

**SUPPLEMENTARY MATERIAL**

**ATMOSPHERIC OCCURRENCE, TRANSPORT AND  
DEPOSITION OF POLYCHLORINATED BIPHENYLS AND  
HEXACHLOROBENZENE IN THE MEDITERRANEAN  
AND BLACK SEAS**

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## **SUPPLEMENTARY MATERIAL**

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## **SUPPLEMENT A: SAMPLE COLLECTION**

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FIGURE S1: Mediterranean Sea sub-basins reference map

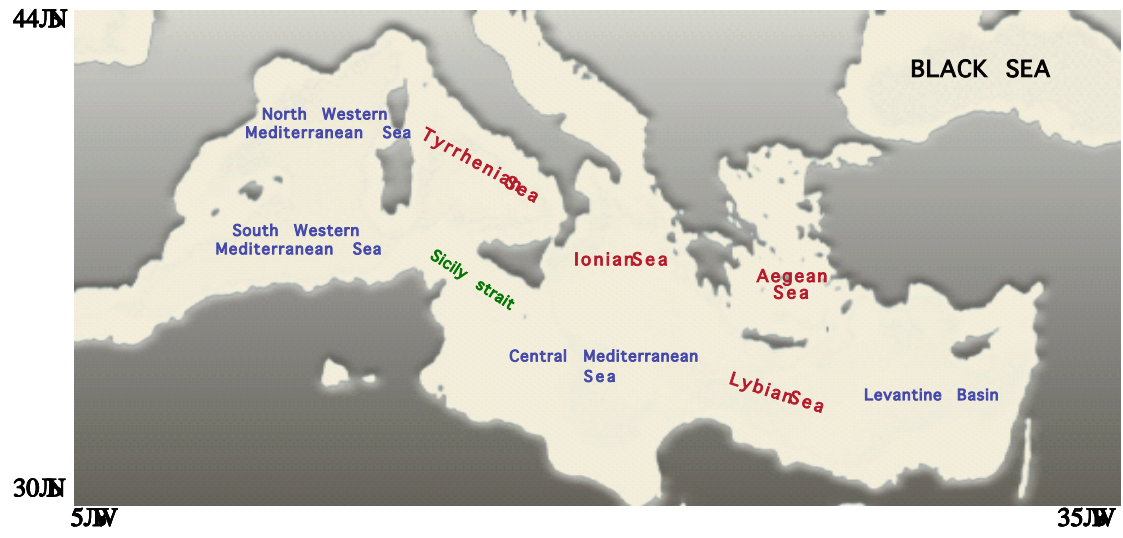
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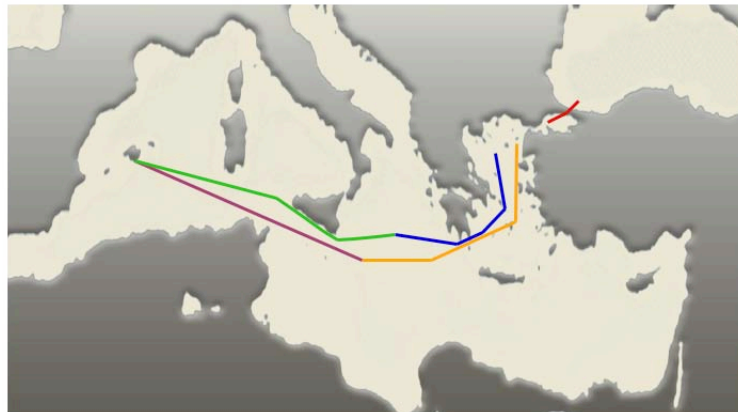
**FIGURE S1: Mediterranean Sea sub-basins reference map**








**TABLE S1: Description of the collected air samples (date and location, sampled volume, amount of aerosols).**

<b>SAMPLE</b>	<b>Date</b>	<b>Time</b>	<b>End Date</b>	<b>End Time</b>	<b>FROM Lat [°N]</b>	<b>FROM Long [°E]</b>	<b>TO Lat [°N]</b>	<b>TO Long [°E]</b>	<b>SAMPLED VOLUME (m<sup>3</sup>)</b>	<b>AEROSOL AMOUNT (mg m<sup>-3</sup>)</b>
<b>T1</b>	2/6/2006	9:50 pm	3/6/2006	10:30 am	39,608	2,291	41,169	2,288	379	0,099
<b>T2</b>	4/5/2007	10:30 am	5/5/2007	9:00 pm	40,59	2,12	39,54	3,86	282	0,041
<b>T3</b>	4/6/2007	12:00 pm	5/6/2007	9:30 am	39,06	5,76	39,56	2,64	441	0,056
<b>STIIa</b>	5/7/2006	7:30 pm	5/7/2006	5:30 am	38,44	3,65	38,42	3,61	361	-
<b>TIIb</b>	4/7/2006	9:30 pm	5/7/2006	5:30 am	38,40	3,61	38,43	3,64	300	0,155
<b>T4</b>	4/7/2006	7:00 am	4/7/2006	9:00 pm	38,14	4,54	38,40	3,61	490	0,149
<b>STIIa</b>	2/7/2006	9:00 pm	2/7/006	9:40 pm	37,95	5,47	37,97	5,39	383	-
<b>STIIb</b>	3/7/2006	9:40 pm	4/7/2006	6:30 am	37,97	5,39	38,13	4,54	330	0,189
<b>T5</b>	2/7/2006	11:00 am	2/7/2006	8:00 pm	37,78	6,91	37,92	5,59	353	0,119
<b>T6</b>	6/6/2006	9:00 am	6/6/2006	7:00 pm	37,92	11,33	37,29	12,87	305	0,111
<b>T7</b>	6/6/2006	7:00 pm	7/6/2006	9:00 am	37,29	12,87	36,79	14,25	262	0,184
<b>T8</b>	7/6/2006	9:00 am	6/7/2006	5:30 pm	36,79	14,25	36,46	16,10	230	0,070
<b>T9</b>	7/6/2006	9:40 pm	8/6/2006	7:30 am	36,46	16,10	36,53	17,48	359	0,105
<b>T10</b>	8/6/2006	7:40 am	8/6/2006	9:30 pm	36,53	17,48	36,73	18,99	447	-
<b>STIIIa</b>	25/6/2006	5:30 am	25/6/2006	7:00 pm	35,72	20,74	35,69	20,77	448	0,137
<b>STIIIb</b>	25/6/2006	8:30 pm	26/6/2006	9:00 am	35,68	20,79	35,67	20,05	304	0,234
<b>T11</b>	13/5/2007	9:30 am	14/5/2007	8:45 am	35,08	19,40	34,28	21,02	947	0,035
<b>T12</b>	14/5/2007	9:00 am	15/5/2007	9:00 am	34,28	21,02	33,11	24,73	950	0,083
<b>T13</b>	15/5/2007	9:00 am	16/5/2007	9:00 am	33,10	24,34	32,46	27,26	814	0,134
<b>T14</b>	16/5/2007	9:00 am	17/5/2007	7:00 am	32,46	27,26	31,29	29,21	414	0,099
<b>T15</b>	17/5/2007	7:00 am	17/5/2007	11:15 am	31,29	29,21	31,188	29,796	446	0,143
<b>T16</b>	21/5/2007	2:00 am	21/5/2007	3:00 pm	31,441	29,736	32,585	29,436	307	0,071
<b>T17</b>	21/5/2007	4:00 pm	22/5/2007	5:15 pm	33,69	29,12	36,08	28,22	848	0,037
<b>T18</b>	12/6/2006	6:30 am	12/6/2006	8:35 pm	36,23	22,35	36,68	23,80	493	-
<b>T19</b>	12/6/2006	9:20 pm	13/6/2006	6:45 am	36,75	23,92	37,52	25,26	350	
<b>T20</b>	13/6/2006	08.30 am	13/6/2006	8:00 pm	37,551	25,285	38,080	24,707	567	0,068
	14/6/2006	09.30 am	14/6/2006	8:00 pm	37,551	25,285	38,080	24,707		
<b>T21</b>	13/6/2006	10:15 pm	14/6/2006	7:00 am	38,22	24,83	40,08	26,34	679	0,090
<b>T22</b>	23/6/2006	8:00 am	23/6/2006	6:00 pm	38,457	25,223	37,056	25,924	304	0,170
<b>T23</b>	22/6/2006	8:00 pm	23/6/2006	5.30 pm	38,457	25,223	37,056	25,924	576	0,086
<b>T24</b>	16/6/2006	6:25 am	16/6/2006	4:30 pm	41,49	29,71	41,04	29,02	373	0,107
<b>STIVa</b>	19/6/2007	7:00 am	19/6/2007	2:30 pm	41,87	30,07	41,89	30,03	277	-
<b>STIVb</b>	19/6/2006	2:30 pm	19/6/2006	9:30 pm	41,89	30,03	41,91	29,98	294	0,143
<b>STIVc</b>	19/6/2006	9:30 pm	20/6/2006	6:00 am	41,91	29,98	41,88	29,84	328	0,272
<b>T25</b>	20/6/2006	7:00 am	20/6/2006	6:00 pm	41,90	29,64	41,13	29,07	371	0,109






**FIGURE S2: Map of the air samples collected with the second and third high volume air samplers during the 2006 cruise.**



	SAMPLE	SAMPLING YEAR
	Ti / Tii / Tiii	2006
	Tiv / Tv / Tvi	2006
	Tvii	2006
	Tviii / Tix / Tx	2006
	Txi / Txii / Txiii	2006

**FIGURE S3: Map of the air samples collected with the second and third high volume air samplers during the 2007 cruise.**



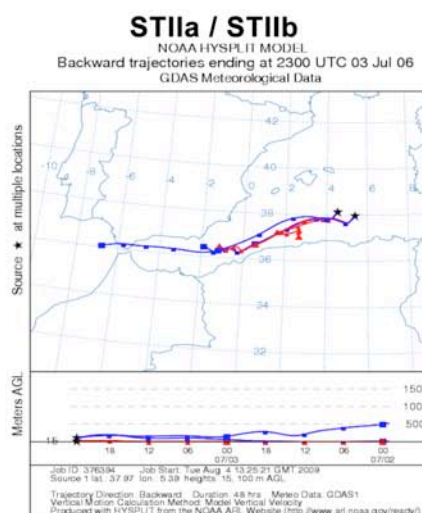
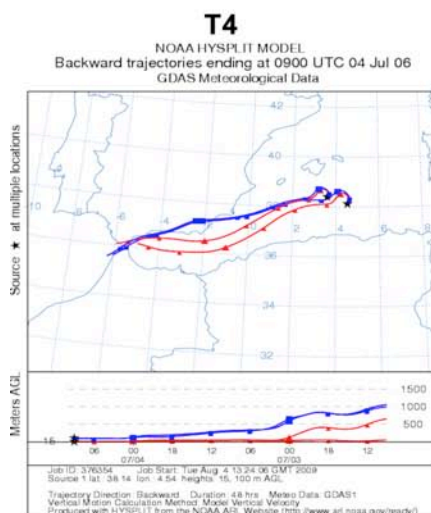
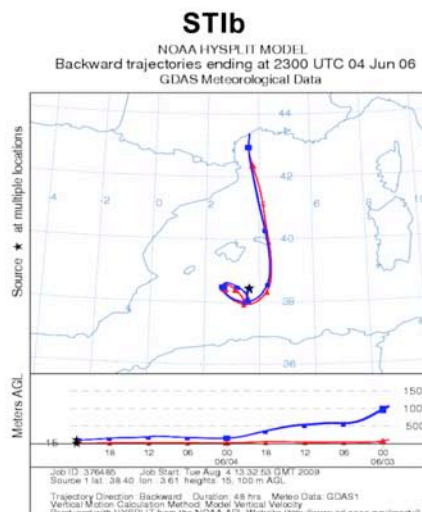
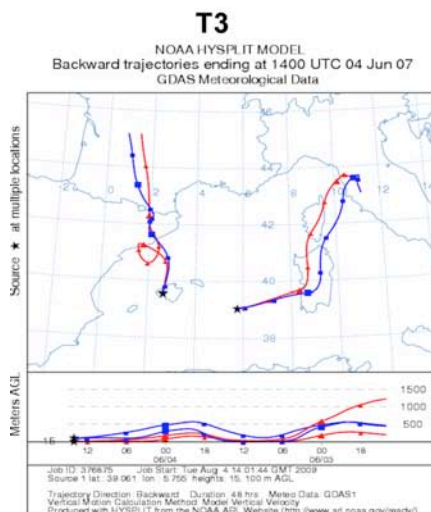
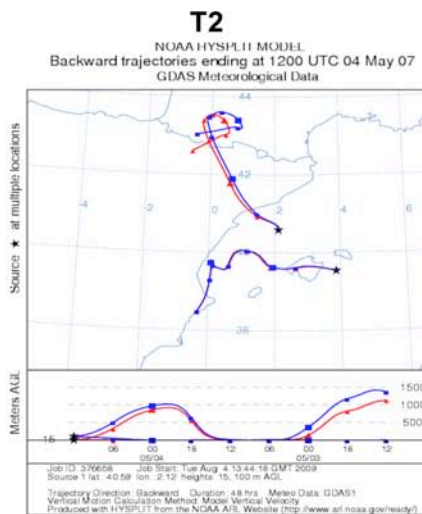
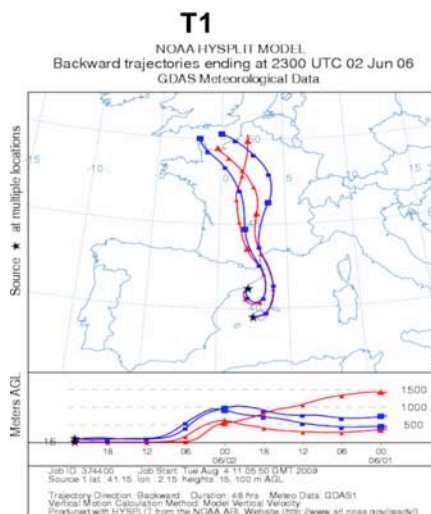
	SAMPLE	SAMPLING YEAR
	Txiv	2007
	Txv	2007
	Txvi	2007
	Txvii	2007
	Txviii	2007

