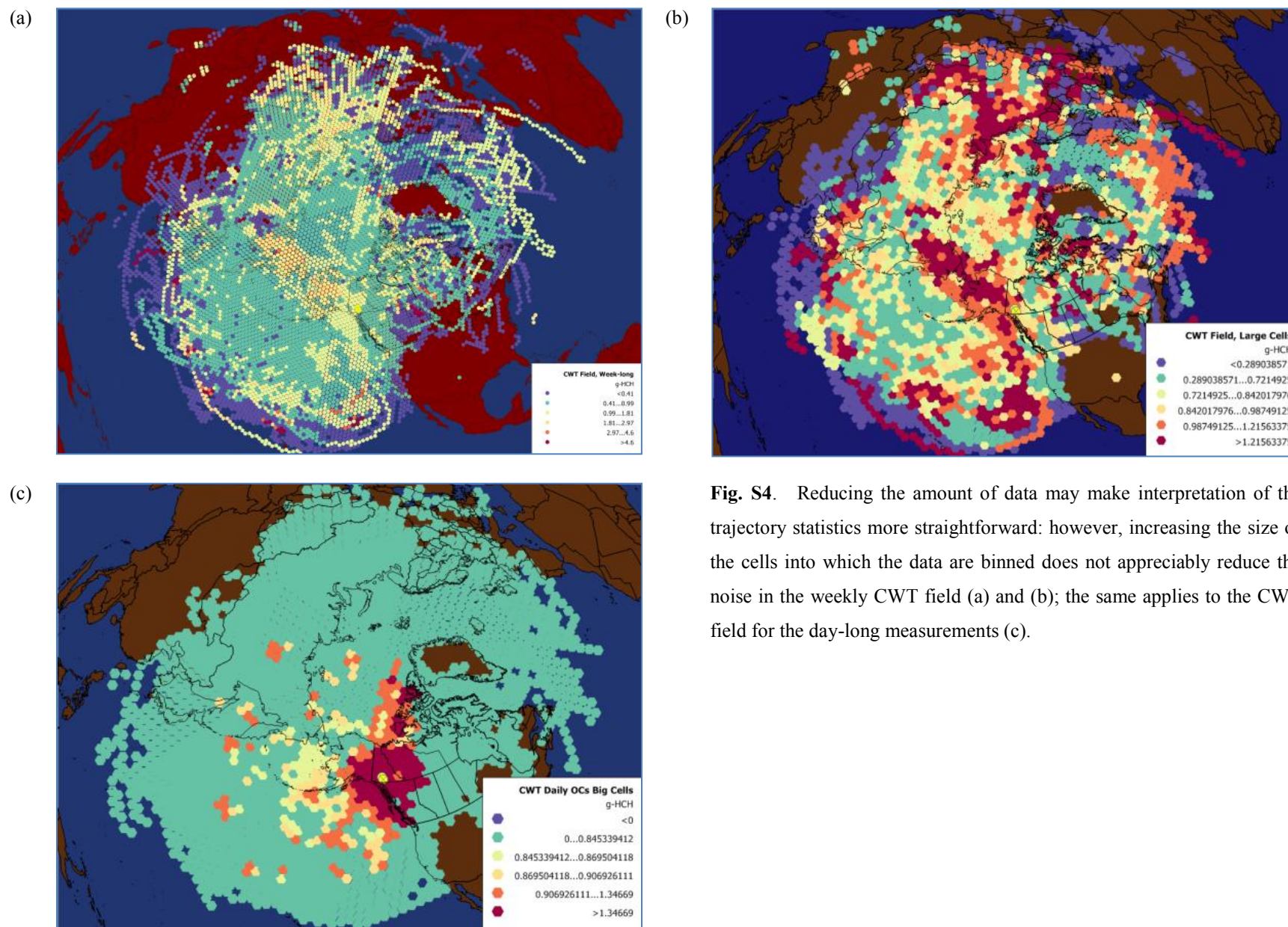
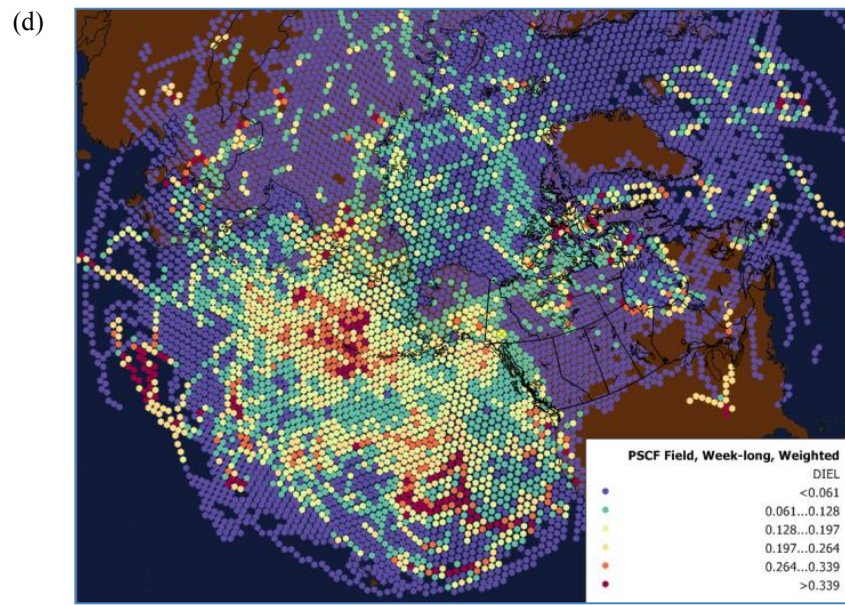
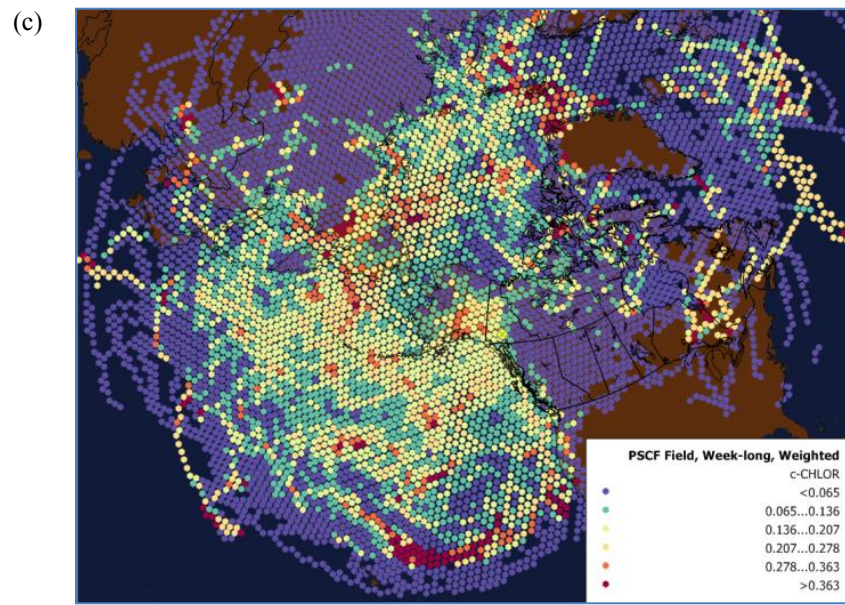
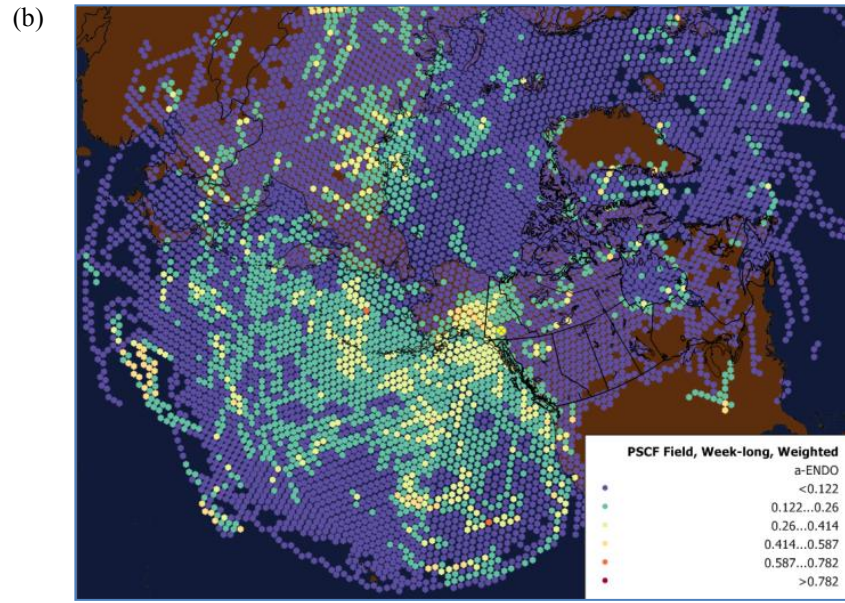
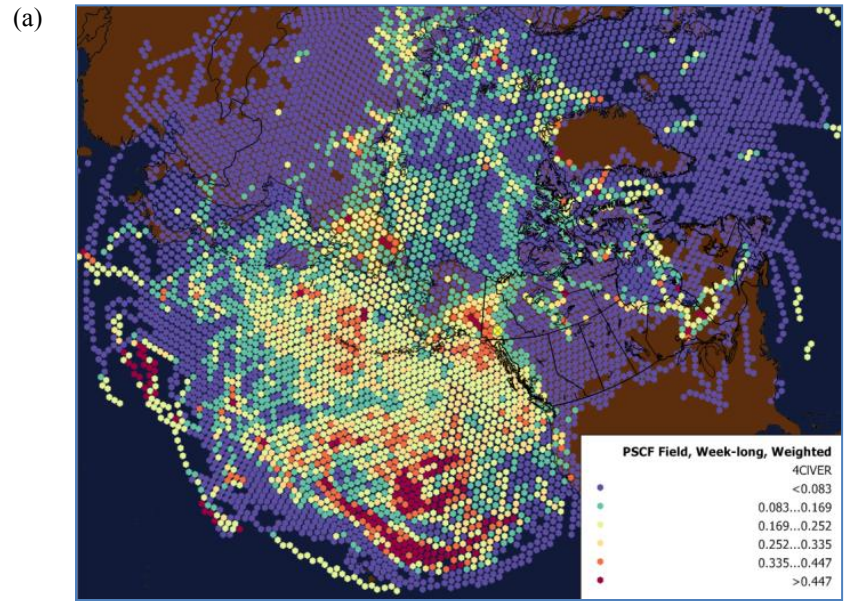


Fig. S4. Reducing the amount of data may make interpretation of the trajectory statistics more straightforward: however, increasing the size of the cells into which the data are binned does not appreciably reduce the noise in the weekly CWT field (a) and (b); the same applies to the CWT field for the day-long measurements (c).



