SUPPORTING INFORMATION:

Soil-Air Exchange Controls on Background Atmospheric Concentrations of Organochlorine Pesticides

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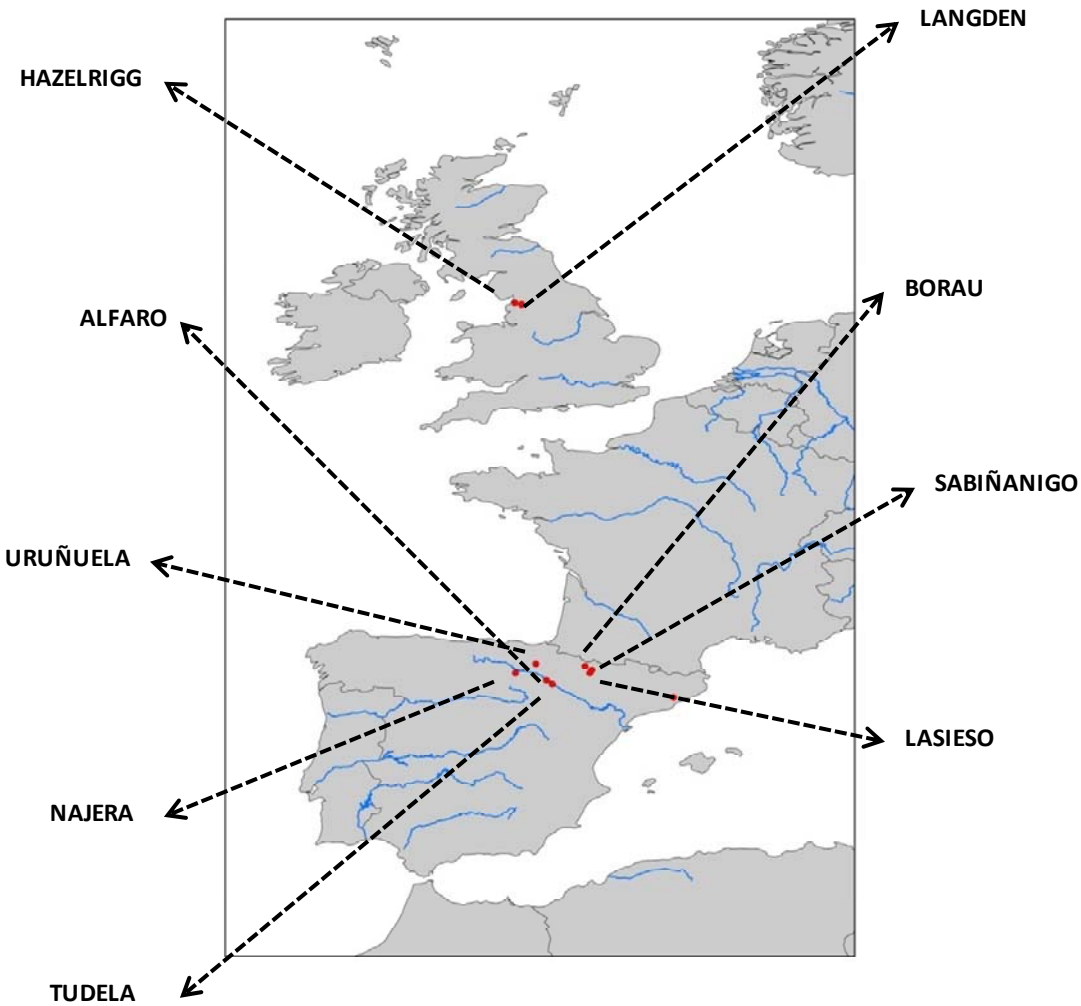