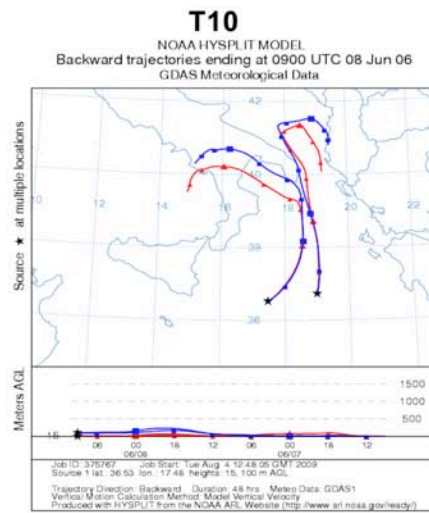
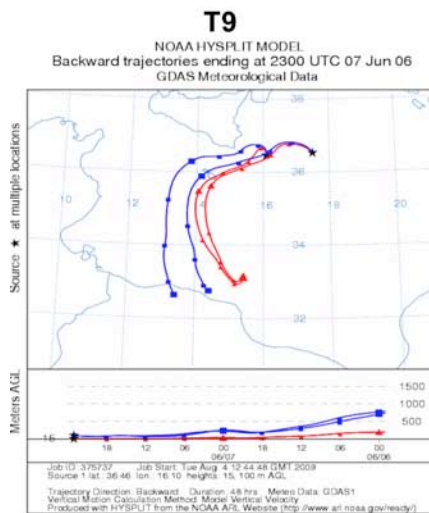
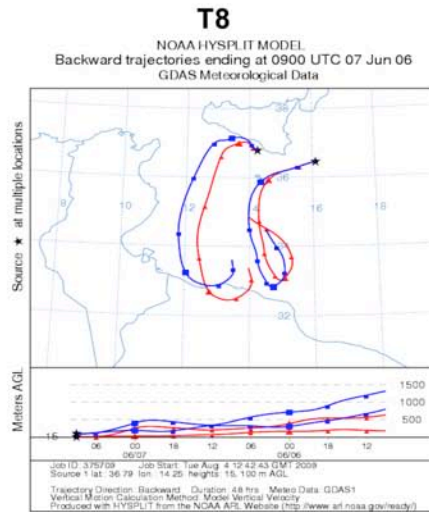
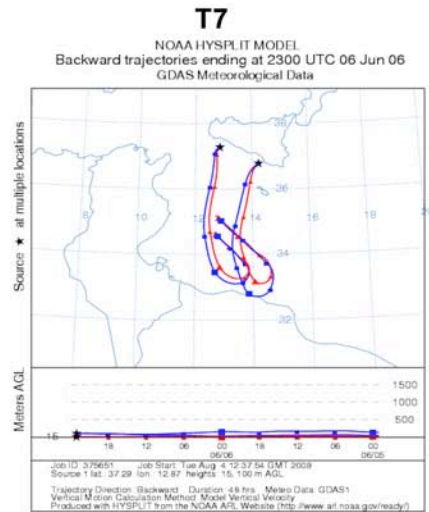
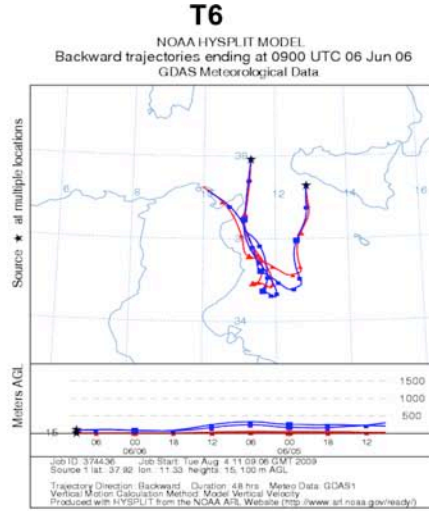
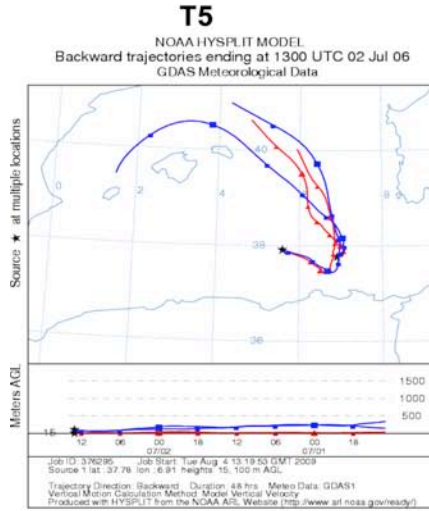


**TABLE S2: Description of the air samples collected with the second and third high volume samplers (date and location).**

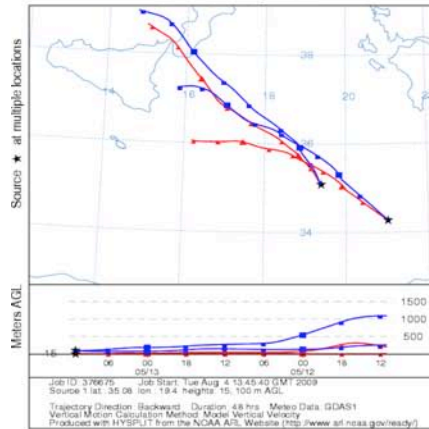
<b>Sample</b>	<b>Date</b>	<b>End Date</b>	<b>Sampling Time</b>	<b>Lat [° N]</b>	<b>Long [° E]</b>	<b>Lat [° N]</b>	<b>Long [° E]</b>
<b>Ti</b>	4 June 2006	8 June 2006	Day	39,305	2,739	36,590	18,583
<b>Tii</b>	3 June 2006	7 June 2006	Night	39,305	2,739	36,590	18,583
<b>Tiii</b>	4 June 2006	8 June 2006	Day + Night	39,305	2,739	36,590	18,583
<b>Tiv</b>	9 June 2006	14 June 2006	Day	36,590	18,583	39,929	25,968
<b>Tv</b>	8 June 2006	12 June 2006	Night	36,590	18,583	39,929	25,968
<b>Tvi</b>	8 June 2006	12 June 2006	Day + Night	36,590	18,583	39,929	25,968
<b>Tvii</b>	15 June 2006	19 June 2006	Day + Night	40,713	27,865	41,370	29,115
<b>Tviii</b>	22 June 2006	26 June 2006	Day	39,580	24,661	35,657	16,533
<b>Tix</b>	22 June 2006	26 June 2006	Night	39,580	24,661	35,657	16,533
<b>Tx</b>	22 June 2006	26 June 2006	Day + Night	39,580	24,661	35,657	16,533
<b>Txi</b>	27 June 2006	4 July 2006	Day	35,657	16,535	39,305	2,739
<b>Txii</b>	30 June 2006	4 July 2006	Night	35,657	16,535	39,305	2,739
<b>Txiii</b>	27 June 2006	4 July 2006	Day + Night	35,657	16,535	39,305	2,739
<b>Txiv</b>	5 May 2007	12 May 2007	Day + Night	39,361	2,757	36,168	16,148
<b>Txv</b>	12 May 2007	16 May 2007	Day + Night	36,168	16,148	32,650	27,591
<b>Txvi</b>	16 May 2007	25 May 2007	Day + Night	32,631	27,617	36,157	24,742
<b>Txvii</b>	25 May 2007	31 May 2007	Day + Night	36,157	27,617	37,370	12,986
<b>Txviii</b>	31 May 2007	6 June 2007	Day + Night	37,377	12,971	41,203	2,279

# SUPPLEMENT B: BACK TRAJECTORIES

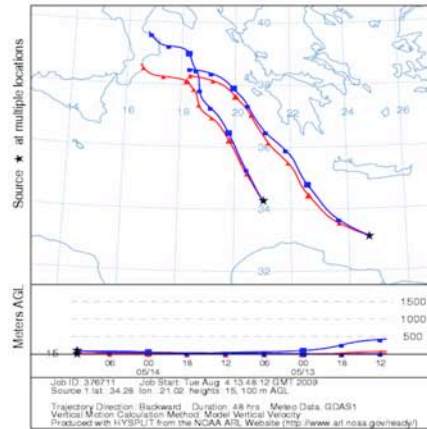




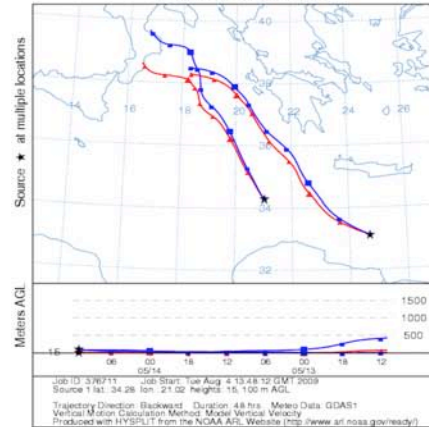
**T11**  
 NOAA HYSPLIT MODEL  
 Backward trajectories ending at 1100 UTC 13 May 07  
 GDAS Meteorological Data



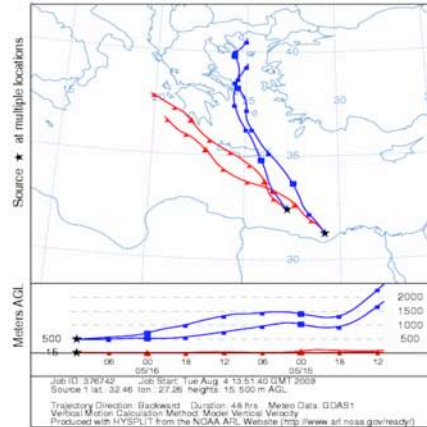
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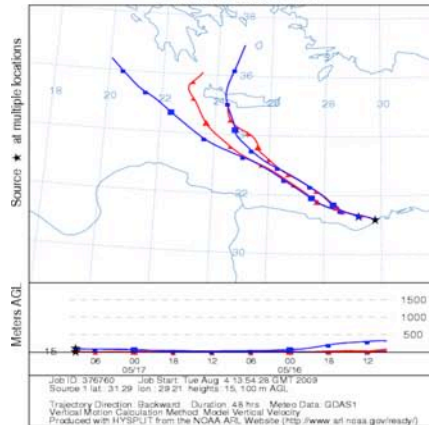
**T13**  
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 GDAS Meteorological Data



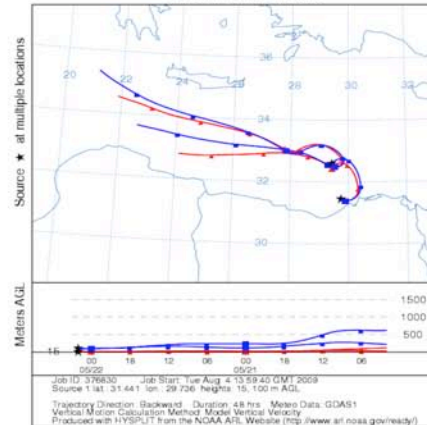
**T14**  
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 GDAS Meteorological Data



**T15**  
 NOAA HYSPLIT MODEL  
 Backward trajectories ending at 0900 UTC 17 May 07  
 GDAS Meteorological Data



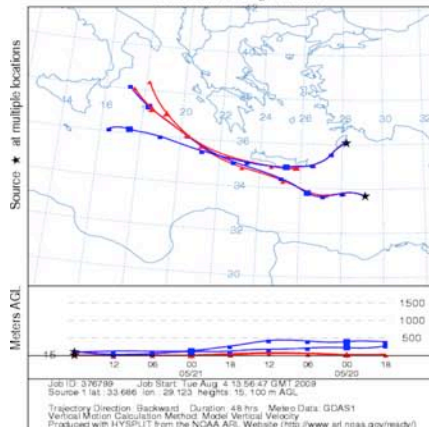
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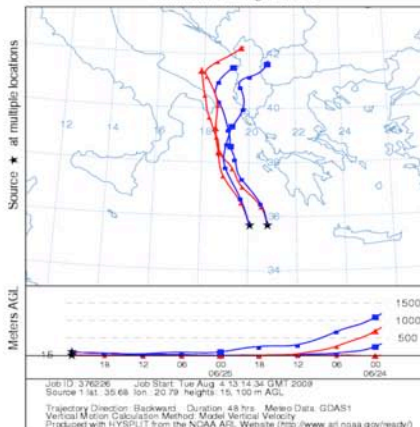
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Backward trajectories ending at 1800 UTC 21 May 07  
GDAS Meteorological Data



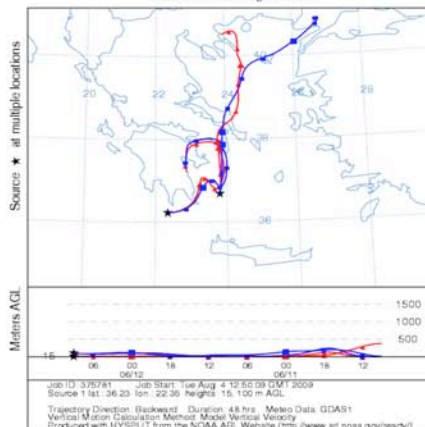
### ST11b

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Backward trajectories ending at 2300 UTC 25 Jun 06  
GDAS Meteorological Data



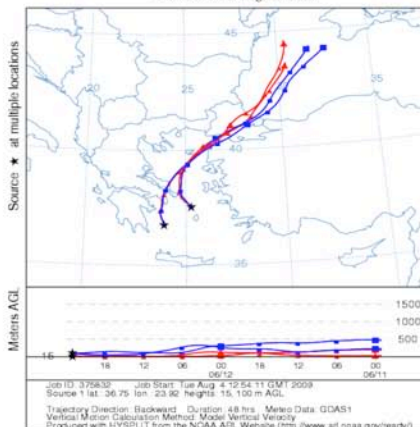
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GDAS Meteorological Data