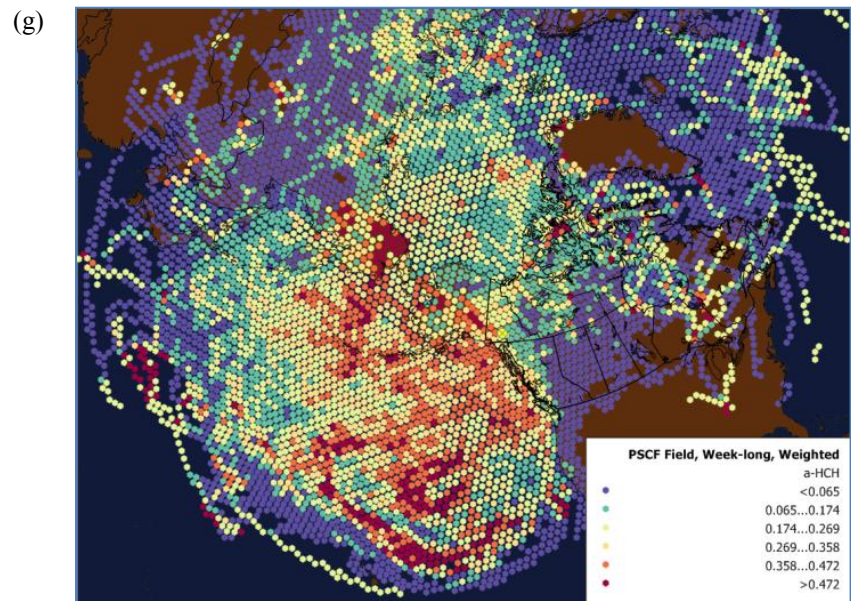
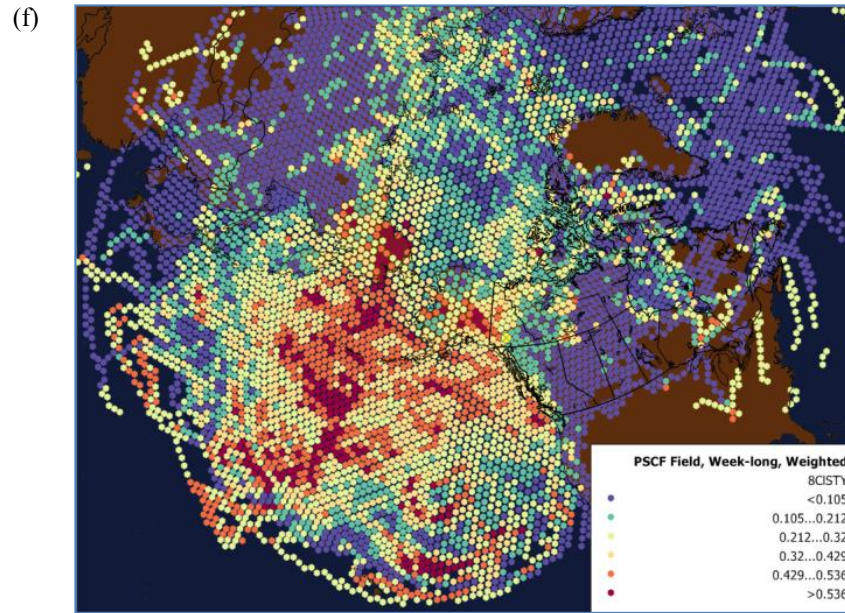
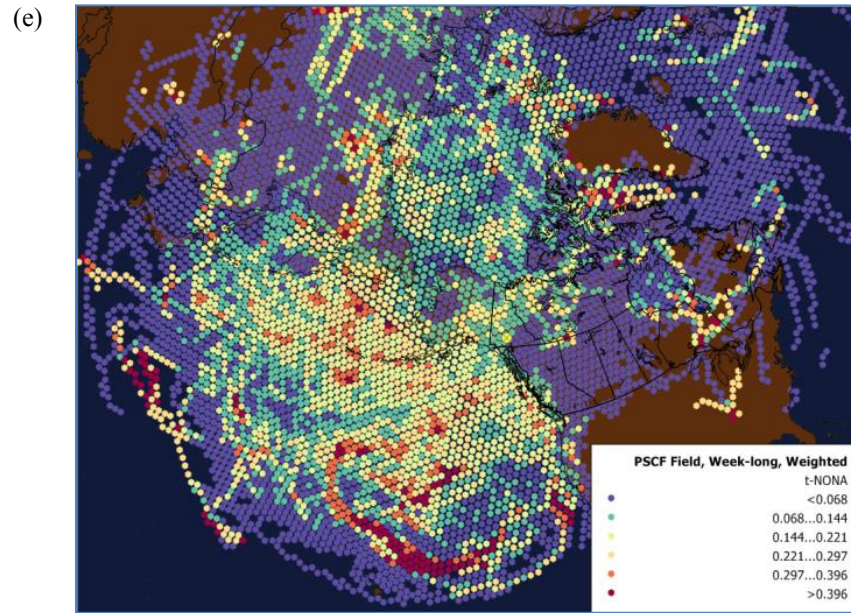


Fig. S5. Even after applying a weighting scheme to reduce over-normalisation, Potential Source Contribution Function (PSCF) fields for most analytes revealed little in the way of probable source regions, but in general highlighted broad swaths of oceans, especially a band of the Pacific near the equator (a-e); the Bering Strait and Gulf of Alaska were highlighted as potential sources of Octachlorostyrene (8CISTY)(f) and α -hexachlorocyclohexane (a-HCH)(g).

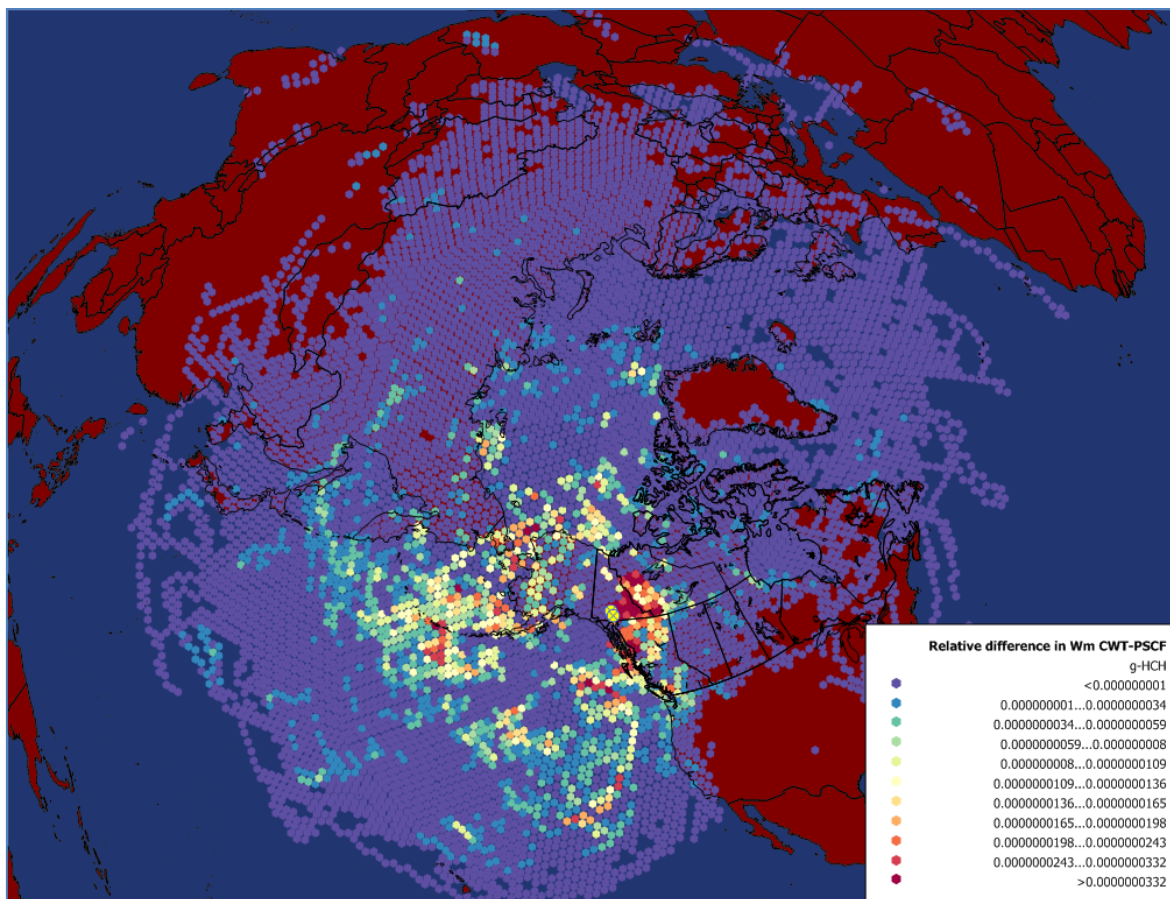


Fig. S6. After scaling, the differences between $W_{m,CWT}$ and $W_{m,PSCF}$ are quite small and are largest close to the arrival point.