Figure S1. Map of selected background areas

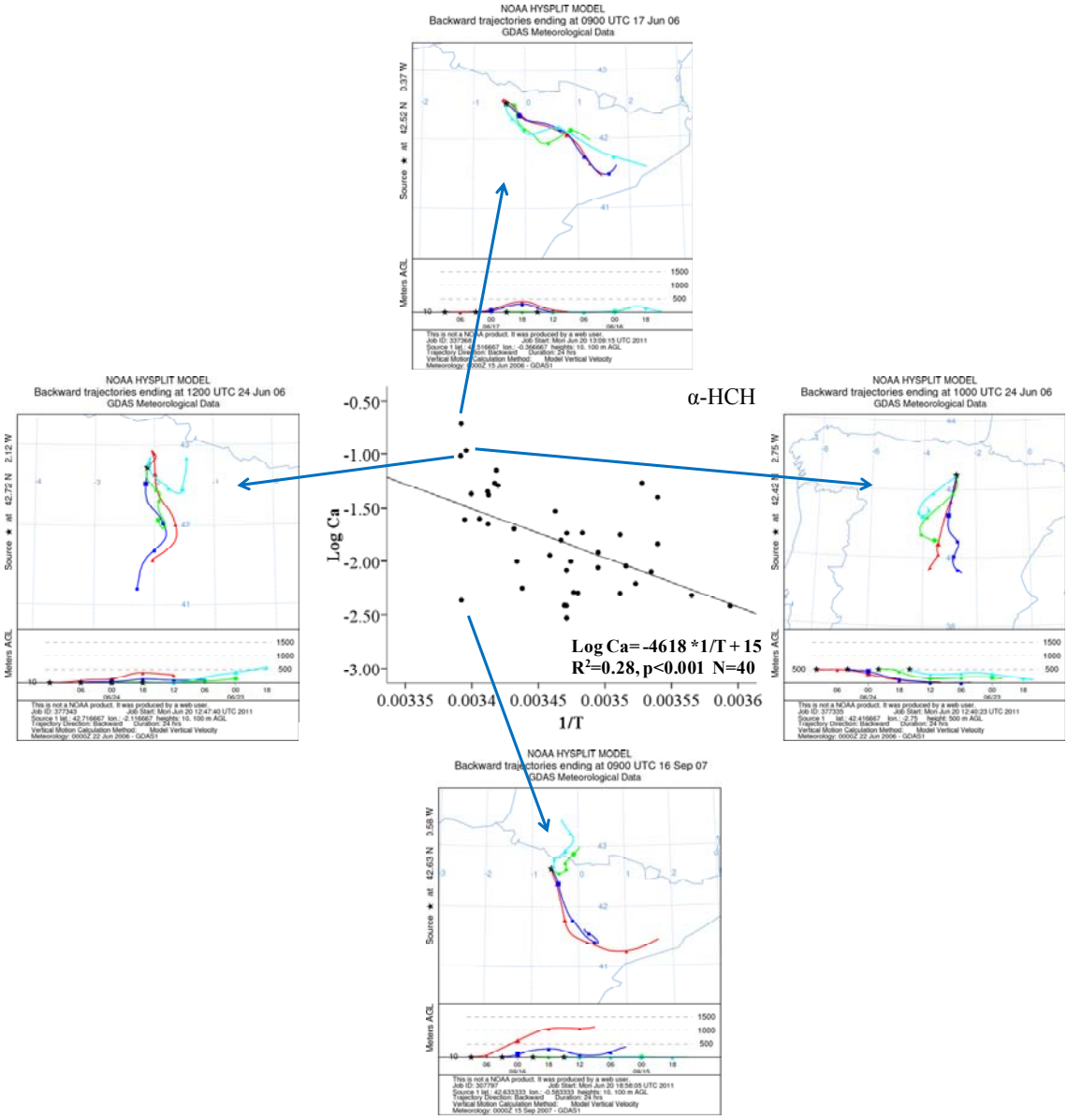

Figure S2a-Examples of back trajectories for selected sites for α-HCH

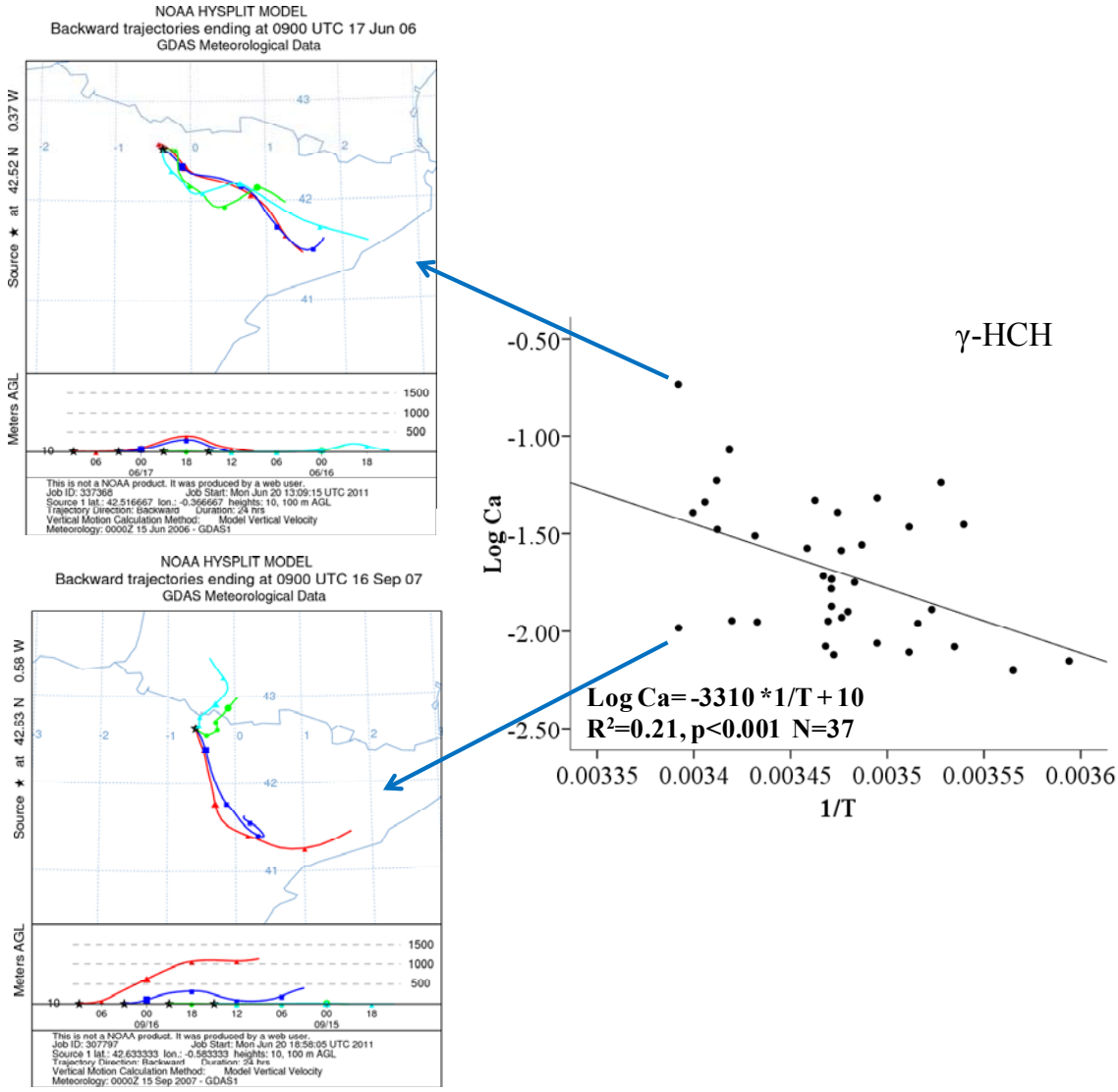