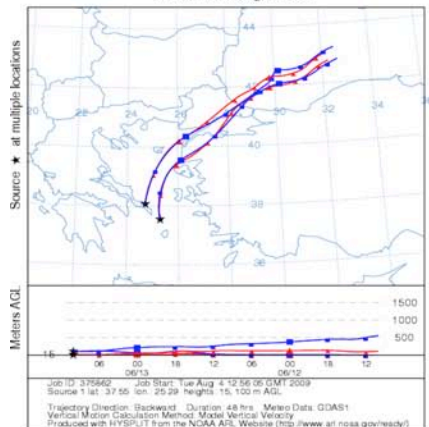
### T19

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GDAS Meteorological Data



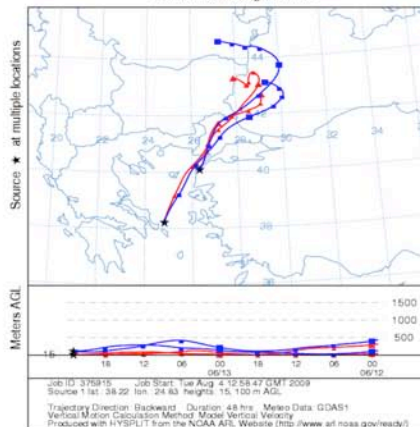
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GDAS Meteorological Data



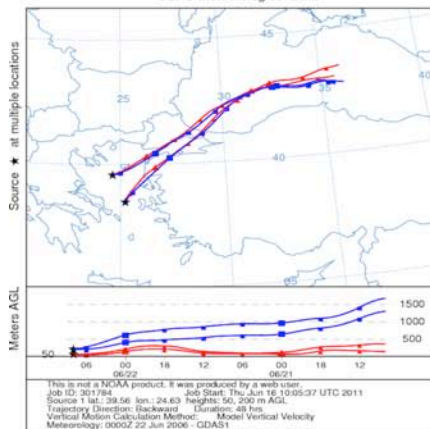
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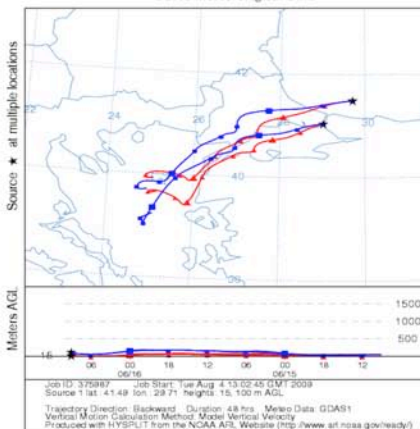
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GDAS Meteorological Data



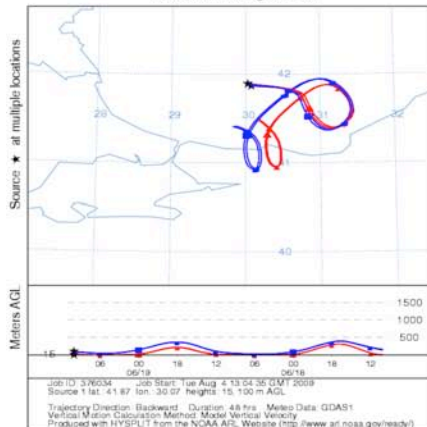
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GDAS Meteorological Data



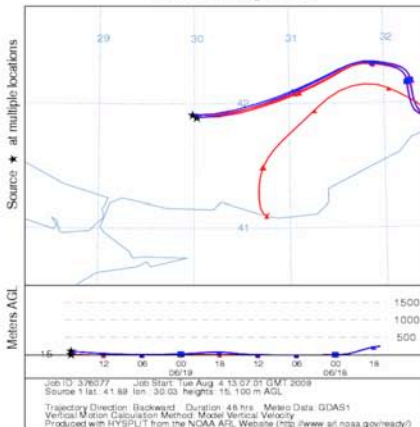
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GDAS Meteorological Data



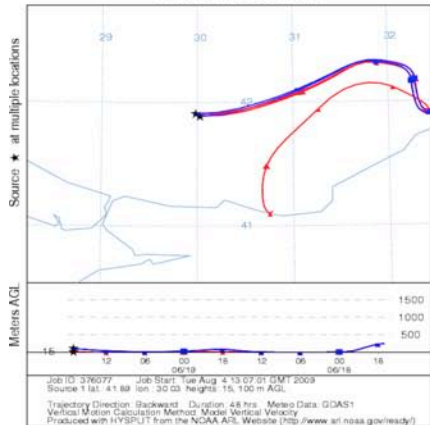
### STIVb

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GDAS Meteorological Data



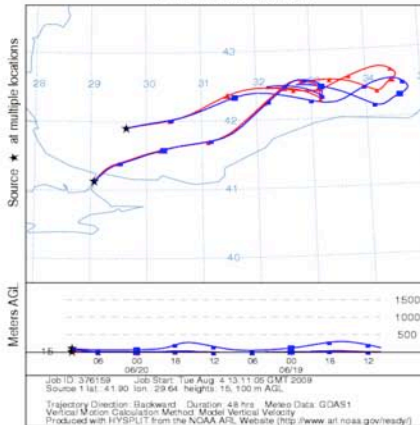
### STIVc

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Backward trajectories ending at 1700 UTC 19 Jun 06  
GDAS Meteorological Data



### T25

NOAA HYSPLIT MODEL  
Backward trajectories ending at 1000 UTC 20 Jun 06  
GDAS Meteorological Data



## **SUPPLEMENT C: ANALYTICAL PROCEDURE**

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TABLE S3: Instrumental detection limits (IDLs)

FIGURE S4: Recoveries (in %) of the analytical procedure for the two different types of matrices

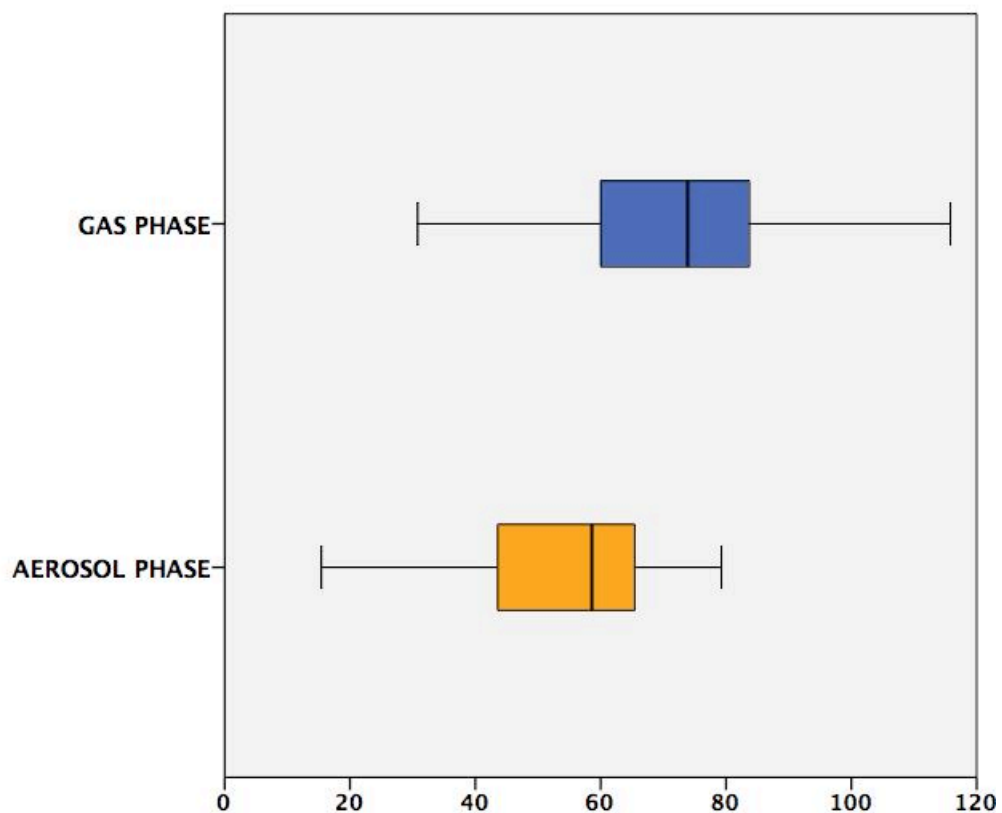
TABLE S4: Field and procedural blanks for each sample matrix.

**TABLE S3: Instrumental detection limits (IDLs) were determined from the lowest standard in the calibration curve. A mean air sampling of 500 m<sup>3</sup> and 0.05 g of particles per filter was applied to derived IDLs.**

	<b>GAS PHASE SAMPLES</b>	<b>AEROSOL SAMPLES</b>
<b>UNITS</b>	<b>pg m<sup>-3</sup></b>	<b>ng g<sup>-1</sup></b>
<b>HCB</b>	0,007	0,069
<b>PCB17</b>	0,012	0,124
<b>PCB18</b>	0,071	0,710
<b>PCB28</b>	0,015	0,148
<b>PCB31</b>	0,001	0,010
<b>PCB33</b>	0,060	0,597
<b>PCB44</b>	0,010	0,102
<b>PCB49</b>	0,054	0,540
<b>PCB52</b>	0,004	0,040
<b>PCB70</b>	0,003	0,034
<b>PCB74</b>	0,007	0,068
<b>PCB82</b>	0,010	0,097
<b>PCB87</b>	0,006	0,055
<b>PCB95</b>	0,023	0,234
<b>99/101</b>	0,161	1,608
<b>105/132</b>	0,008	0,082
<b>PCB110</b>	0,002	0,018
<b>PCB118</b>	0,022	0,220
<b>PCB128</b>	0,019	0,190
<b>PCB138</b>	0,010	0,103
<b>PCB149</b>	0,003	0,026
<b>PCB151</b>	0,003	0,031
<b>PCB153</b>	0,090	0,897
<b>156/171</b>	0,038	0,375
<b>PCB158</b>	0,003	0,032
<b>PCB169</b>	0,004	0,043
<b>PCB170</b>	0,010	0,105
<b>PCB177</b>	0,006	0,064
<b>PCB180</b>	0,004	0,039
<b>PCB183</b>	0,019	0,193
<b>PCB187</b>	0,007	0,067
<b>PCB191</b>	0,009	0,091
<b>PCB194</b>	0,002	0,020
<b>PCB195</b>	0,006	0,065
<b>201/199</b>	0,056	0,557
<b>PCB205</b>	0,006	0,061
<b>PCB206</b>	0,055	0,551
<b>PCB208</b>	0,077	0,768
<b>PCB209</b>	0,056	0,559



**FIGURE S4: Recoveries (in %) of the analytical procedure for the two different types of matrices.**



The box plot graph represents the minimum, the lowest quartile, the median, the upper quartile and the maximum value of the % of recovery of each sample matrix.



The calculated values for each matrix are referred to the average amount sampled: 500 m<sup>3</sup> for gas phase samples and 0,05 g of particles for aerosol samples. One blank was analysed every five samples (the number of analysed blanks for each matrix is indicated below). Samples were corrected using the mean blank value shown in the table.

## SUPPLEMENT D: ATMOSPHERIC OCCURRENCE OF PCBS AND HCB

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TABLE S5: Organochlorine compound concentrations in gas phase samples given in  $\text{pg m}^{-3}$

TABLE S6: Organochlorine compound concentrations in aerosol phase samples given in  $\text{pg m}^{-3}$

TABLE S7: Organochlorine compound concentrations in aerosol phase samples given in  $\text{ng g}^{-3}$

TABLE S8: Detailed concentrations of mono-ortho-substituted PCB congener concentrations in gas phase samples given in  $\text{pg m}^{-3}$

TABLE S9: Detailed concentrations of mono-ortho-substituted PCB congener concentrations in aerosol samples given in  $\text{pg m}^{-3}$

FIGURE S5: Spatial distribution of the  $\sum_{7\text{ICES}}\text{PCB}$  congeners in gas phase samples in  $\text{pg m}^{-3}$

FIGURE S6: Spatial distribution of  $\sum_{7\text{ICES}}\text{PCB}$  congeners in aerosols given in  $\text{pg m}^{-3}$  (upper panel) and  $\text{ng g}^{-1}$  (lower panel).

FIGURE S7: Spatial distribution of mono-ortho-substituted PCB congeners in the gas phase samples in  $\text{pg m}^{-3}$

FIGURE S8: Spatial distribution of mono-ortho-substituted PCB congeners in the aerosol samples in  $\text{pg m}^{-3}$

FIGURE S9: Spatial distribution of HCB isomers in the gas phase samples in  $\text{pg m}^{-3}$