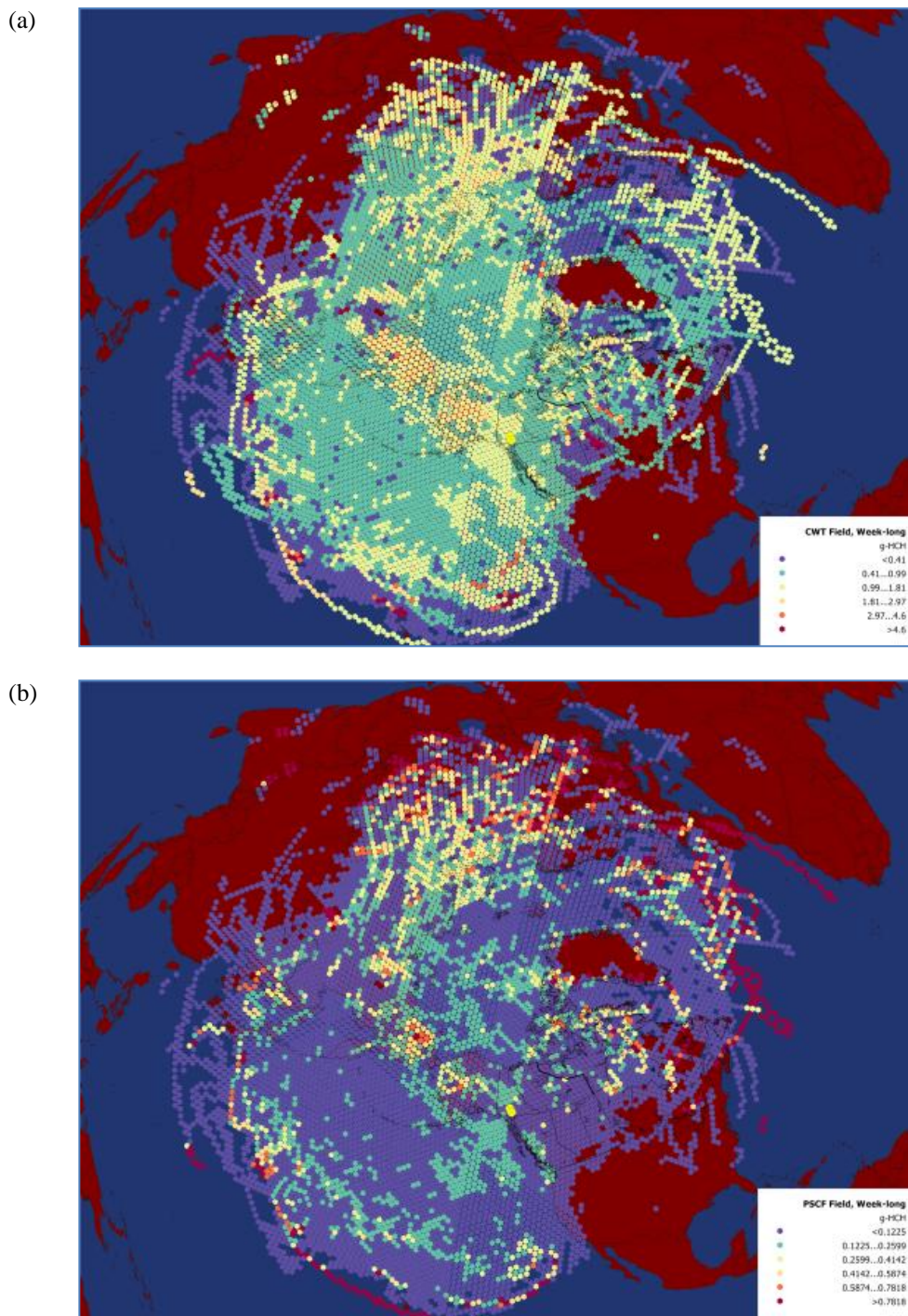


Fig. S7. CWT highlights Alaska and north-eastern Russia as potential sources of γ -hexachlorocyclohexane (g-HCH) to Little Fox Lake (a); PSCF highlights the same areas, but is easier to interpret (b); a blend of the two techniques, using $W_{m,CWT}$ and $N_{m,CWT}$ highlights that the normalisation scheme both highlights likely source areas and cells with low numbers of endpoints due to division by small numbers (c); in CWT it has been common to remove cells with low numbers of endpoints to account for the division by small numbers, but this technique is here applied to the PSCF field resulting in a relatively clear picture of potential sources (d).

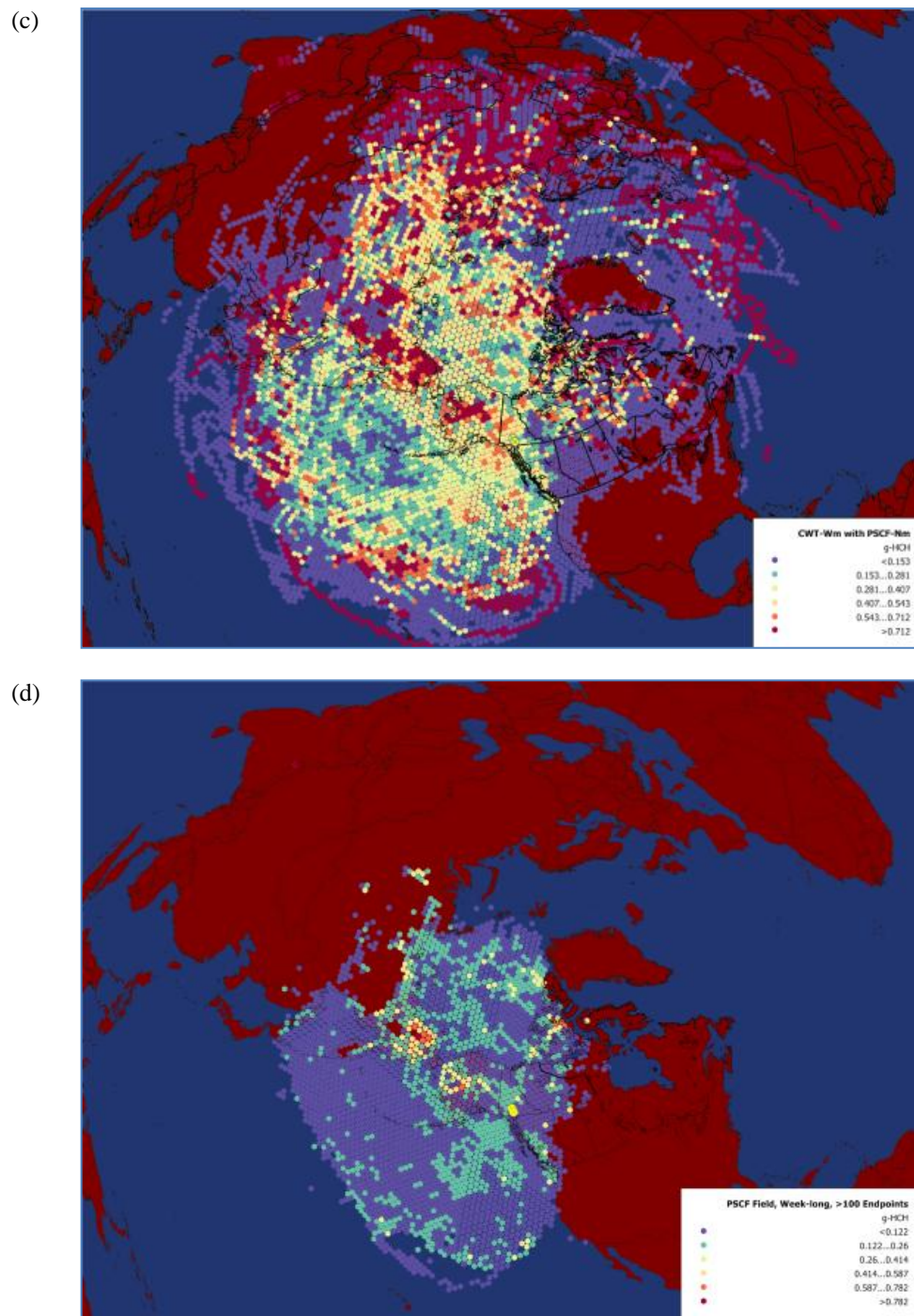
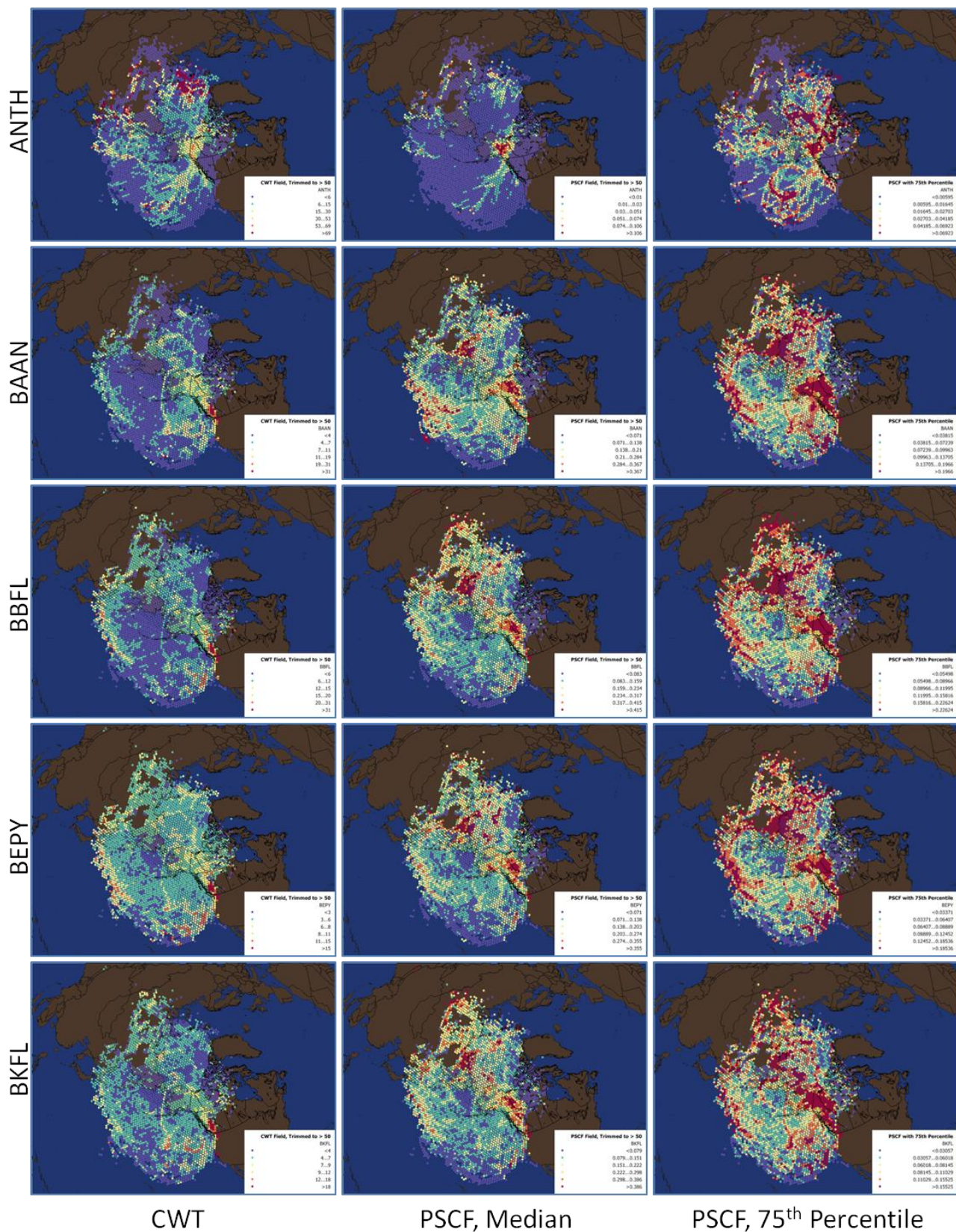
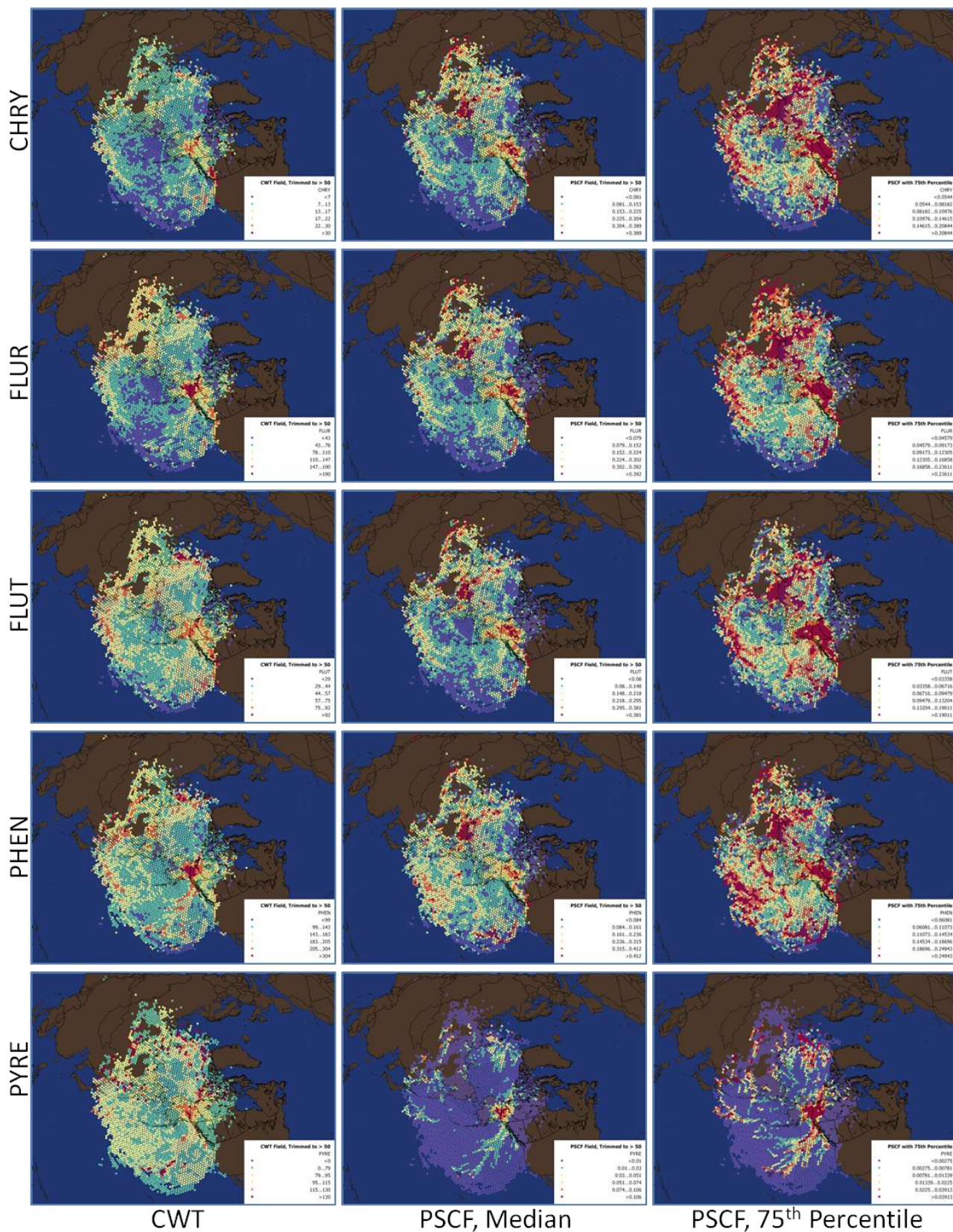