Figure S2b- Examples of back trajectories for selected sites for γ-HCH

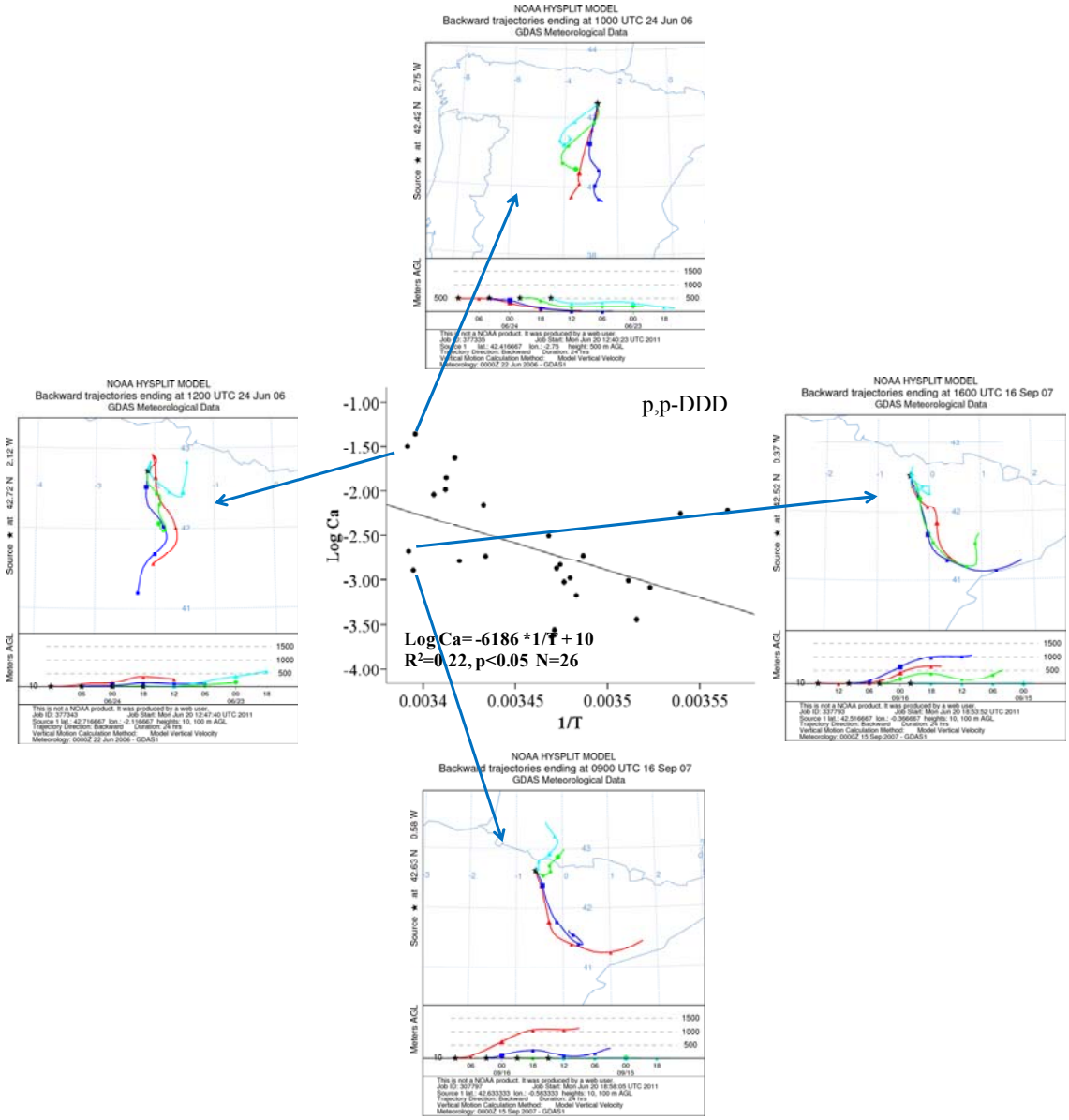

Figure S2c- Examples of back trajectories for