**TABLE S5: Organochlorine compound concentrations in gas phase samples given in  $\text{pg m}^{-3}$ .**

	T1	T2	STIa	STIb	T4	STIIa	STIIb	T5	T6	T7	T8	T9	T10	STIIIb	T11	T12	T13	T14	T15	T17	T18	T19	T20	T21	T22	T23	STIVa	STIVb	STIVc	T25
<b><math>\alpha</math>-HCH</b>	6,80	24,10	15,29	4,56	6,28	21,13	14,19	15,88	0,51	-	-	38,20	7,10	32,44	-	12,74	3,59	87,55	19,27	8,77	29,07	84,83	36,00	48,92	38,83	33,22	73,14	48,40	16,42	74,21
<b><math>\gamma</math>-HCH</b>	41,66	296,08	11,20	2,74	25,28	124,87	20,91	22,55	6,09	133,75	0,88	16,38	59,57	13,61	-	63,73	67,51	127,36	41,25	24,88	-	3,31	4,33	4,89	22,88	-	60,11	13,95	4,65	12,33
<b><math>\delta</math>-HCH</b>	375,61	305,46	-	-	7,97	47,49	-	-	-	-	-	-	12,38	-	30,57	13,57	9,89	-	9,39	29,23	48,37	-	-	-	-	44,89	-	-	-	-
<b>HCB</b>	87,85	200,92	3,66	2,43	59,34	418,30	50,93	23,33	36,56	-	25,14	61,77	178,54	16,01	-	8,80	5,16	50,17	30,97	7,16	73,14	25,19	10,85	11,23	46,08	106,11	57,46	100,23	13,72	26,55
<b>PCB 17</b>	24,44	2,07	26,41	-	0,59	5,21	29,42	1,55	6,58	1,94	-	0,85	10,14	8,28	-	17,98	6,75	-	12,17	-	3,24	9,18	3,86	4,14	11,48	-	15,85	0,11	70,13	4,68
<b>PCB 18</b>	37,85	57,72	14,94	0,89	-	14,85	23,43	-	11,97	1,16	-	1,64	8,10	22,71	9,04	170,30	9,37	-	115,71	-	6,66	16,32	9,86	14,77	15,14	-	8,72	-	43,92	15,15
<b>PCB 28</b>	-	-	10,36	-	2,65	-	45,54	3,62	0,88	16,89	-	8,93	8,03	20,29	-	3,45	4,29	-	-	-	-	72,82	22,15	8,87	27,56	-	14,41	6,00	53,99	22,48
<b>PCB 33</b>	34,43	52,13	-	7,72	10,90	26,35	48,35	24,42	9,49	9,46	11,01	-	8,27	44,33	10,03	-	12,51	-	-	-	11,75	22,04	10,42	8,78	48,56	13,17	-	11,27	-	80,43
<b>PCB 49</b>	15,73	27,54	27,62	4,27	4,70	8,66	15,79	9,96	-	4,14	-	2,66	14,81	47,73	-	10,09	-	12,90	-	-	18,01	29,47	3,45	-	35,19	0,97	-	-	14,19	36,19
<b>PCB 52</b>	41,99	100,97	-	7,36	8,20	64,39	24,46	13,40	-	-	14,99	-	6,85	53,68	2,67	-	21,68	15,94	-	12,64	16,32	50,92	12,21	19,18	-	11,02	-	14,63	-	
<b>PCB 74</b>	-	62,12	9,37	-	-	30,80	19,74	7,89	-	-	-	-	-	-	5,09	19,60	11,78	24,37	6,83	7,44	7,16	-	-	-	14,18	16,92	-	-	21,65	10,15
<b>PCB 82</b>	-	10,10	15,48	52,42	4,62	-	19,19	9,88	7,23	15,70	21,51	-	74,29	-	-	0,36	-	-	-	-	10,04	29,04	7,07	3,28	27,69	-	21,15	8,05	109,27	10,09
<b>PCB 87</b>	9,05	-	1,17	-	-	6,10	-	-	-	5,67	-	-	-	-	59,40	85,25	-	-	1,98	-	1,71	4,48	1,98	-	1,38	-	7,17	-	-	-
<b>99/101</b>	17,97	123,11	22,85	0,57	3,52	43,51	44,25	16,22	-	8,65	0,50	5,36	-	22,77	7,30	24,25	-	1,52	0,02	1,20	28,91	40,40	12,05	7,35	46,65	-	32,19	13,28	12,62	
<b>105/132</b>	-	105,45	-	-	-	15,92	-	-	6,61	3,82	6,00	-	-	-	35,97	24,31	26,18	34,30	21,21	18,60	-	7,65	4,70	3,37	11,85	-	14,36	8,83	-	10,09
<b>PCB 110</b>	-	-	8,06	3,32	1,89	5,73	9,01	1,78	3,08	5,19	7,28	3,77	7,07	11,07	1,77	2,44	-	1,67	1,89	-	-	18,67	8,18	23,42	16,11	3,93	16,71	5,77	12,85	7,83
<b>PCB 118</b>	-	15,78	4,90	4,27	4,28	14,91	-	-	4,31	3,11	5,14	-	2,68	10,56	14,78	11,24	-	2,96	0,97	-	6,81	10,58	4,36	5,64	6,08	17,80	3,93	7,25	-	5,44
<b>PCB 128</b>	-	-	-	-	-	-	-	-	4,00	-	6,12	-	2,15	-	1,72	3,36	1,25	1,43	0,65	0,80	1,63	-	3,26	-	-	-	4,47	-	4,54	6,89
<b>PCB 138</b>	26,26	56,01	7,68	7,35	1,24	20,76	14,84	13,05	26,49	18,10	33,69	12,79	19,69	12,33	14,98	13,95	14,91	22,94	9,38	7,40	22,47	15,48	10,65	5,73	24,05	17,00	42,22	20,03	22,02	23,21
<b>PCB 149</b>	26,27	98,95	17,79	9,19	9,83	43,40	19,49	23,55	22,02	13,85	26,46	11,61	13,60	21,27	10,84	15,37	12,16	23,51	15,19	7,05	27,55	14,94	10,42	8,02	26,84	-	56,79	25,93	17,48	20,97
<b>PCB 151</b>	44,62	7,31	0,54	-	-	0,57	1,64	-	-	11,37	-	0,03	2,66	5,85	-	1,71	-	-	-	-	4,25	20,68	-	1,23	6,81	-	6,64	-	3,91	0,71
<b>PCB 153</b>	35,19	68,36	-	7,42	9,48	29,59	19,87	21,80	27,59	18,63	39,21	14,79	22,38	14,96	20,17	18,47	15,15	22,48	10,13	7,41	21,18	16,74	8,36	6,86	20,25	24,78	45,36	24,00	35,16	26,69
<b>156/171</b>	-	-	-	-	-	-	-	-	-	-	16,37	-	-	-	-	-	-	-	-	-	-	-	-	9,77	-	-	-	-	-	-
<b>PCB 158</b>	-	-	-	-	-	-	4,16	-	-	-	4,48	-	-	-	-	1,94	-	1,91	-	0,28	-	-	-	-	-	-	3,28	-	2,31	1,78
<b>PCB 169</b>	-	-	-	-	-	-	-	-	18,73	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<b>PCB 170</b>	8,63	2,33	-	3,69	2,01	6,95	5,63	6,90	6,33	7,19	17,84	3,16	15,01	9,15	5,87	3,66	4,11	4,07	1,97	1,89	11,56	10,76	9,72	5,39	12,80	11,25	11,95	7,86	14,08	14,40
<b>PCB177</b>	-	1-	3,42	4,03	2,66	9,40	4,74	7,79	12,17	5,98	14,97	5,57	10,71	-	4,68	3,61	3,08	4,00	2,26	1,68	7,17	3,02	6,69	3,59	12,31	8,83	11,58	6,24	12,31	10,50
<b>PCB 180</b>	16,68	18,26	7,04	-	-	-	8,39	18,57	36,42	20,25	51,31	14,45	41,05	16,61	17,51	14,46	13,38	17,24	8,27	7,23	35,86	27,47	25,92	15,52	23,05	29,12	36,65	21,02	42,14	28,93



**TABLE S6: Organochlorine compound concentrations in aerosol phase samples given in pg m<sup>-3</sup>**

	T1	T2	T3	STIb	T4	STIIb	T5	T6	T7	T8	T9	T10	STIIIa	STIIb	T11	T12	T13	T14	T15	T16	T17	T18	T20	T21	T22	T23	T24	STIVb	STIVc	T25	
HCB	0,11	0,28	0,13	0,27	0,21	-	0,29	0,06	0,15	0,16	-	0,19	0,26	0,18	-	0,10	-	-	-	-	-	0,28	0,19	0,07	-	0,24	0,72	-	-	-	
PCB 17	-	-	0,12	-	-	-	-	-	-	-	-	0,58	-	-	-	-	-	-	-	-	-	-	0,57	-	-	0,40	-	-	-	-	
PCB 18	-	-	0,03	0,92	0,49	-	0,55	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
PCB 28	-	-	0,21	0,47	0,31	0,43	-	0,58	0,41	0,63	-	0,29	0,40	-	-	-	-	-	0,93	0,54	-	-	0,32	0,42	0,58	0,26	-	-	-	-	
PCB 31	-	-	0,25	0,69	0,52	0,65	0,62	0,24	1,69	-	-	-	0,34	-	-	0,20	0,29	-	-	-	-	-	-	0,31	-	0,37	-	-	-	-	
PCB 33	-	-	0,06	-	0,38	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0,20	-	0,36	-	-	-	-	
PCB 44	0,08	-	0,10	1,53	1,16	1,71	1,24	0,01	-	-	0,63	0,37	0,41	-	-	-	0,47	-	-	-	-	-	0,36	0,23	-	0,23	-	0,56	-	0,39	
PCB 49	-	-	0,10	2,80	1,67	2,30	1,60	-	-	-	0,11	0,45	0,12	-	-	-	0,07	-	-	-	-	-	0,40	-	-	0,12	-	-	-	-	
PCB 52	0,98	-	-	-	3,50	3,34	-	1,09	1,25	1,59	1,85	1,01	1,31	0,95	-	-	0,84	-	-	-	-	1,35	0,95	0,74	-	0,82	2,93	1,96	1,15	0,76	
PCB 70	-	-	-	1,97	1,35	1,63	1,42	1,03	3,26	-	0,84	0,53	0,65	0,68	-	-	0,54	-	-	-	-	0,52	0,51	0,65	0,85	0,38	-	0,80	0,92	0,45	
PCB 82	0,08	0,54	-	1,61	1,04	1,68	1,22	0,24	-	-	0,67	0,28	0,48	0,16	0,05	0,01	0,37	0,01	-	0,06	-	-	0,23	0,19	0,15	0,09	-	0,66	0,65	0,41	
PCB 87	-	-	-	0,63	0,45	0,83	0,65	-	-	-	0,08	-	0,09	-	-	-	0,21	-	-	-	-	0,05	-	-	-	-	-	-	0,15	-	
PCB 95	-	0,68	-	3,11	2,16	3,10	2,73	-	-	-	0,89	0,65	0,63	0,13	0,17	-	0,61	-	-	0,28	-	0,28	0,57	-	0,34	0,12	-	0,66	0,47	0,34	
99/101	-	1,84	-	7,30	5,08	7,90	6,74	-	-	-	3,07	1,72	1,91	0,98	0,15	0,05	1,91	0,01	-	0,41	-	1,64	1,12	-	1,81	0,62	0,83	3,01	2,73	1,50	
105/132	1,51	0,75	0,17	1,61	0,81	1,40	0,99	1,30	1,26	2,15	0,78	0,51	0,75	0,47	-	-	0,43	0,37	-	-	0,15	-	0,35	0,92	0,54	0,28	-	1,08	1,03	0,65	
PCB 110	0,32	0,27	0,31	1,06	0,35	0,39	0,37	-	0,34	0,02	0,35	0,24	0,20	0,20	0,07	0,09	0,13	0,18	1,30	0,40	0,09	0,22	0,15	0,35	0,23	0,23	-	0,25	0,13	0,24	
PCB 118	-	0,81	-	1,92	0,86	1,90	1,33	-	-	-	0,59	0,10	0,21	-	-	0,11	0,21	0,17	0,02	0,33	-	0,06	0,03	-	0,42	0,13	-	0,77	1,03	0,29	
PCB 128	-	0,74	0,03	-	0,22	0,31	0,27	-	0,66	-	-	-	-	-	0,16	0,15	-	0,46	1,00	0,62	-	-	-	-	-	-	-	-	0,34	-	
PCB 138	3,74	3,05	0,12	1,52	0,85	1,71	1,59	0,69	1,40	0,83	1,62	0,64	0,92	0,39	0,28	0,20	0,65	1,28	1,23	1,06	0,04	0,70	0,25	0,33	1,12	0,24	-	2,20	2,82	0,75	
PCB 149	1,42	2,22	0,29	2,36	1,25	2,65	2,13	0,61	1,23	0,74	1,45	0,58	0,91	0,47	-	0,07	0,59	0,52	0,06	0,42	0,03	0,92	0,39	0,45	1,08	0,50	-	1,59	1,67	0,65	
PCB 151	0,13	0,60	0,01	1,07	0,52	1,24	0,58	-	0,05	0,03	0,43	0,20	0,38	0,05	-	-	0,12	0,09	-	0,17	-	0,29	0,15	0,08	0,20	0,08	-	0,47	0,36	-	
PCB 153	2,87	3,00	0,15	1,37	0,79	1,46	1,29	1,10	0,95	0,98	1,44	0,58	0,74	0,25	0,27	0,19	0,57	1,23	1,39	1,16	0,05	1,13	0,25	0,35	1,01	0,30	0,50	1,97	2,14	0,42	
156/171	2,30	-	-	-	0,46	-	0,92	0,91	0,80	0,96	1,04	-	0,69	0,66	0,25	-	0,38	0,70	-	-	-	0,61	0,41	1,29	0,76	0,42	-	-	0,40	-	
PCB 158	-	0,26	-	-	0,17	0,22	0,20	-	0,21	-	-	-	-	0,13	-	-	-	-	-	-	-	-	-	-	-	0,22	-	-	0,28	0,33	0,12
PCB 169	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
PCB 170	4,37	2,36	0,17	-	0,23	-	0,35	1,01	-	1,37	0,60	1,46	-	0,18	0,27	0,21	0,35	1,52	1,94	0,79	0,13	0,86	0,34	0,61	0,41	0,23	-	0,54	0,55	0,42	
PCB177	2,05	1,13	0,08	-	0,25	-	-	0,77	0,94	0,55	-	0,30	0,22	-	-	-	0,21	0,48	-	-	-	0,32	-	-	-	0,23	-	-	-	-	
PCB 180	10,30	4,77	0,46	0,72	0,37	0,66	-	-	-	2,90	1,68	2,72	-	-	0,57	0,46	0,66	2,99	4,16	1,49	0,31	-	0,61	-	1,18	-	3,26	1,65	1,66	0,80	