Fig. S7. (Cont.).





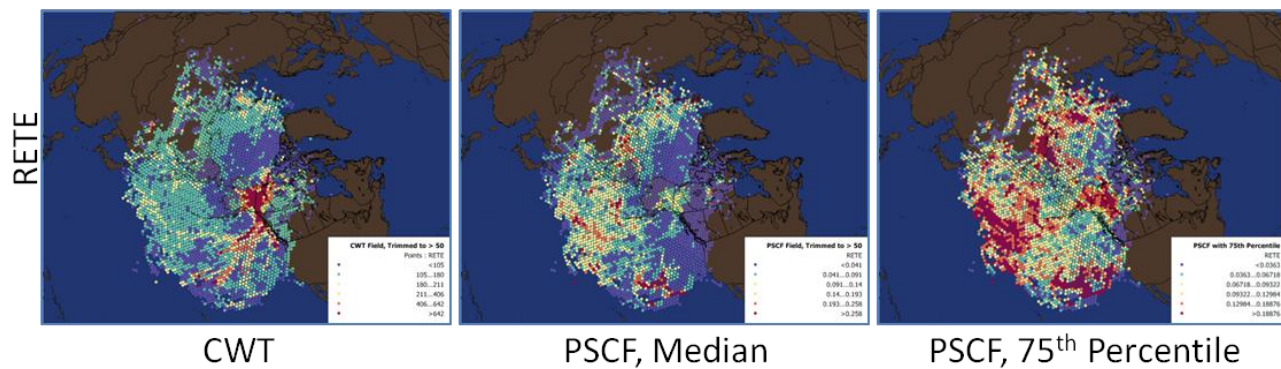


Fig. S8. Maps for CWT, PSCF using the median measurement as the threshold for inclusion and PSCF using the 75th percentile measurement as the threshold for inclusion for polycyclic aromatic hydrocarbons measured in week-long samples at Little Fox Lake.

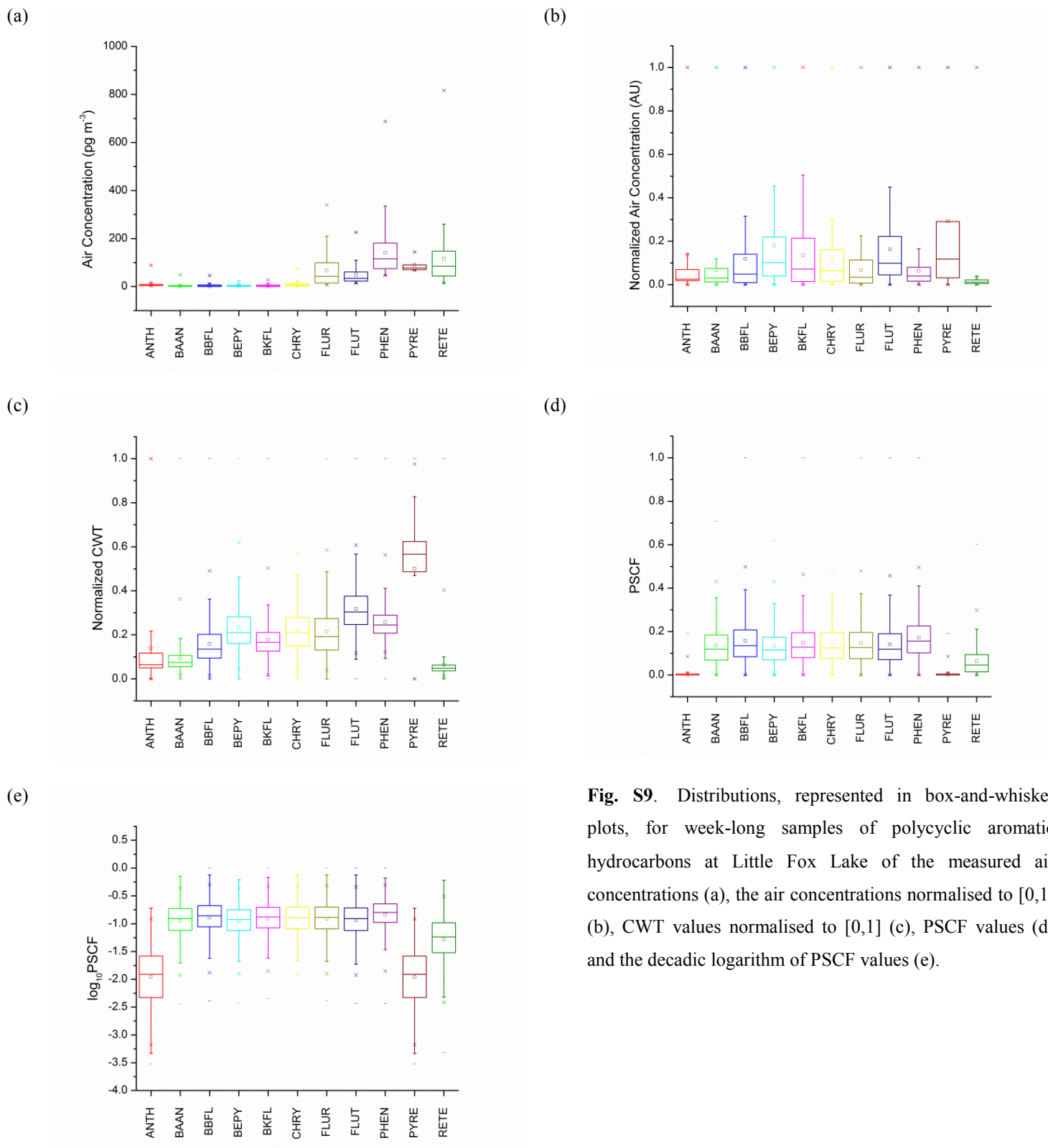


Fig. S9. Distributions, represented in box-and-whisker plots, for week-long samples of polycyclic aromatic hydrocarbons at Little Fox Lake of the measured air concentrations (a), the air concentrations normalised to [0,1] (b), CWT values normalised to [0,1] (c), PSCF values (d) and the decadic logarithm of PSCF values (e).

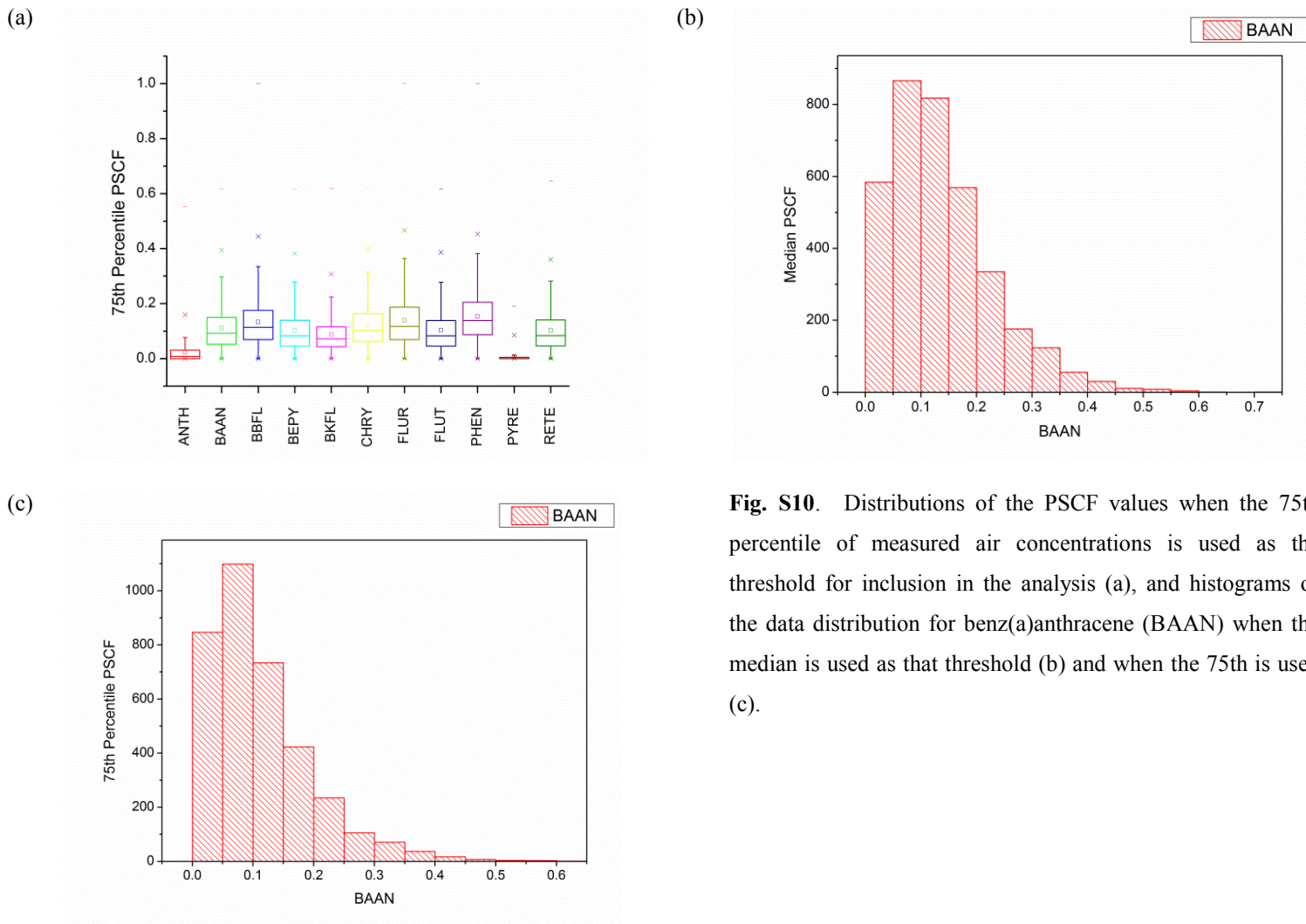


Fig. S10. Distributions of the PSCF values when the 75th percentile of measured air concentrations is used as the threshold for inclusion in the analysis (a), and histograms of the data distribution for benz(a)anthracene (BAAN) when the median is used as that threshold (b) and when the 75th is used (c).