selected sites for p,p´-DDD

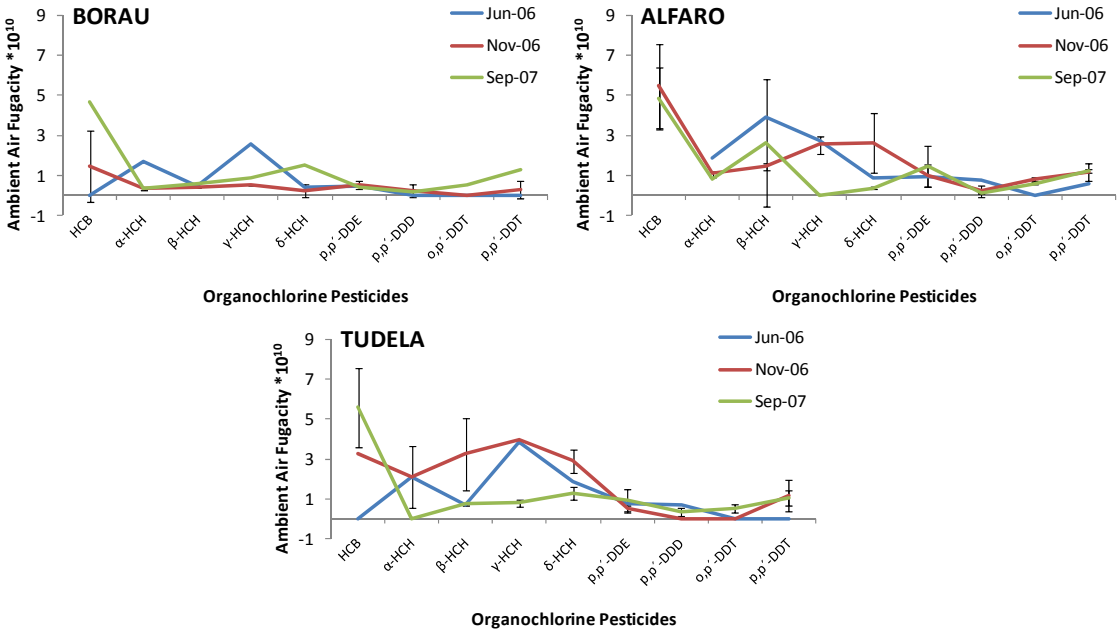

Figure S3. Ambient air fugacity*10^{10} for selected sampling sites of the Ebro river basin (Spain) for Organochlorine Pesticides