**TABLE S7: Organochlorine compound concentrations in aerosol phase samples given in ng g<sup>-1</sup>**

	T1	T2	T3	STIb	T4	STIb	T5	T6	T7	T8	T9	STIIa	STIIb	T11	T12	T13	T14	T15	T16	T17	T20	T21	T22	T23	T24	STIVb	STIVc	T25
<b>HCB</b>	1,15	6,72	2,36	1,72	1,39	-	2,42	0,56	0,82	2,20	-	1,87	0,76	-	1,17	-	-	-	-	-	2,77	0,77	-	2,80	6,70	-	-	-
<b>PCB 17</b>	-	-	2,21	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	8,42	-	-	4,64	-	-	-	-
<b>PCB 18</b>	-	-	0,53	5,96	3,29	-	4,60	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<b>PCB 28</b>	-	-	3,73	3,06	2,11	2,26	-	5,27	2,23	8,98	-	2,92	-	-	-	-	6,53	7,61	-	4,75	4,71	3,43	3,04	-	-	-	-	-
<b>PCB 31</b>	-	-	4,48	4,45	3,45	3,43	5,21	2,17	9,20	-	-	2,49	-	-	2,45	2,20	-	-	-	-	-	3,48	-	4,28	-	-	-	-
<b>PCB 33</b>	-	-	1,11	-	2,54	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	2,19	-	4,17	-	-	-	-
<b>PCB 44</b>	0,85	-	1,78	9,89	7,78	9,06	10,45	0,08	-	-	5,97	2,99	-	-	-	3,51	-	-	-	-	5,40	2,54	-	2,68	-	3,90	-	3,58
<b>PCB 49</b>	-	-	1,75	18,07	11,19	12,17	13,42	-	-	-	1,04	0,86	-	-	-	0,52	-	-	-	-	5,98	-	-	1,44	-	-	-	-
<b>PCB 52</b>	9,87	-	-	-	23,42	17,71	-	9,79	6,82	22,51	17,66	9,57	4,07	-	-	6,28	-	-	-	-	14,04	8,22	-	9,51	27,43	13,70	4,21	6,98
<b>PCB 70</b>	-	-	-	12,68	9,06	8,66	11,90	9,31	17,78	-	7,99	4,71	2,92	-	-	4,03	-	-	-	-	7,61	7,27	4,99	4,42	-	5,57	3,37	4,09
<b>PCB 82</b>	0,84	13,02	-	10,42	6,97	8,89	10,26	2,17	-	-	6,38	3,50	0,70	1,38	0,14	2,77	0,10	-	0,84	-	3,47	2,09	0,91	0,99	-	4,61	2,38	3,77
<b>PCB 87</b>	-	-	-	4,08	3,02	4,40	5,45	-	-	-	0,81	0,68	-	-	-	1,53	-	-	-	-	-	-	-	-	-	-	0,56	-
<b>PCB 95</b>	-	16,46	-	20,05	14,45	16,44	22,91	-	-	-	8,47	4,60	0,54	4,86	-	4,53	-	-	3,91	-	8,38	-	2,02	1,37	-	4,61	1,75	3,12
<b>99/101</b>	-	44,43	-	47,13	33,98	41,87	56,63	-	-	-	29,27	13,91	4,19	4,39	0,61	14,27	0,11	-	5,75	-	16,64	-	10,63	7,24	7,80	21,04	10,04	13,68
<b>105/132</b>	15,31	18,03	2,98	10,36	5,41	7,44	8,29	11,75	6,84	30,57	7,40	5,48	2,02	-	-	3,19	3,77	-	-	3,93	5,20	10,28	3,19	3,31	-	7,59	3,79	5,98
<b>PCB 110</b>	3,28	6,56	5,64	6,87	2,35	2,05	3,15	-	1,84	0,22	3,35	1,44	0,86	2,03	1,12	0,98	1,78	9,08	5,70	2,48	2,28	3,93	1,33	2,69	-	1,78	0,48	2,18
<b>PCB 118</b>	-	19,53	-	12,36	5,76	10,09	11,21	-	-	-	5,60	1,55	-	-	1,32	1,58	1,71	0,17	4,60	-	0,46	-	2,49	1,47	-	5,36	3,80	2,65
<b>PCB 128</b>	-	17,93	0,53	-	1,48	1,62	2,28	-	3,61	-	-	-	-	4,70	1,77	-	4,63	6,96	8,82	-	-	-	-	-	-	-	1,24	-
<b>PCB 138</b>	37,76	73,50	2,13	9,78	5,72	9,07	13,33	6,22	7,61	11,82	15,43	6,73	1,69	8,19	2,38	4,87	12,88	8,56	14,94	1,04	3,68	3,71	6,56	2,83	-	15,40	10,35	6,85
<b>PCB 149</b>	14,39	53,59	5,22	15,24	8,40	14,03	17,94	5,51	6,68	10,48	13,85	6,60	2,03	-	0,83	4,44	5,28	0,43	6,01	0,73	5,79	5,01	6,33	5,82	-	11,11	6,12	5,95
<b>PCB 151</b>	1,27	14,41	0,25	6,92	3,50	6,59	4,92	-	0,27	0,41	4,07	2,78	0,20	-	-	0,92	0,90	-	2,42	-	2,22	0,88	1,17	0,88	-	3,31	1,34	-
<b>PCB 153</b>	29,00	72,32	2,73	8,82	5,30	7,71	10,82	9,88	5,19	13,96	13,75	5,41	1,07	7,89	2,25	4,26	12,42	9,72	16,40	1,33	3,71	3,89	5,97	3,47	4,71	13,81	7,86	3,83
<b>156/171</b>	23,26	-	-	-	3,09	-	7,73	8,20	4,33	13,57	9,94	5,04	2,82	7,35	-	2,82	7,02	-	-	-	6,05	14,35	4,48	4,92	-	-	1,46	-
<b>PCB 158</b>	-	6,17	-	-	1,11	1,17	1,72	-	1,14	-	-	-	0,55	-	-	-	-	-	-	-	-	-	1,29	-	-	1,96	1,21	1,14
<b>PCB 169</b>	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<b>PCB 170</b>	44,18	56,92	3,01	-	1,55	-	2,93	9,14	-	19,41	5,69	-	0,77	7,81	2,52	2,62	15,34	13,56	11,19	3,61	5,00	6,79	2,40	2,63	-	3,81	2,03	3,84
<b>PCB177</b>	20,69	27,35	1,43	-	1,71	-	-	6,95	5,14	7,74	-	1,64	-	-	-	1,60	4,81	-	-	-	-	-	-	2,67	-	-	-	-
<b>PCB 180</b>	104,09	114,99	8,18	4,65	2,50	3,47	-	-	-	41,18	16,01	-	-	16,41	5,62	4,92	30,23	29,09	21,13	8,34	9,08	-	6,94	-	30,50	11,57	6,12	7,31



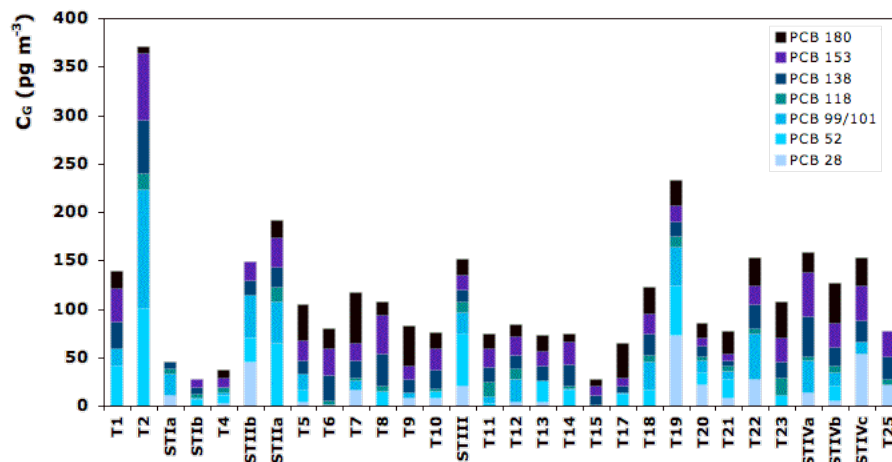
**TABLE S8: Detailed concentrations of mono-ortho-substituted PCB congener concentrations in gas phase samples given in  $\text{pg m}^{-3}$**

	PCB 105	PCB 114	PCB 118	PCB 123	PCB 156	PCB 157	PCB 167	PCB 189
<b>Ti</b>	0.624	0.071	1.528	0.015	0.428	0.039	0.207	0.043
<b>Tii</b>	0.839	0.093	1.916	0.040	0.452	0.057	0.217	0.035
<b>Tiii</b>	0.385	0.049	0.938	0.019	0.325	0.031	0.145	0.040
<b>Tiv</b>	0.380	0.064	0.980	0.036	0.227	0.042	0.104	0.038
<b>Tv</b>	0.436	0.041	1.182	0.020	0.205	0.021	0.092	0.018
<b>Tvi</b>	0.373	0.043	0.954	0.020	0.201	0.020	0.090	0.029
<b>Tvii</b>	0.989	0.142	2.416	0.056	0.374	0.037	0.194	0.040
<b>Tviii</b>	0.693	0.107	1.802	0.038	0.319	0.031	0.168	0.035
<b>Tix</b>	0.681	0.105	1.819	0.053	0.399	0.033	0.183	0.046
<b>Tx</b>	0.674	0.102	1.861	0.037	0.334	0.030	0.173	0.043
<b>Txi</b>	0.608	0.096	1.652	0.029	0.323	0.030	0.176	0.035
<b>Txii</b>	0.401	0.059	1.070	0.016	0.206	0.021	0.099	0.020
<b>Txiii</b>	0.543	0.076	1.407	0.032	0.215	0.021	0.110	0.025
<b>Txiv</b>	1.183	0.098	3.030	0.050	0.421	0.064	0.214	0.026
<b>Txv</b>	0.616	0.061	1.546	0.029	0.281	0.046	0.144	0.022
<b>Txvi</b>	0.454	0.051	1.208	0.021	0.211	0.028	0.108	0.019
<b>Txvii</b>	0.348	0.043	0.976	0.018	0.153	0.021	0.075	0.018
<b>Txviii</b>	0.398	0.047	1.256	0.020	0.159	0.022	0.088	0.016

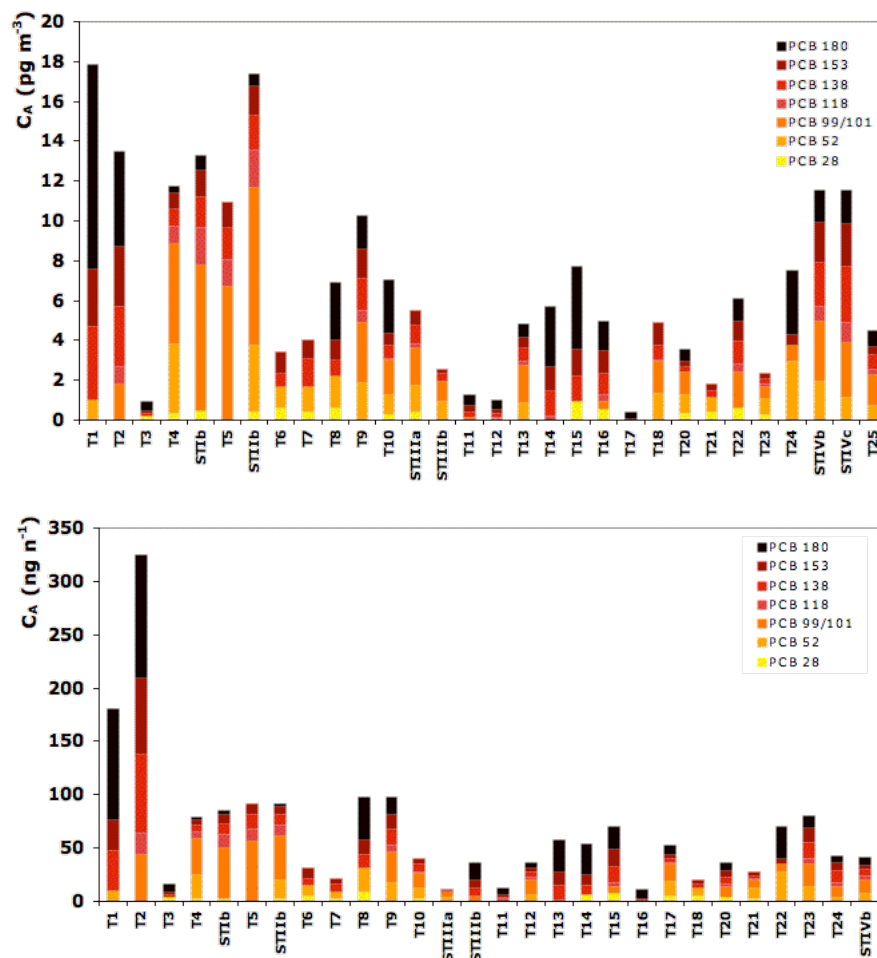
**TABLE S9: Detailed concentrations of mono-ortho-substituted PCB congener concentrations in aerosol samples given in  $\text{pg m}^{-3}$**

	PCB 105	PCB 114	PCB 118	PCB 123	PCB 156	PCB 157	PCB 167	PCB 189
<b>Ti</b>	0,303	0,021	0,825	0,017	0,184	0,025	0,093	0,025
<b>Tii</b>	0,155	0,018	0,394	0,011	0,176	0,013	0,073	0,029
<b>Tiii</b>	0,039	0,005	0,087	0,001	0,035	0,005	0,013	0,006
<b>Tiv</b>	0,078	0,010	0,216	0,004	0,105	0,011	0,042	0,013
<b>Tv</b>	0,121	0,012	0,276	0,002	0,106	0,012	0,045	0,010
<b>Tvi</b>	0,047	0,005	0,117	0,002	0,046	0,005	0,021	0,007
<b>Tvii</b>	0,064	0,009	0,148	0,002	0,053	0,008	0,023	0,009
<b>Tviii</b>	0,136	0,015	0,338	0,003	0,155	0,014	0,066	0,013
<b>Tix</b>	0,148	0,019	0,363	0,006	0,152	0,018	0,068	0,018
<b>Tx</b>	0,035	0,002	0,087	0,002	0,029	0,005	0,016	0,003
<b>Txi</b>	0,106	0,012	0,284	0,002	0,118	0,013	0,057	0,011
<b>Txii</b>	0,090	0,010	0,221	0,004	0,092	0,009	0,038	0,009
<b>Txiii</b>	0,023	0,002	0,063	0,001	0,022	0,002	0,009	0,003
<b>Txiv</b>	0,024	0,002	0,053	0,001	0,017	0,003	0,007	0,004
<b>Txv</b>	0,018	0,001	0,050	-	0,013	0,004	0,007	0,004
<b>Txvi</b>	0,011	-	0,029	-	0,011	0,002	0,005	0,002
<b>Txvii</b>	0,018	0,002	0,049	0,001	0,017	0,003	0,007	0,006
<b>Txviii</b>	0,023	0,002	0,064	-	0,022	0,004	0,009	0,004