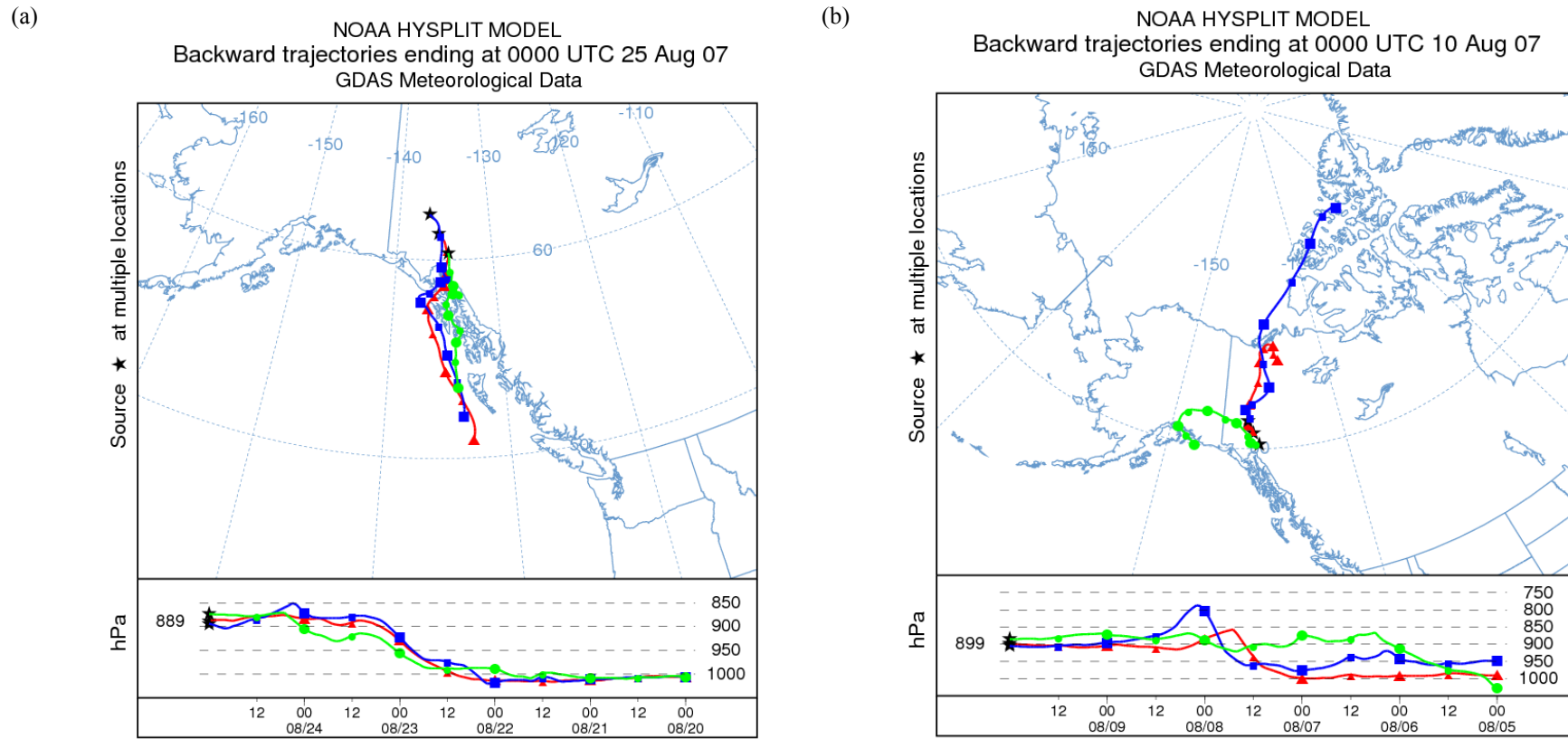


Fig. S11. Back-trajectories started 1° North and 1° South of Little Fox Lake can take slightly different paths, and be used to represent some of the uncertainty inherent in the trajectory analysis (a); however, trajectories are quite sensitive to starting conditions and can propagate in very different manners (b).

Table S12. Correlations between PAH measurements and the significances thereof, with the significant correlations ($P < 0.01$) in bold

		ANTH	BAAN	BBFL	BEPY	BKFL	CHRY	FLUR	FLUT	PHEN	PYRE	RETE
ANTH	R^2	1	0.35	0.07	0.28	0.07	0.79	0.64	0.79	0.86	0.96	0.92
	Sig.		1.74E-01	7.89E-01	2.94E-01	7.91E-01	1.05E-04	5.24E-03	1.09E-04	8.13E-06	2.81E-03	1.36E-07
BAAN	R^2	0.35	1	0.79	0.80	0.79	0.78	0.58	0.66	0.41	0.16	0.34
	Sig.	1.74E-01		0.00E+00	0.00E+00	3.33E-15	0.00E+00	4.13E-09	1.02E-09	1.09E-04	6.73E-01	2.81E-03
BBFL	R^2	0.07	0.79	1	0.86	0.84	0.73	0.74	0.65	0.40	-0.10	0.25
	Sig.	7.89E-01	0.00E+00		0.00E+00	0.00E+00	9.99E-15	2.22E-16	1.54E-09	1.39E-04	7.99E-01	3.26E-02
BEPY	R^2	0.28	0.80	0.86	1	0.80	0.73	0.84	0.64	0.57	-0.02	0.49
	Sig.	2.94E-01	0.00E+00	0.00E+00		1.82E-14	4.11E-12	0.00E+00	3.82E-08	3.69E-07	9.62E-01	6.44E-05
BKFL	R^2	0.07	0.79	0.84	0.80	1	0.68	0.61	0.52	0.29	-0.21	0.17
	Sig.	7.91E-01	3.33E-15	0.00E+00	1.82E-14		1.37E-10	3.72E-08	1.30E-05	1.78E-02	5.92E-01	1.76E-01
CHRY	R^2	0.79	0.78	0.73	0.73	0.68	1	0.89	0.95	0.73	0.78	0.65
	Sig.	1.05E-04	0.00E+00	9.99E-15	4.11E-12	1.37E-10		0.00E+00	0.00E+00	1.69E-14	1.37E-02	2.37E-10
FLUR	R^2	0.64	0.58	0.74	0.84	0.61	0.89	1	0.90	0.76	0.79	0.61
	Sig.	5.24E-03	4.13E-09	2.22E-16	0.00E+00	3.72E-08	0.00E+00		0.00E+00	0.00E+00	1.94E-02	4.75E-09
FLUT	R^2	0.79	0.66	0.65	0.64	0.52	0.95	0.90	1	0.83	0.86	0.69
	Sig.	1.09E-04	1.02E-09	1.54E-09	3.82E-08	1.30E-05	0.00E+00	0.00E+00		0.00E+00	2.84E-03	1.38E-10
PHEN	R^2	0.86	0.41	0.40	0.57	0.29	0.73	0.76	0.83	1	0.86	0.92
	Sig.	8.13E-06	1.09E-04	1.39E-04	3.69E-07	1.78E-02	1.69E-14	0.00E+00	0.00E+00			6.46E-03
PYRE	R^2	0.96	0.16	-0.10	-0.02	-0.21	0.78	0.79	0.86	0.86	1	0.92
	Sig.	2.81E-03	6.73E-01	7.99E-01	9.62E-01	5.92E-01	1.37E-02	1.94E-02	2.84E-03	6.46E-03		1.41E-03
RETE	R^2	0.92	0.34	0.25	0.49	0.17	0.65	0.61	0.69	0.92	0.92	1
	Sig.	1.36E-07	2.81E-03	3.26E-02	6.44E-05	1.76E-01	2.37E-10	4.75E-09	1.38E-10	0.00E+00	1.41E-03	