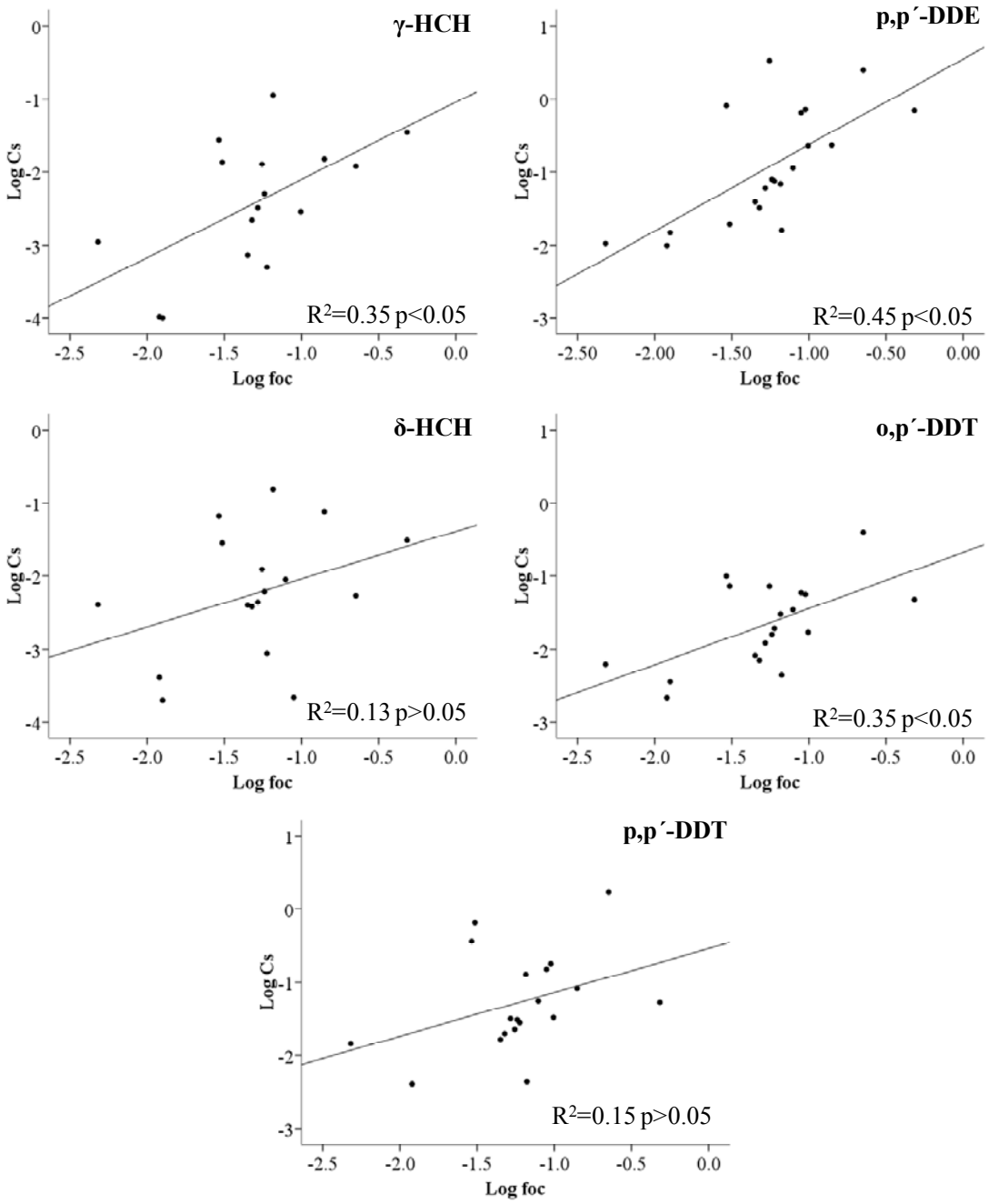

Figure S4. Log Cs (soil concentration in ng gdw-1) versus Log f_{OC} for selected HCH isomers and DDT metabolites