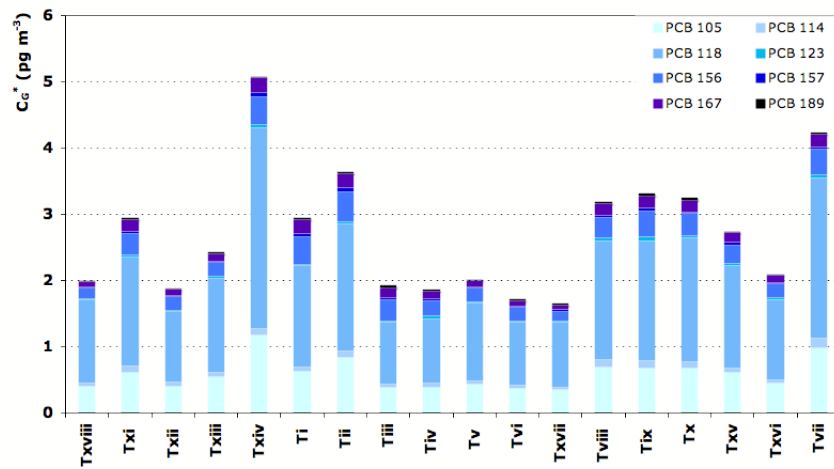
**FIGURE S5: Spatial distribution of  $\sum_{7ICES}PCBs$  in the gas phase samples in  $pg\ m^{-3}$ .**



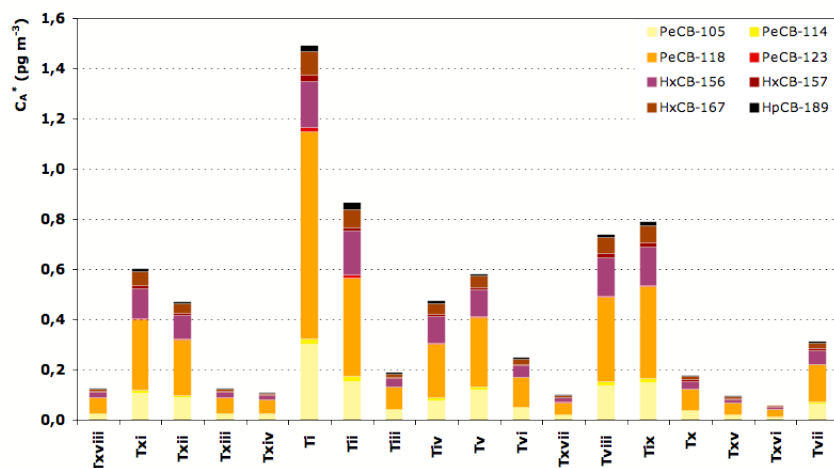
**FIGURE S6: Spatial distribution of  $\sum_{7ICES}PCBs$  in aerosols given in  $pg\ m^{-3}$  (upper panel) and  $ng\ g^{-1}$  (lower panel).**



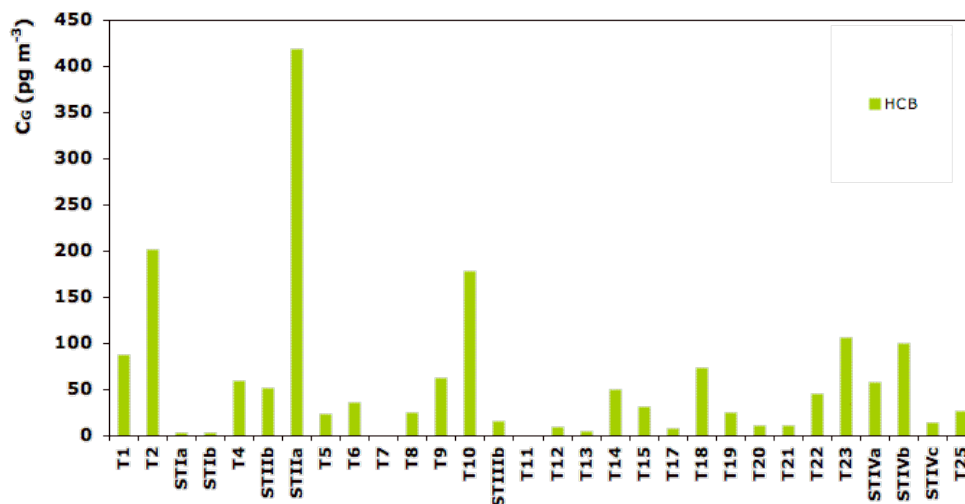
**FIGURE S7: Spatial distribution of mono-ortho-substituted PCB congeners in the gas phase samples in  $\text{pg m}^{-3}$ .**



**FIGURE S8: Spatial distribution of mono-ortho-substituted PCB congeners in the aerosol samples in  $\text{pg m}^{-3}$ .**



**FIGURE S9: Spatial distribution of HCB isomers in the gas phase samples in  $\text{pg m}^{-3}$ .**



## **SUPPLEMENT E: AEROSOL BULK COMPOSITION AND AEROSOL-GAS PARTITIONING OF PCB AND HCB**

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TEXT: PCA analysis for aerosol samples

TABLE S10: Summarize of component loadings (correlations between PCBs and principal component) for aerosol samples.

FIGURE S10: Principal component plot for aerosol phase samples made using normalized volumetric concentrations ( $\text{pg m}^{-3}$ ),

FIGURE S11: Principal component plot for aerosol phase samples generated with particle-normalized concentrations (in  $\text{ng g}^{-1}$ )

FIGURE S12: Dust loadings and dust concentrations

FIGURE S13: Aerosol-particle partitioning of PCBs and HCB. Log-log relationship between aerosol-gas partition coefficient ( $K_P$ ) and octanol-air partitioning coefficient ( $K_{OA}$ ) for PCBs and HCB.  $K_{OA}$  values were corrected for temperature.

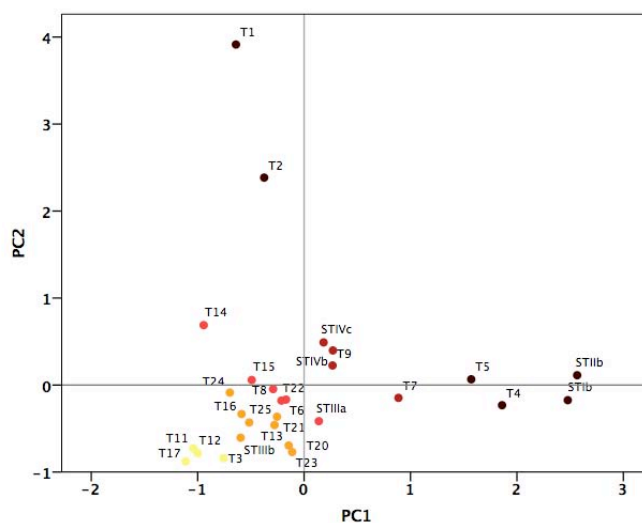
### **TEXT: PCA analysis for aerosol samples**

Principal Component Analysis (PCA) has been used to detect relationships among the quantified variables. PCA has been performed on normalized (subtraction of mean and division by standard deviation) data using statistical software (SPSS 13.0). Principal Components (PC) are formed by linear combinations of the original variables taken as orthogonal to one another. The first factor (PC1) accounts for the maximum amount of variance and subsequent factors successively explain smaller quantities of the original variance.

**TABLE S10: Summary of component loadings (correlations between PCBs and principal component) for aerosol samples.**

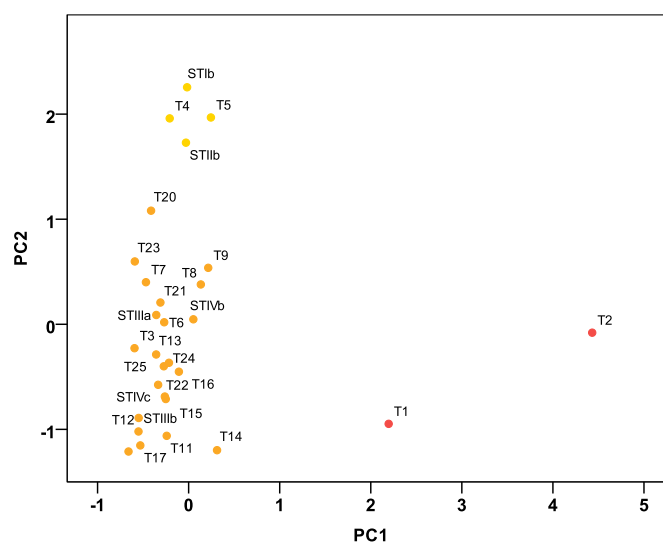
	Component	
	1	2
suma 3cl	0.667	-0.072
suma 4cl	0.868	0.206
suma 5cl	0.864	0.330
suma 6cl	0.257	0.911
suma 7 cl	-0.485	0.824
suma <8cl	-0.447	0.826
tsp	0.590	0.196
T°	0.680	0.004

**FIGURE S10: Principal component plot for aerosol phase samples made using normalized volumetric concentrations ( $\text{pg m}^{-3}$ ),**



The first axis (PC1) explains the maximum amount of variation within the data set (41%) while PC2 explains 30% of the original variance. For gas phase samples, PC1 seems to separate between those samples with higher amount of less chlorinated compounds (congeners with 4 and 5 chlorine atoms). The concentration of 3 chlorinated congeners and the temperature variations seem to have also some influence in this PC. Conversely, PC2 separates samples by the presence of high chlorinated PCBs (congeners with 6, 7 and 8 chlorine atoms).

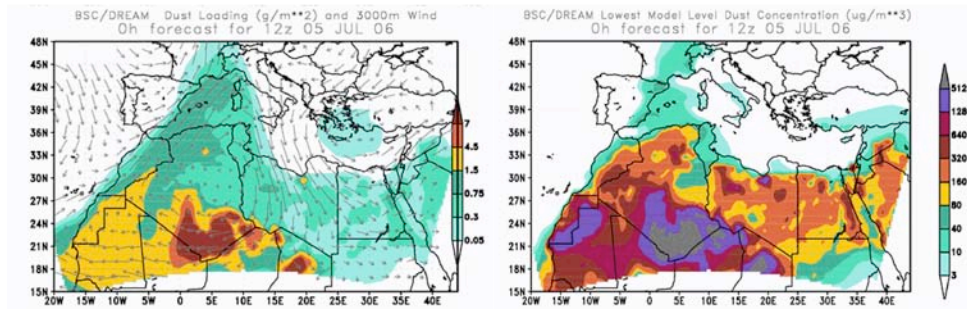
**FIGURE S11: Principal component plot for aerosol phase samples generated with particle-normalized concentrations (in  $\text{ng g}^{-1}$ )**



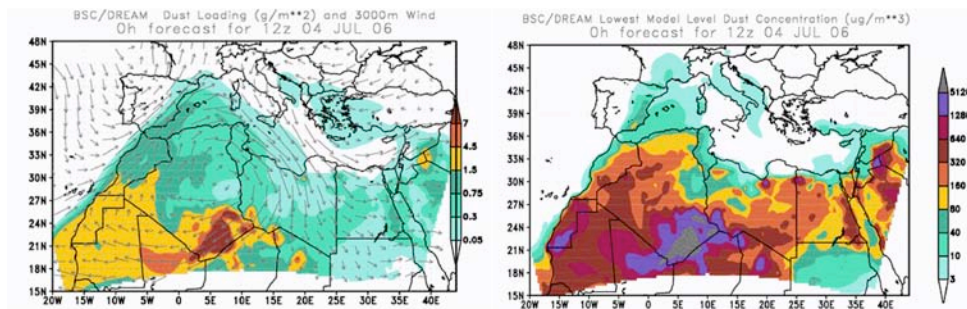


**FIGURE S12: Dust loadings and dust concentrations. Samples subjected to dust intrusions (ST1b, T4, STIIB and T5). T2 and T6 are two examples of samples not affected by Saharan dust loadings.**