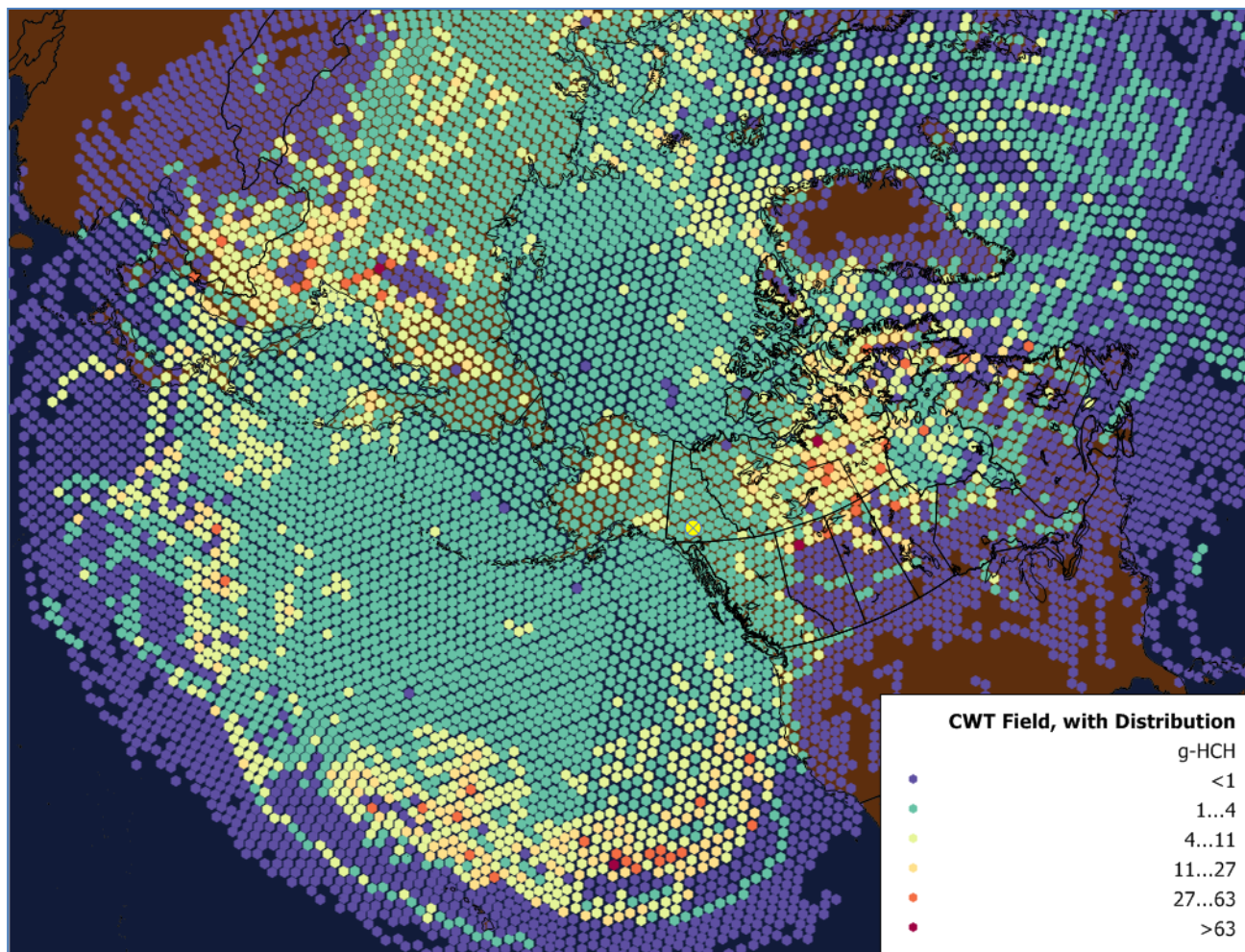


Fig. S12. When the back-trajectories arriving in the six cells surrounding Little Fox Lake were combined with those from Little Fox Lake the CWT analysis is smoothed, but a source region suggested for γ -hexachlorocyclohexane (g-HCH) – Alaska – remains highlighted. However, so do several cells at points quite distant from the arrival area.

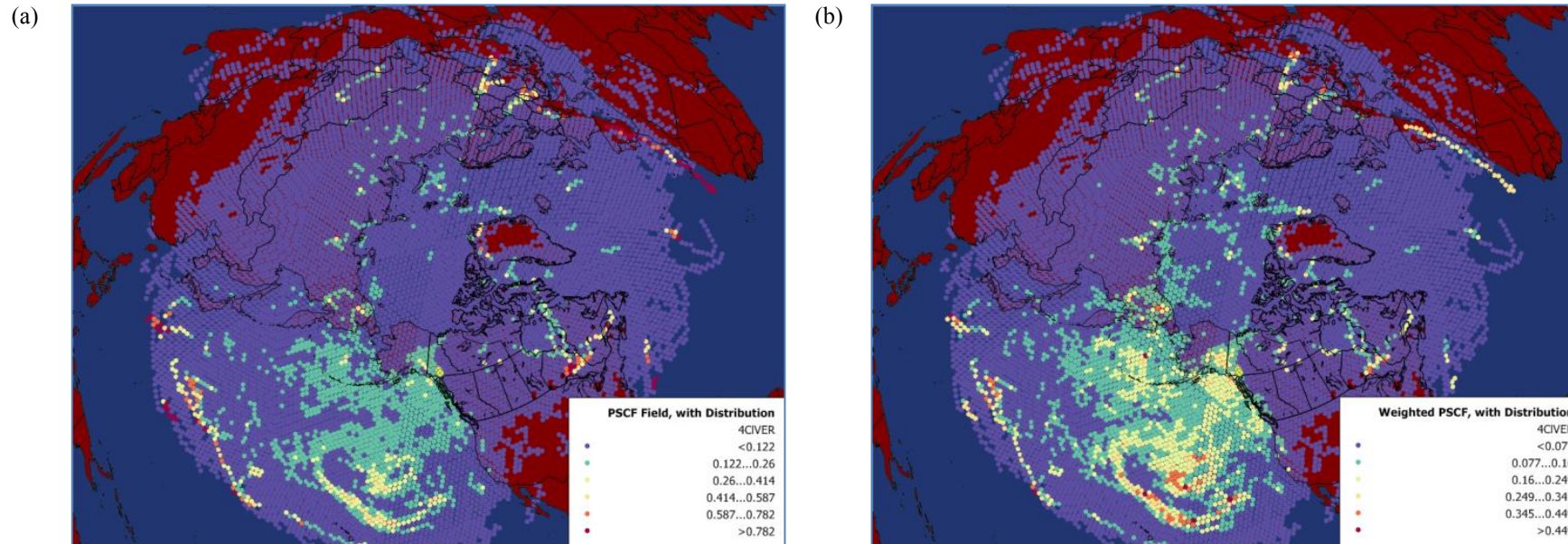


Fig. S13. For tetrachloroveratrole (4CIVER), including the trajectories for the 6 cells surrounding Little Fox Lake to represent uncertainty masks the large potential source area over the Pacific somewhat (a), but applying the weighting scheme highlights the area more strongly again (b), suggesting a strong meteorological connection.

Concentration distributions

Table S13. Distributions of the concentrations and the logarithm of the concentrations of the analytes that appeared in more than 14% of samples, and the results of three tests for the normality of those distributions

Compound	Concentration		Shapiro–			log-Concentration		Kolmogorov–		
	Skewness	Kurtosis	Wilk	Lilliefors	Smirnov	Skewness	Kurtosis	Wilk	Lilliefors	Smirnov
α -HCH	0.55	-0.76	N	N	N	-0.71	-0.61	N	N	N
β -HCH	0.78	-0.62	N	Y	Y	-0.45	-0.90	Y	Y	Y
γ -HCH	3.28	11.50	N	N	N	0.22	0.27	Y	Y	Y
8CISTY	0.45	0.25	N	Y	Y	-1.69	4.52	N	N	Y
c-CHLOR	1.10	0.75	N	N	Y	-0.05	-0.31	Y	Y	Y
t-NONA	1.52	2.01	N	N	N	0.39	-0.15	Y	Y	Y
DIEL	0.52	-0.36	Y	Y	Y	-0.06	-0.52	Y	Y	Y
<i>p,p'</i> -DDE	-0.75	1.53	N	N	Y	-2.08	5.33	N	N	N
5CIANI	2.35	4.59	N	N	N	0.96	-0.28	N	N	Y
α -ENDO	1.73	4.33	N	N	Y	-0.60	-0.31	N	N	Y
4CIVER	1.10	0.63	N	N	N	-0.36	-0.79	N	Y	Y
6CIBD	4.69	24.94	N	N	N	0.90	0.51	N	N	Y

Table S13 contains the results of an analysis of the distributions of the concentration data. Compounds that have a reasonably high frequency of detection can be divided into two broad categories: those with a nearer to normal distribution and those with a nearer to log-normal (that is, exponential) distribution. Conceptually, compounds which are measured at a true ‘background’ site – one that is truly remote and is not directly influenced by emissions of SVOCs from any one area – ought to have a narrow, normal distribution of concentrations. Air arriving at that site from any direction should have approximately the same loading of contaminants. In contrast, if concentrations at a site are influenced by emissions in a distinct source area, causing the arriving air to be especially high in contaminants because of a meteorological event that couples it to a source, then it ought to have an exponential distribution, with many low level measurements and a decreasing number of higher concentration measurements. This is because each sample is a measurement of air parcels arriving over an entire week from multiple directions: if samples were limited to individual transport pathways or directions (e.g. Offenthaler et al.^[4]) a bimodal distribution would be anticipated as each sample would have either a high or a low concentration. This ignores the effect of varying distances to these distinct source regions, and the dilution that accompanies longer transport to the sampling site. Only the first two distributions are considered here.

The measurements at Little Fox Lake do not fall neatly in these two categories. Table S13 gives the skewness, kurtosis and the results of three normality tests – the Shapiro–Wilk test, the Lilliefors test and the Kolmogorov–Smirnov test – both on the concentrations and the base-ten logarithm of the concentrations of those compounds found in more than 15 % of samples that are not suspected to have ‘broken through’. The three tests disagree more often than not. Of the three the Kolmogorov–Smirnov has been argued to be the least robust,^[5] and is the one that most often provides a positive result for normality. In this discussion it is taken that a set that passes more tests is more normal, which is supported by visual examination of the histograms in Fig. S14.

Two compounds, *para,para'*-dichlorodiphenyldichloroethene and octachlorostyrene, exhibit more normal distributions without a logarithmic transformation, and the magnitudes of both skewness and kurtosis increase with the transformation, suggesting Little Fox Lake is a background site for these compounds. It would be expected that, as the metabolite of a long-restricted compound, *para,para'*-dichlorodiphenyldichloroethene would not have distinct source areas, because it is able to undergo long-range transport and it may now be distributed quite evenly in the environment. It should be recalled, however, that *para,para'*-dichlorodiphenyldichloroethene was also only seen in 17 samples, so the statistical analysis is not overly robust. Octachlorostyrene, in contrast, is not produced intentionally, but is a by-product of electrolytic chlorine production.^[6] Despite not being banned, per se, levels of this

contaminant in the environment appear to be decreasing (e.g. Karst-Riddoch et al.^[7]), thus it may be expected to behave similarly to a banned compound.

trans-Nonachlor and γ -hexachlorocyclohexane are the only compounds that fail all three normality tests prior to log transformation, and pass all three after, and the magnitudes of skewness and kurtosis decrease by an order of magnitude. This hints that they may have distinct source areas that influence the sampling site. However, it is not expected that they should demonstrate very different behaviour than *para,para'*-dichlorodiphenyldichloroethene because *trans*-nonachlor is also banned and γ -hexachlorocyclohexane is restricted to very limited use.

β -Hexachlorocyclohexane, *cis*-chlordane, Pentachloroanisole, tetrachloroveratrole and hexachlorobutadiene all undergo moderate increases in normality with the log transformation. There is a very large decrease in absolute skewness and kurtosis for hexachlorobutadiene, but it still only passes the Kolmogorov–Smirnov test of normality. Tetrachloroveratrole also passes the Lilliefors test after log transformation. This by-product of the chlorine bleaching in pulp and paper may well have a distinct source area or areas that act as the main source of it to the air at Little Fox Lake.

α -Endosulfan and α -hexachlorocyclohexane measurements are neither normally, nor exponentially distributed, although the skewness and kurtosis both decrease between distributions of the concentration and log-concentration data for α -endosulfan. It has been reported that oceans now act as a source of α -hexachlorocyclohexane to the atmosphere,^[8] which would explain why Little Fox Lake is neither a background site, nor under the influence of a distinct source area of α -hexachlorocyclohexane.

Dieldrin, seen in only half of the samples at levels above method detection limit, passes all three normality tests both before and after log transformation. The magnitude of the skewness decreases, but the absolute kurtosis increases with the transformation. Tetrachloroveratrole also exhibits this latter behaviour.

In all cases in this work, large changes in the skewness and kurtosis of the data sets upon log transformation are a relatively good indicator of whether the data are more normally or more log-normally distributed. This may serve as a quick measure of whether a site is a background site or receives sporadic contributions from a small number of sources.

CWT and concentration distribution

The distribution analysis suggests that *para,para'*-dichlorodiphenyldichloroethene ought not to have individual sources revealed by the trajectory analysis. However, of the compounds detected in more than a few percent of samples, it stands out from the other analytes, particularly in that it shows high CWT values are largely restricted to cells over the Arctic Ocean. The map only of W_m (Fig. S15a) reflects that

this compound was only seen in around a 5th of the samples: it has a much smaller area with non-zero cells than the other compounds. The large number of non-detects provides binary-like data to the analysis, much like the treatment built into PSCF. Despite sharing similar distribution characteristics, the maps for octachlorostyrene (Fig. S15b,c) share few characteristics with those for *para,para'*-dichlorodiphenyldichloroethene.

The very low detection frequencies of methoxychlor and *ortho,para'*-dichlorodiphenyldichloroethane also provide binary type inputs to the analysis. The unfiltered CWT map for methoxychlor suggests that a small patch of ocean off the coast of California is a source to the air at Little Fox Lake, which is an unlikely scenario; this area is highlighted for most compounds, likely because it is connected meteorologically to Little Fox Lake. After cells with fewer than 100 trajectory segment endpoints are removed (Fig. S16a), no areas appear as strong sources, indicating that a single measurement is insufficient for potential source identification using CWT.

Dieldrin, with a normal and log-normal distribution of concentrations, had a map with remarkably little difference from the other analytes (Fig. S16b). This was not expected, because dieldrin was found above detection limits in only half of the samples, suggesting it might be associated only with air from certain places or directions. However, all measurements are very near the MDL so very small differences in concentration that have nothing to do with air-mass origin may cause it to be above or below this threshold. Tetrachloroveratrole, which passed an additional normality test after log transformation, appeared in most samples, and well above its method detection limit, shared the trait that with a log transformation their absolute skewness decreased while their absolute kurtosis increased. However, the CWT maps of the two are neither notably different from each other, nor together markedly different from the majority of analytes.

The distribution of air concentration analysis suggests that both γ -hexachlorocyclohexane and *trans*-nonachlor should have a few discernible sources. The main text contains a figure of the CWT field for γ -hexachlorocyclohexane with only cells with endpoint counts greater than 100. It suggests two different sources of γ -hexachlorocyclohexane to the air at Little Fox Lake: Chukotka (in north-eastern Russia) and Alaska. The CWT field for *trans*-nonachlor (Fig. S13) is somewhat similar to those of γ -hexachlorocyclohexane although the relative CWT values for the two potential source regions are smaller.

No patterns of particular interest appeared in any of the other SVOCs examined in this work – while their CWT maps were subtly different from one another, the noise was too great or the variability too small for individual sources to be identified.

The statistical distributions of concentrations did not prove to be a reliable predictor of whether a compound had or did not have source regions revealed by the trajectory analysis, nor whether its CWT field displayed weak or strong spatial heterogeneity. It predicted that γ -hexachlorocyclohexane and *trans*-nonachlor should be markedly different from the other analytes, but only γ -hexachlorocyclohexane was visually different. It hinted that *p,p'*-DDE and octachlorostyrene might share some defining characteristics, but only *para,para'*-dichlorodiphenyldichloroethene stood out from the group. The long distance of the site from likely source regions means that back trajectories that connect them are highly uncertain, that if source regions exist they may be at similar distances, and that many analytes arrive at near method detection limit concentrations, all of which may confound such an analysis. In all, Little Fox Lake is a well located site for measuring the background concentration of most SVOCs in the atmosphere.

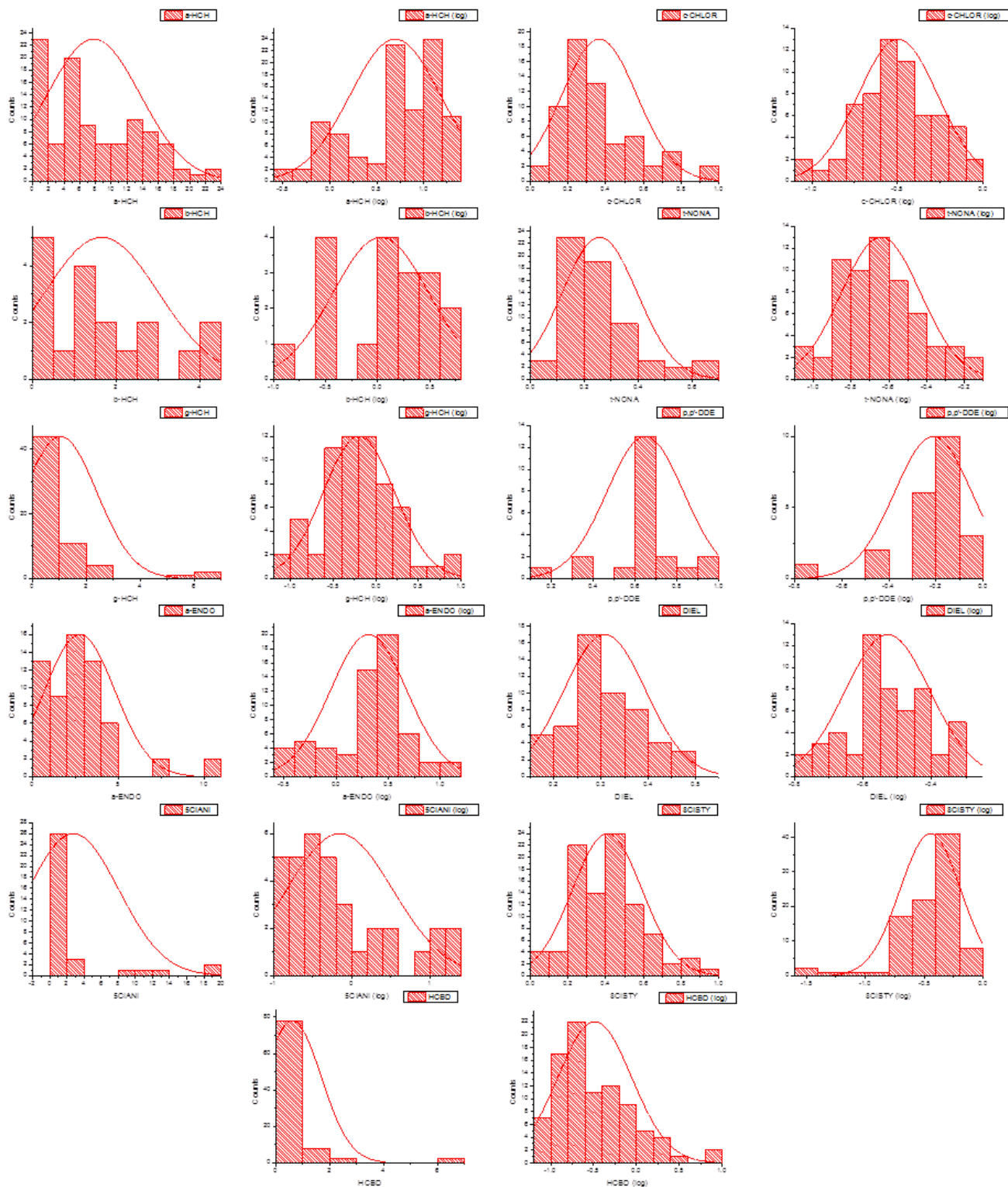


Fig. S14. Histograms of the air concentrations of the SVOCs that appeared above method detection limit in at least 15% of samples, and histograms of the decadic logarithms of those concentrations, generated using OriginPro 8.5.1 Student Version from OriginLabs.

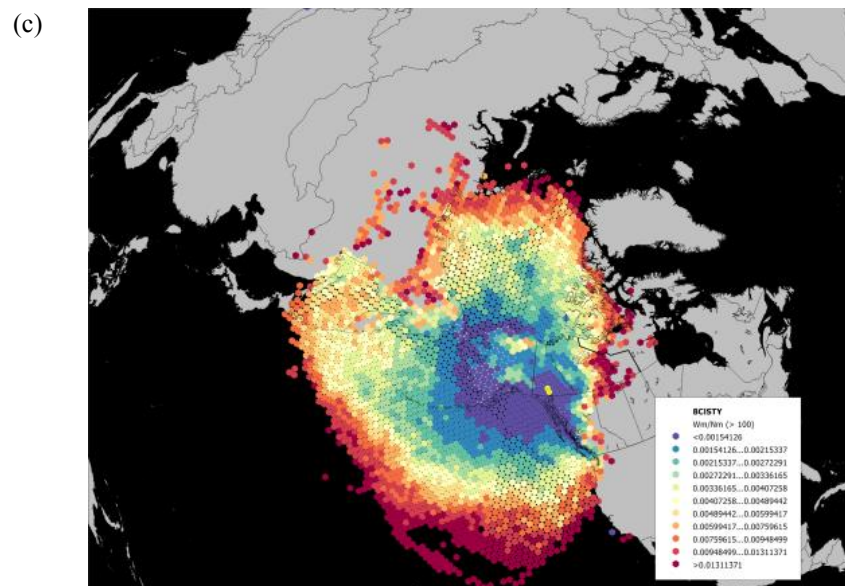
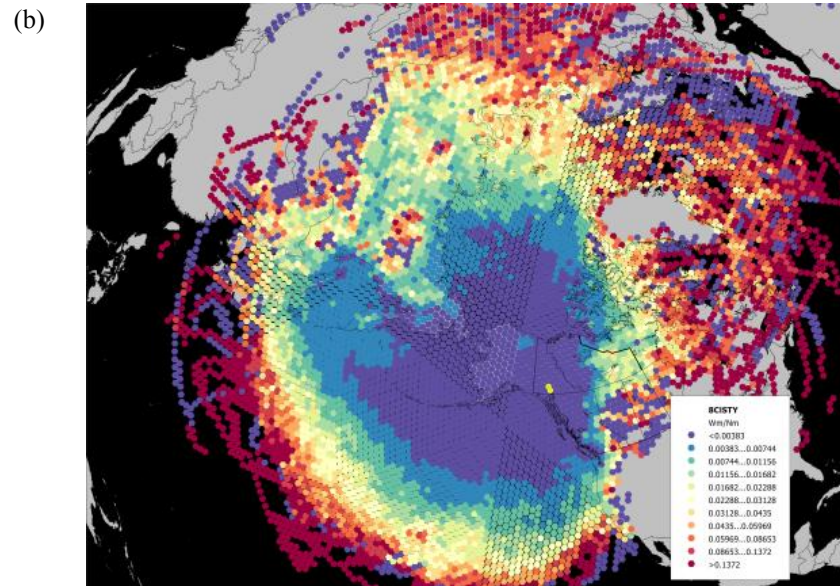
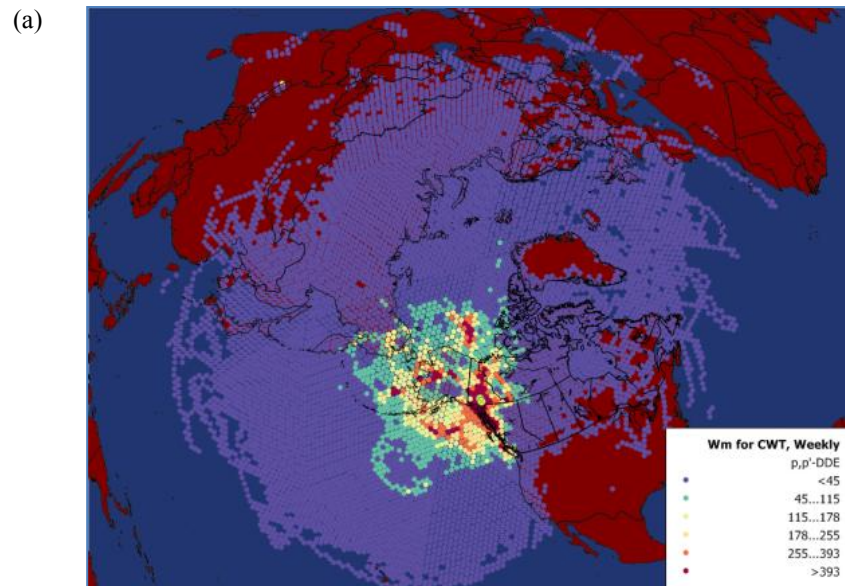