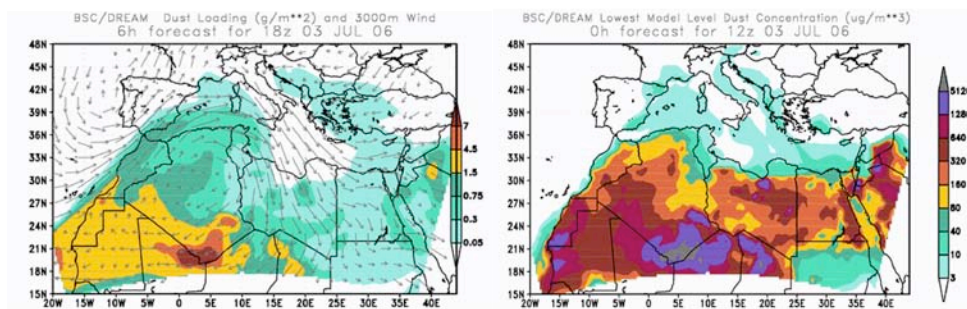
**SAMPLE ST1b**



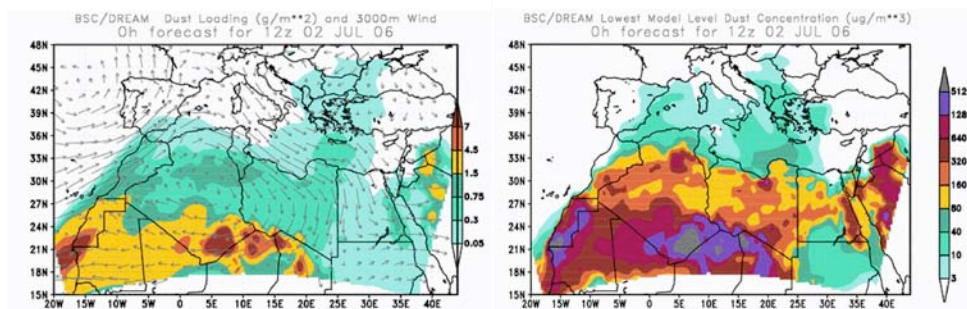
**SAMPLE T4**



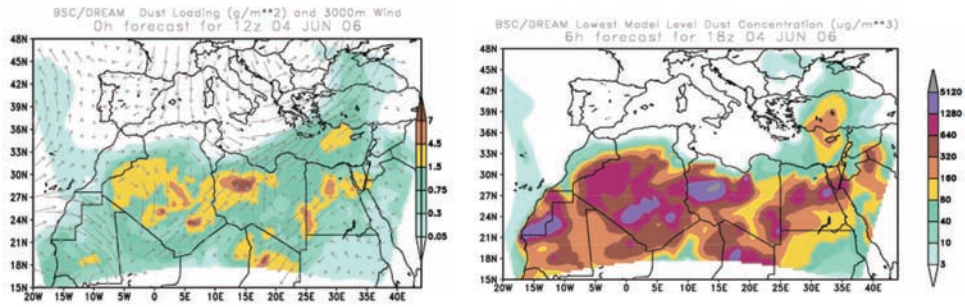
**SAMPLE STIIB**



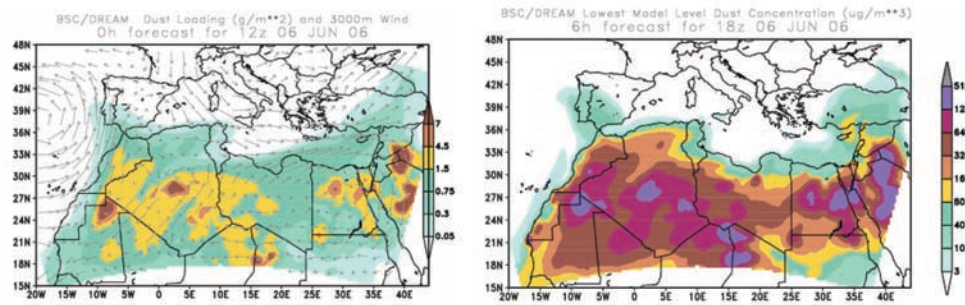
**SAMPLE T5**



### SAMPLE T3

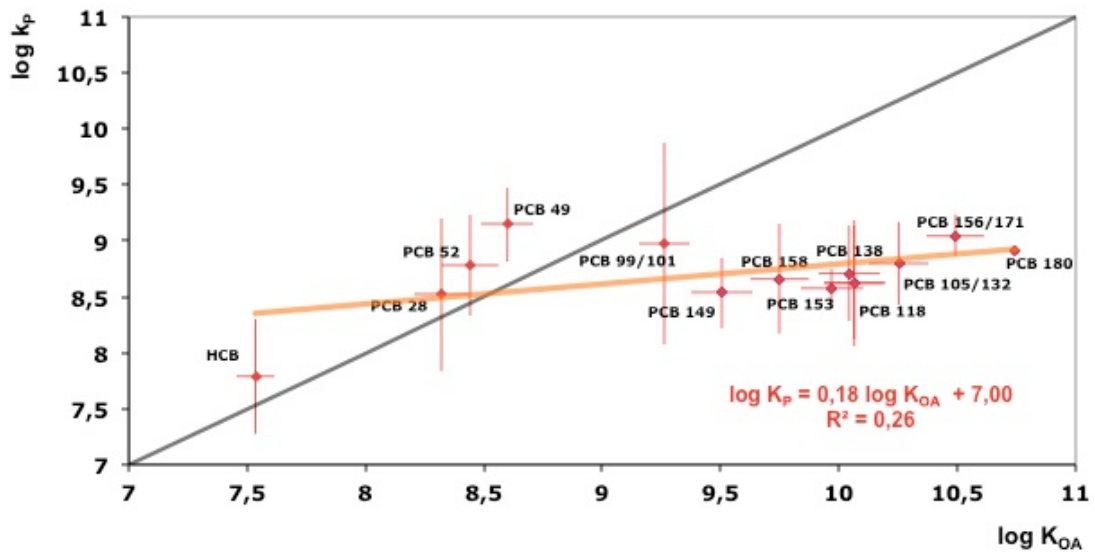


### SAMPLE T6



Source: <http://www.bsc.es/earth-sciences/mineral-dust-forecast-system/bsc-dream8bforecast/north-africa-europe-and-middle-ea-0>

**FIGURE S13: Aerosol-particle partitioning of PCBs and HCB. Log-log relationship between aerosol-gas partition coefficient ( $K_P$ ) and octanol-air partitioning coefficient ( $K_{OA}$ ) for PCBs and HCB.  $K_{OA}$  values where corrected for temperature.**



## **SUPPLEMENT F: FACTORS AFFECTING OCL OCCURRENCE IN MEDITERRANEAN MARINE ATMOSPHERIC ENVIRONMENT**

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TEXT: PCA analysis for gas phase samples

TABLE S11: Summary of component loadings (correlations between PCBs and principal component) for gas phase samples.

FIGURE S14: Principal component plot for gas phase samples generated using normalized data in  $\text{pg m}^{-3}$

TEXT: Clausius-Clapeyron equation

TEXT: atmospheric resident times

TABLE S12: Dry deposition fluxes per sample in  $\text{ng m}^{-2} \text{d}^{-1}$

TABLE S13: detailed air-water fugacity ratios

FIGURE S15: Box-plot of the air-water fugacity ratios for the individual PCB congeners

TABLE S14: Detailed net diffusive air-water net exchange flux in  $\text{ng m}^{-2} \text{d}^{-1}$

FIGURE S16: Plankton biomass in  $\text{mg L}^{-1}$  vs atmospheric gas phase concentrations in  $\text{pg m}^{-3}$  for four PCB congeners. For the time periods with the higher plankton biomass, the gas phase concentrations were at the lower end of those observed.

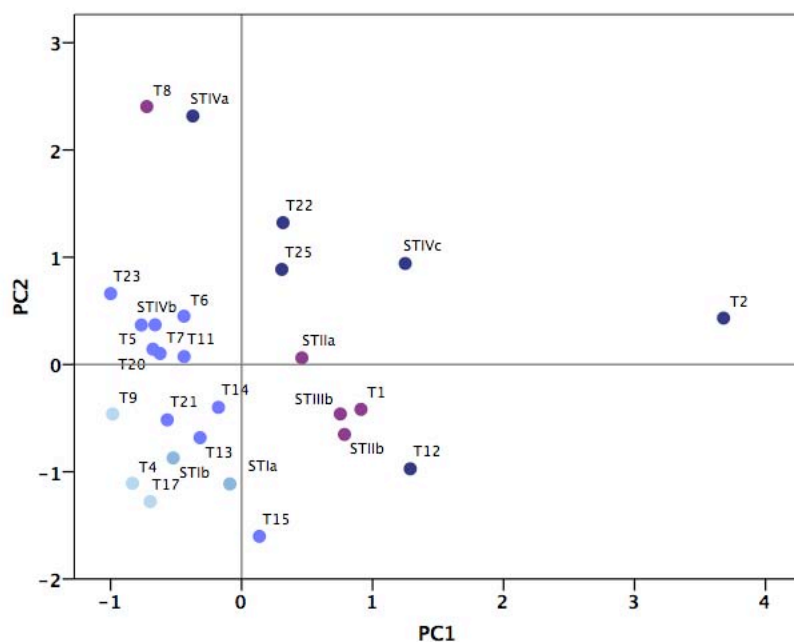
### **TEXT: PCA analysis for gas phase samples**

Principal Component Analysis (PCA) has been used to detect relationships among the quantified variables. PCA has been performed on normalized (subtraction of mean and division by standard deviation) data using statistical software (SPSS 13.0). Principal Components (PC) are formed by linear combinations of the original variables taken as orthogonal to one another. The first factor (PC1) accounts for the maximum amount of variance and subsequent factors successively explain smaller quantities of the original variance.

**TABLE S11: Summary of component loadings (correlations between PCBs and principal component) for gas phase samples.**

	Component		
	1	2	3
suma 3cl	0.725	0.022	0.188
suma 4cl	0.792	-0.04	0.161
suma 5cl	0.787	0.295	-0.029
suma 6cl	0.597	0.626	-0.272
suma 7 cl	0.260	0.910	-0.232
suma <8cl	-0.212	0.709	0.597
tsp	0.129	-0.217	0.700
T°	0.146	0.011	0.764

**FIGURE S14: Principal component plot for gas phase samples generated using normalized data in  $\text{pg m}^{-3}$**



The two first principal components (PC1 and PC2), explaining 35% and 23% of the variability respectively, separate samples in a gradient of concentrations. Samples T4, T9, T17 and STI show the lowest  $C_G$  for  $\Sigma_{41}\text{PCB}$ , while T2, T22, T25, STV and T12 where those samples with the highest  $C_G$ . A third PC of the analysis explaining the 15% of the variation of the samples was highly influenced by the temperature and the total amount of particles present for each sampling period.

## TEXT: Clausius-Clapeyron equation

The partial pressure (P) of a semivolatile compound in air is related to the enthalpy of air-surface exchange ( $\Delta H_{a-w}$ ) by the Clausius-Clapeyron equation:

$$\ln P = \frac{\Delta H_{a-w}}{R} \left( \frac{1}{T} \right) + const$$

where P is the partial pressure of the compound (Pa),  $\Delta H_{a-w}$  is a environmental phase-transition energy of the compound ( $\text{kJ mol}^{-1}$ ), R is the gas constant ( $8.314 \cdot 10^{-3} \text{ Pa m}^3 \text{ mol}^{-1} = \text{kJmol}^{-1}$ ) and T is the temperature (K). Partial pressures of each individual PCB were calculated from measured gas phase concentrations. The Clausius-Clapeyron equation can be expressed graphically as plot of  $\ln P$  vs  $1/T$ . In this study, the temperature did not affect significantly the concentrations of all OCl studied.

	Slope (m)	R <sup>2</sup>	p
HCB	1213	0,002	> 0,05
PCB 17	-11781	0,302	> 0,05
PCB 18	-9569	0,550	> 0,05
PCB 28	-2983	0,018	> 0,05
PCB 33	-18520	0,441	> 0,05
PCB 49	-13208	0,209	> 0,05
PCB 52	2611	0,06	> 0,05
PCB 74	1087	0,02	> 0,05
PCB 75	-1752	0,07	> 0,05
PCB 82	-13227	0,065	> 0,05
PCB87	20114	0,158	> 0,05
PCB 99/101	-21065	0,051	> 0,05
PCB 105/132	-16385	0,228	> 0,05
PCB 128	-25390	0,302	> 0,05
PCB 138	-27058	0,694	> 0,05
PCB 149	-42915	0,896	>0,05
PCB 151	-25484	0,135	> 0,05
PCB 153	-2957	0,017	> 0,05
PCB 177	-2642	0,011	> 0,05
PCB 180	5127	0,053	> 0,05
PCB 183	-5155	0,05	> 0,05
PCB 187	3809	0,041	> 0,05
PCB 191	-1544	0,005	> 0,05
PCB 194	-4066	0,017	> 0,05
PCB 199/201	-1210	0,004	> 0,05

### TEXT: atmospheric resident times

According to equation (5) of the main text, the atmospheric resident times ( $R$ , d) could be calculated as follows:

$$R = \frac{C_{TOTAL}}{F_{OH} + F_{DD} + F_{WD} + F_{AWdep} - F_{AWvol}} \cdot AML \quad (5)$$

where  $C_{TOTAL}$  is the total concentration of POPs in the atmosphere (gas and aerosol phase),  $AML$  is the considered atmospheric mixed layer height, and  $F_{OH}$ ,  $F_{DD}$ ,  $F_{WD}$ ,  $F_{AWdep}$  and  $F_{AWvol}$  are the atmospheric OH degradation flux, atmospheric dry deposition flux, atmospheric wet deposition flux, diffusive deposition flux and diffusive volatilization flux, respectively ( $\text{ng m}^{-2} \text{d}^{-1}$ ). Wet deposition was in this case not considered due to lack of rainfall events during the sampling cruise.  $F_{DD}$  was calculated following equation (1) from the main text, whereas  $F_{OH}$ ,  $F_{AWdep}$  and  $F_{AWvol}$  were parametrized as follows:

$$F_{OH} = (1 - \emptyset) \cdot C_{TOTAL} \cdot r_{OH} \cdot AML \quad (F1)$$

$$F_{AWdep} = k_{AW} \frac{(1 - \emptyset) \cdot C_{TOTAL}}{H^1} \quad (F2)$$

$$F_{AWvol} = (k_{AW} - k_{ADW}) \cdot \frac{(1 - \emptyset) \cdot C_{TOTAL}}{H^1} \quad (F3)$$

where  $\emptyset$  is the fraction of OCl bound to the aerosols in the atmosphere,  $r_{OH}$  is the compound specific OH radical degradation decay rate ( $\text{d}^{-1}$ ) calculated following equation (7) of the main text and  $k_{ADW}$  is the air-deep water mass transfer coefficient estimated as reported elsewhere (Dachs et al., 2002).

Equation (6) from the main text could be derived from equations (5), (1), (F1), (F2) and (F3).