Fig. S15. Maps of $W_{m,CWT}$ for *para,para'*-dichlorodiphenyl-dichloroethene highlight the low number of detects of this compound (a). CWT field for octachlorostyrene (b), even when trimmed (c) is dissimilar to that of *para,para'*-dichlorodiphenyl-dichloroethene despite sharing similar statistical distribution characteristics.

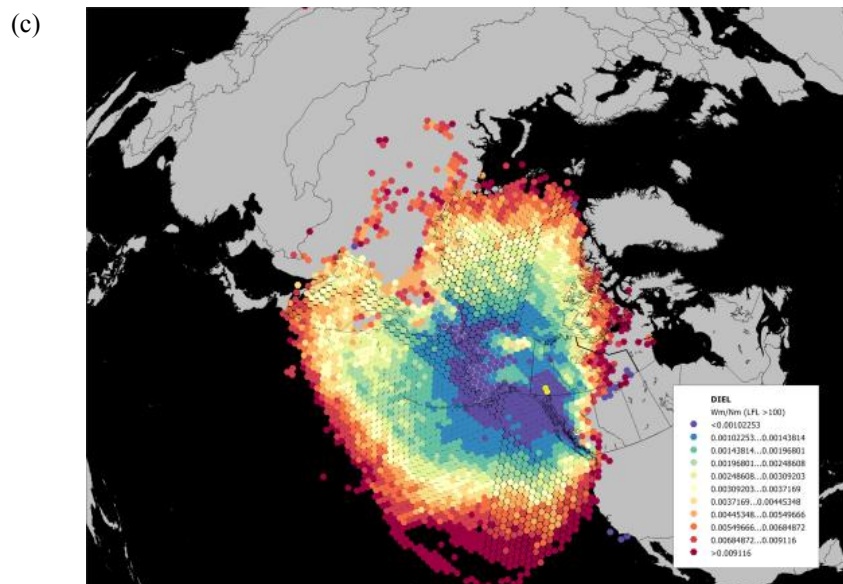
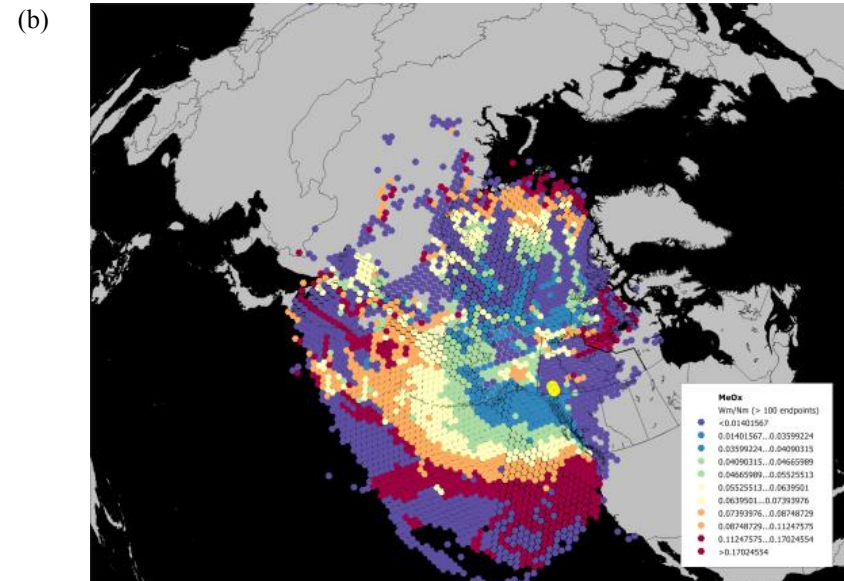
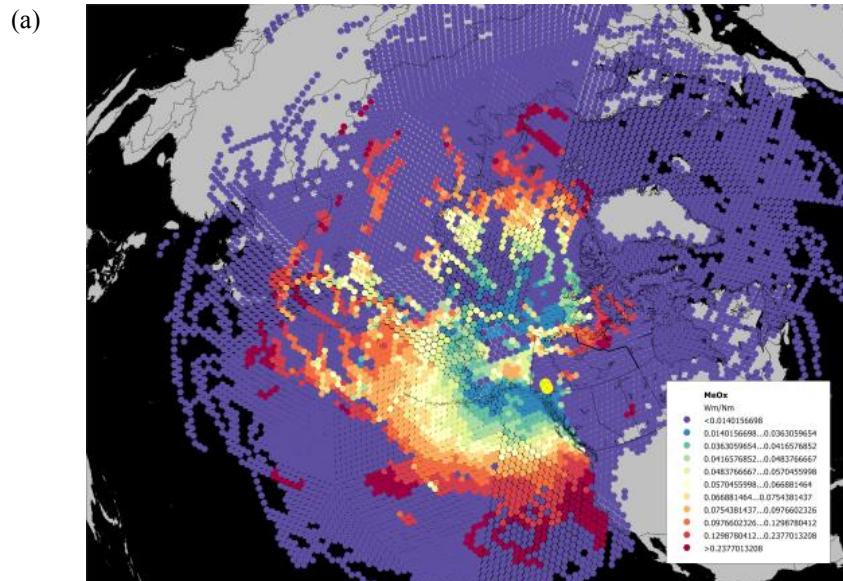


Fig. S16. Map of the CWT field for methoxychlor (a), and with cells with fewer than 100 trajectory segment endpoints removed (b). Concentric ellipses are evident with potential sources around Anchorage, Alaska. Although the peak of methoxychlor concentration in air was strong, the trajectory analysis reveals little when applied to a single observation, and the arbitrary choice of a 100 endpoint per cell cut-off does nothing to aid in interpreting sources of methoxychlor. Dieldrin appears in about half of all samples, but despite having such a binary style data set it shares more similarities with compounds detected in all samples than with methoxychlor (c).

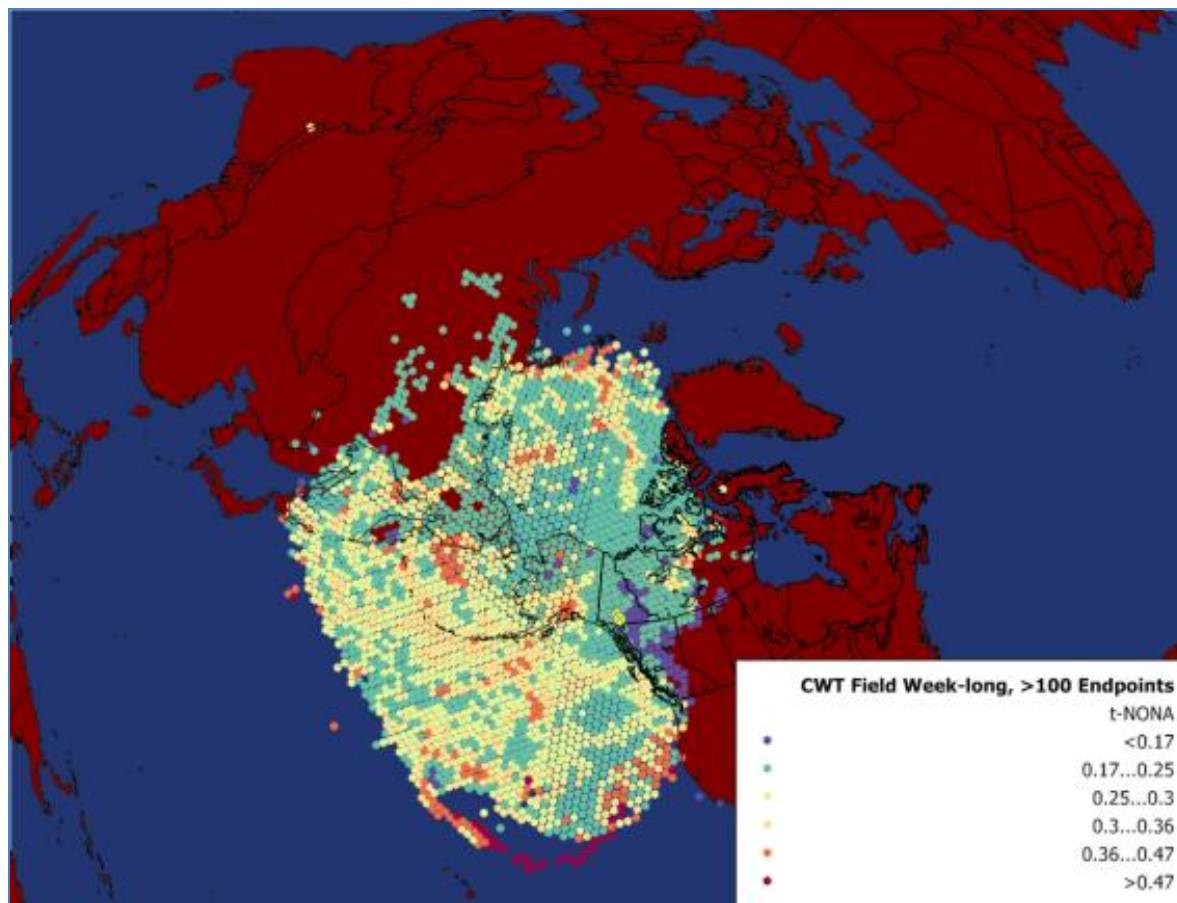


Fig. S17. CWT field for Little Fox Lake with only those cells with endpoint counts greater than 100 for *trans*-nonachlor – which like γ -hexachlorocyclohexane exhibited a log-normal distribution of concentrations in the weekly samples.

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