**TABLE S12: Dry deposition fluxes per sample in ng m<sup>-2</sup> d<sup>-1</sup>**

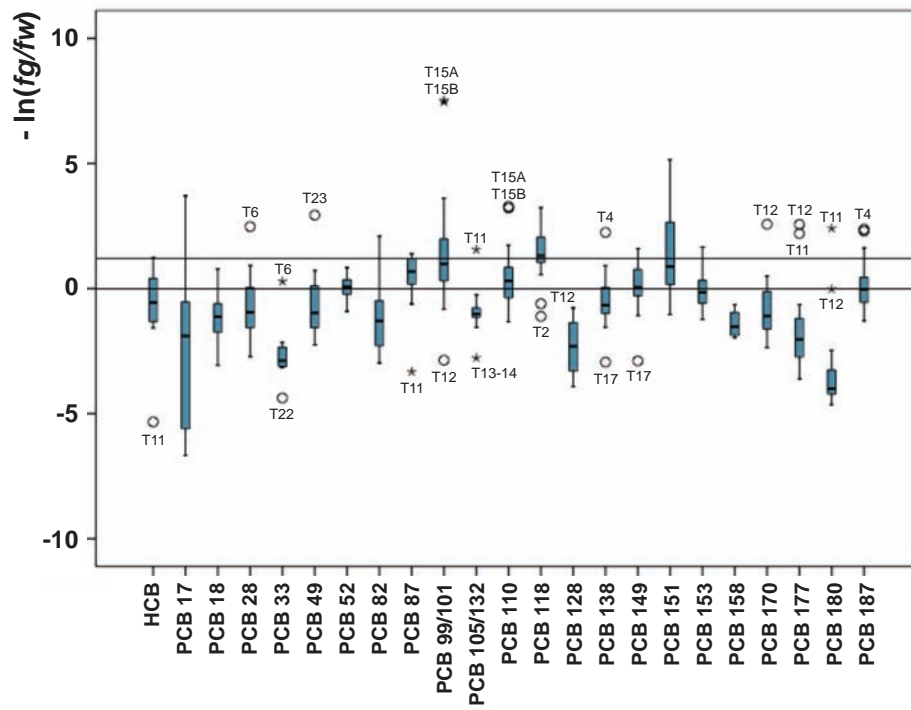
SAMPLE	T1	T2	T3	T4	STIb	T5	STIb	T6	T7	T8	T9	T10	STIHa	STIb	T11	T12	T13	T14	T15	T16	T17	T18	T20	T21	T22	T23	T24	STIVb	STIVc	T25	
<b>VOL (m3)</b>	379	282	441	490	300	353	330	305	262	230	359	447	448	304	947	950	814	414	139	307	848	493	567	679	304	576	373	294	328	371	
<b>WEIGHT (g)</b>	0,0375	0,0117	0,0246	0,0732	0,0465	0,042	0,0623	0,0338	0,0481	0,0162	0,0377	-	0,0615	0,071	0,0327	0,0785	0,109	0,041	0,0199	0,0217	0,0314	-	0,0383	0,0608	0,0517	0,0494	0,0399	0,042	0,0892	0,0406	
<b>mg/m3</b>	0,099	0,041	0,056	0,149	0,155	0,119	0,189	0,111	0,184	0,070	0,105	-	0,137	0,234	0,035	0,083	0,134	0,099	0,143	0,071	0,037	-	0,068	0,090	0,170	0,086	0,107	0,143	0,272	0,109	
<b>HCB</b>	0,020	0,048	0,023	0,036	0,046	0,050	-	0,011	0,026	0,027	-	0,032	0,044	0,031	-	0,017	-	-	-	-	-	0,048	0,032	0,012	-	0,041	0,124	-	-	-	
<b>PCB 17</b>	-	-	0,021	-	-	-	-	-	-	-	-	0,101	-	-	-	-	-	-	-	-	-	-	0,098	-	-	0,069	-	-	-	-	
<b>PCB 18</b>	-	-	0,005	0,085	0,160	0,095	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
<b>PCB 28</b>	-	-	0,036	0,054	0,082	-	0,074	0,101	0,071	0,109	-	0,050	0,069	-	-	-	-	-	0,162	0,093	-	-	0,055	0,073	0,101	0,045	-	-	-	-	
<b>PCB 31</b>	-	-	0,043	0,089	0,119	0,107	0,112	0,042	0,292	-	-	-	0,059	-	-	0,035	0,051	-	-	-	-	-	-	0,054	-	0,063	-	-	-	-	
<b>PCB 33</b>	-	-	0,011	0,066	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0,034	-	0,062	-	-	-	-	
<b>PCB 44</b>	0,015	-	0,017	0,201	0,265	0,215	0,295	0,001	-	-	0,108	0,064	0,071	-	-	-	0,081	-	-	-	-	-	0,063	0,039	-	0,040	-	0,096	-	0,068	
<b>PCB 49</b>	-	-	0,017	0,289	0,484	0,276	0,397	-	-	-	0,019	0,077	0,020	-	-	-	0,012	-	-	-	-	-	0,070	-	-	0,021	-	-	-	-	
<b>PCB 52</b>	0,169	-	-	0,605	-	-	0,578	0,188	0,216	0,274	0,321	0,174	0,227	0,164	-	-	0,145	-	-	-	-	0,232	0,164	0,127	-	0,141	0,507	0,338	0,198	0,132	
<b>PCB 70</b>	-	-	-	0,234	0,340	0,245	0,282	0,178	0,564	-	0,145	0,091	0,112	0,118	-	-	0,093	-	-	-	-	0,089	0,089	0,113	0,147	0,066	-	0,138	0,158	0,077	
<b>PCB 82</b>	0,014	0,093	-	0,180	0,279	0,211	0,290	0,042	-	-	0,116	0,049	0,083	0,028	0,008	0,002	0,064	0,002	-	0,010	-	-	0,041	0,032	0,027	0,015	-	0,114	0,112	0,071	
<b>PCB 87</b>	-	-	-	0,078	0,109	0,112	0,143	-	-	-	0,015	-	0,016	-	-	-	0,035	-	-	-	-	0,009	-	-	-	-	-	-	-	0,026	-
<b>99/101</b>	-	0,319	-	0,877	1,262	1,164	1,366	-	-	-	0,531	0,297	0,330	0,169	0,026	0,009	0,330	0,002	-	0,070	-	0,284	0,194	-	0,312	0,107	0,144	0,519	0,472	0,259	
<b>PCB 95</b>	-	0,118	-	0,373	0,537	0,471	0,536	-	-	-	0,154	0,112	0,109	0,022	0,029	-	0,105	-	-	0,048	-	0,049	0,098	-	0,059	0,020	-	0,114	0,082	0,059	
<b>105/132</b>	0,262	0,129	0,029	0,140	0,277	0,171	0,243	0,225	0,217	0,372	0,134	0,089	0,130	0,081	-	-	0,074	0,065	-	-	0,025	-	0,061	0,159	0,094	0,049	-	0,187	0,178	0,113	
<b>PCB 110</b>	0,056	0,047	0,054	0,061	0,184	0,065	0,067	-	0,058	0,003	0,061	0,042	0,034	0,035	0,012	0,016	0,023	0,030	0,225	0,070	0,016	0,039	0,027	0,061	0,039	0,040	-	0,044	0,023	0,041	
<b>PCB 118</b>	-	0,140	-	0,149	0,331	0,230	0,329	-	-	-	0,102	0,017	0,037	-	-	0,019	0,037	0,029	0,004	0,056	-	0,011	0,005	-	0,073	0,022	-	0,132	0,179	0,050	
<b>PCB 128</b>	-	0,129	0,005	0,038	-	0,047	0,053	-	0,115	-	-	-	-	-	0,028	0,025	-	0,079	0,172	0,108	-	-	-	-	-	-	-	-	-	0,058	-
<b>PCB 138</b>	0,646	0,527	0,020	0,148	0,262	0,274	0,296	0,119	0,241	0,144	0,280	0,111	0,160	0,068	0,049	0,034	0,113	0,220	0,212	0,183	0,007	0,121	0,043	0,057	0,193	0,042	-	0,380	0,487	0,130	
<b>PCB 149</b>	0,246	0,384	0,050	0,217	0,408	0,369	0,458	0,106	0,212	0,128	0,251	0,100	0,157	0,082	-	0,012	0,103	0,090	0,011	0,073	0,005	0,159	0,068	0,078	0,186	0,086	-	0,274	0,288	0,113	
<b>PCB 151</b>	0,022	0,103	0,002	0,090	0,185	0,101	0,215	-	0,009	0,005	0,074	0,035	0,066	0,008	-	-	0,021	0,015	-	0,030	-	0,050	0,026	0,014	0,034	0,013	-	0,082	0,063	-	
<b>PCB 153</b>	0,496	0,518	0,026	0,137	0,236	0,222	0,251	0,189	0,165	0,170	0,249	0,100	0,128	0,043	0,047	0,032	0,099	0,212	0,240	0,200	0,009	0,196	0,043	0,060	0,175	0,051	0,087	0,341	0,369	0,073	
<b>156/171</b>	0,398	-	-	0,080	-	0,159	-	0,157	0,137	0,165	0,180	-	0,120	0,114	0,044	-	0,065	0,120	-	-	-	0,106	0,071	0,222	0,132	0,073	-	-	0,069	-	







**FIGURE S15: Box-plot of the air-water fugacity ratios for the individual PCB congeners**



**TABLE S14: Detailed net diffusive air-water exchange flux in ng m<sup>-2</sup> d<sup>-1</sup>**

SAMPLE	T2	T4	STIb	T6	T7	T8	T9	T10	STIb	T11	T12	T13-T14	T15A	T15B	T17	T18	T19	T20	T21	T22	T23	STIVa	STIVb	STIVc	T25
HCB	-7,09	-0,43	-0,83	-0,67	-	0,24	-0,76	-3,74	0,48	-	-0,60	-0,52	-2,13	-0,22	-0,21	-1,05	-0,09	0,80	0,34	-0,04	-2,50	-1,09	-2,05	0,31	1,96
PCB17	-0,23	-0,04	-2,28	-0,67	-0,15	-	-0,08	-0,91	-0,25	-	-4,51	-0,22	-2,43	-0,31	-	-0,31	-0,91	-0,36	-0,41	-0,98	-	-1,19	0,42	-5,72	0,09
PCB 18	-4,75	-	-1,29	-0,59	-0,09	-	0,15	-0,24	-1,26	-0,40	-32,92	-0,21	-17,47	-2,06	-	-0,19	-1,04	-0,39	-0,92	-0,64	-	-0,15	-	-2,64	-0,82
PCB 28	-	0,20	-2,16	0,65	-0,93	-	-0,30	-0,17	-0,62	-	-0,59	-0,08	-	-	-	-	-3,91	-0,99	0,05	-1,14	0,00	0,63	-0,13	-2,73	-1,00
PCB 33	-5,30	-0,74	-3,35	0,37	-0,86	-0,96	-	-0,67	-2,87	-0,56	-	-0,36	-	-	-	-0,96	-1,86	-0,83	-0,68	-3,67	-1,06	-	-0,89	-	-7,25
PCB 49	-1,79	-0,20	-0,59	-	-0,24	-	-0,15	-0,75	-2,05	-	-1,58	-0,23	-	-	-	-0,52	-0,41	0,28	-	-1,19	1,20	-	-	-0,70	-1,59
PCB 52	-8,08	0,37	0,12	-	-	-0,22	-	0,64	-0,55	-0,11	-	-0,83	-	-	-0,86	-0,10	-2,00	0,21	0,22	-	0,80	-	0,15	0,00	-
PCB 82	-1,05	0,16	-0,88	-0,10	-1,21	-1,66	-	-5,66	-	-	0,96	-	-	-	-	-0,60	-1,62	-0,32	0,74	-1,87	-	-0,94	0,03	-8,17	-0,14
PCB 87	-	-	-	-	-0,13	-	-	-	-	-2,31	-14,93	-	1,12	0,12	-	0,19	0,22	0,18	0,00	0,25	-	0,20	-	-	-
PCB 99/101	-6,21	2,02	2,86	-	1,10	1,52	1,48	-	7,14	1,58	4,55	1,13	7,21	0,73	0,41	0,15	1,60	0,89	3,06	-0,25	-	2,59	2,06	2,10	-
PCB 105/132	-8,68	-	-	-0,09	-0,01	-0,14	-	-	-	-0,71	-1,19	-1,20	-3,18	-0,38	-1,30	-	-0,18	0,20	0,13	-0,18	-	-0,50	-0,31	-	-0,18
PCB 110	-	0,32	0,35	0,22	-0,08	-0,19	0,15	-0,01	0,19	0,18	0,54	0,21	8,47	0,94	-	-	-0,73	0,08	-1,06	-0,32	0,33	0,65	0,62	0,19	1,12
PCB 118	-0,39	2,68	-	0,85	0,48	0,43	-	2,16	3,57	-0,12	-0,25	0,41	3,31	0,33	-	0,68	1,43	0,80	1,65	0,81	1,16	0,86	0,87	-	1,66
PCB 128	-	-	-	-0,24	0,00	-0,35	-	-0,01	-	0,05	-0,13	-0,04	-0,09	-0,01	-0,05	-0,04	-	-0,14	-	-	-	-0,25	-	-0,24	-0,47
PCB 138	-2,90	0,99	0,69	-0,48	-0,03	-0,64	0,37	0,22	2,36	2,40	10,11	0,05	2,24	0,22	-0,28	-0,13	0,46	0,41	0,91	-0,01	0,42	-0,70	0,09	0,02	0,26
PCB 149	-5,10	1,35	0,15	0,07	0,23	-0,25	0,91	0,98	2,18	2,64	10,48	-0,08	1,26	0,12	-0,29	-0,14	0,66	0,62	0,98	0,10	-	-1,26	-0,14	0,21	0,33
PCB 151	-0,31	-	-0,05	-	-0,17	-	0,33	0,27	1,25	-	5,64	-	-	-	-	0,14	0,75	0,00	1,47	0,10	-	0,46	-	0,37	0,61
PCB 153	-2,27	0,77	0,18	-0,10	0,27	-0,25	0,57	0,49	2,58	2,86	12,85	-0,16	0,00	0,00	-0,21	-0,58	0,71	-0,22	1,00	-0,49	0,50	-0,11	0,29	0,02	0,53
PCB 156/191	-	-	-	-	-	-0,39	-	-	-	-	-	-	-	-	-	-	-	-	0,11	-	-	-	-	-	-
PCB 158	-	-	-0,12	-	-	-0,03	-	-	-	-	-0,21	-0,02	-	-	-0,01	-	-	-	-	-	-	0,00	-	0,05	0,14
PCB 169	-	-	-	-1,38	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
PCB 170	-0,08	0,11	-0,08	0,16	-0,21	-0,49	0,12	-0,16	0,01	1,83	8,36	-0,04	0,14	0,01	-0,05	-0,19	-0,06	-0,13	0,09	-0,17	-0,07	-0,10	-0,12	-0,28	-0,23
PCB 177	-0,29	-0,03	-0,09	-0,28	-0,13	-0,34	-0,01	-0,10	-	1,35	6,18	-0,05	-0,13	-0,01	-0,04	-0,12	-0,03	-0,11	-0,05	-0,21	-0,18	-0,24	-0,12	-0,25	-0,26
PCB 180	-0,36	-	-0,06	-0,41	0,07	-0,42	0,10	-0,26	0,29	5,12	23,95	0,01	0,60	0,05	-0,07	-0,40	-0,22	-0,22	-0,01	-0,13	-0,24	-0,27	-0,06	-0,36	-0,11
PCB 183	-0,37	-	-0,07	-0,09	-0,03	-0,10	0,02	-0,01	0,11	1,08	5,09	-0,04	-0,05	-0,01	-0,03	-0,04	0,00	-0,02	0,01	-0,04	-0,01	-0,07	-0,02	-0,04	0,01
PCB 187	-0,87	0,90	0,20	-0,02	0,01	-0,15	0,18	0,13	0,45	2,63	12,27	0,12	1,70	0,15	0,05	0,14	0,05	0,20	0,11	0,15	0,01	-0,04	-	-0,04	0,13

**FIGURE S16: Plankton biomass in  $\text{mg L}^{-1}$  vs. atmospheric gas phase concentrations in  $\text{pg m}^{-3}$  for four PCB congeners. For the time periods with the higher plankton biomass, the gas phase concentrations were at the lower end of those observed.**