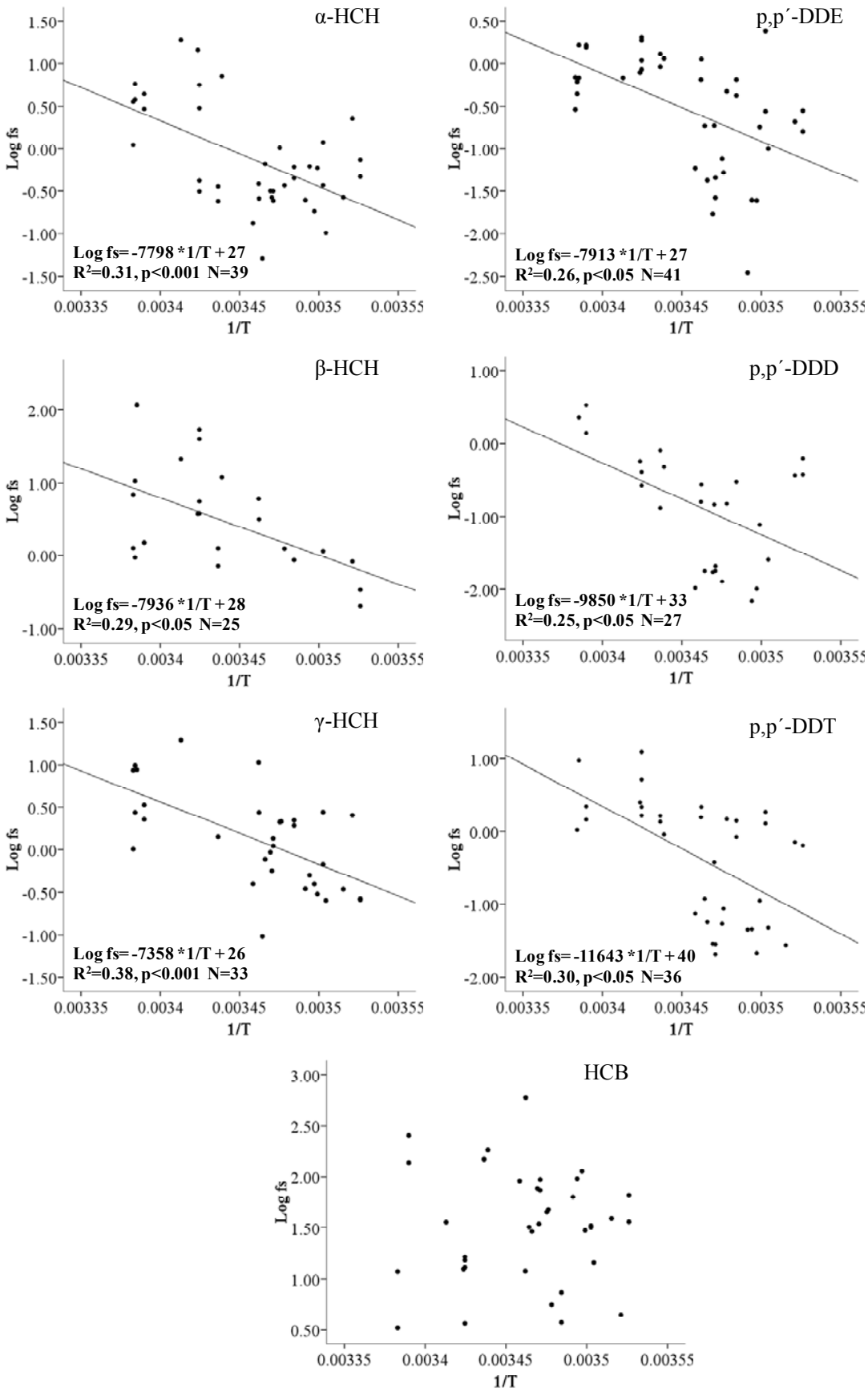

Figure S5. Soil fugacity (Log fs*10^{10}) regressed against 1/T

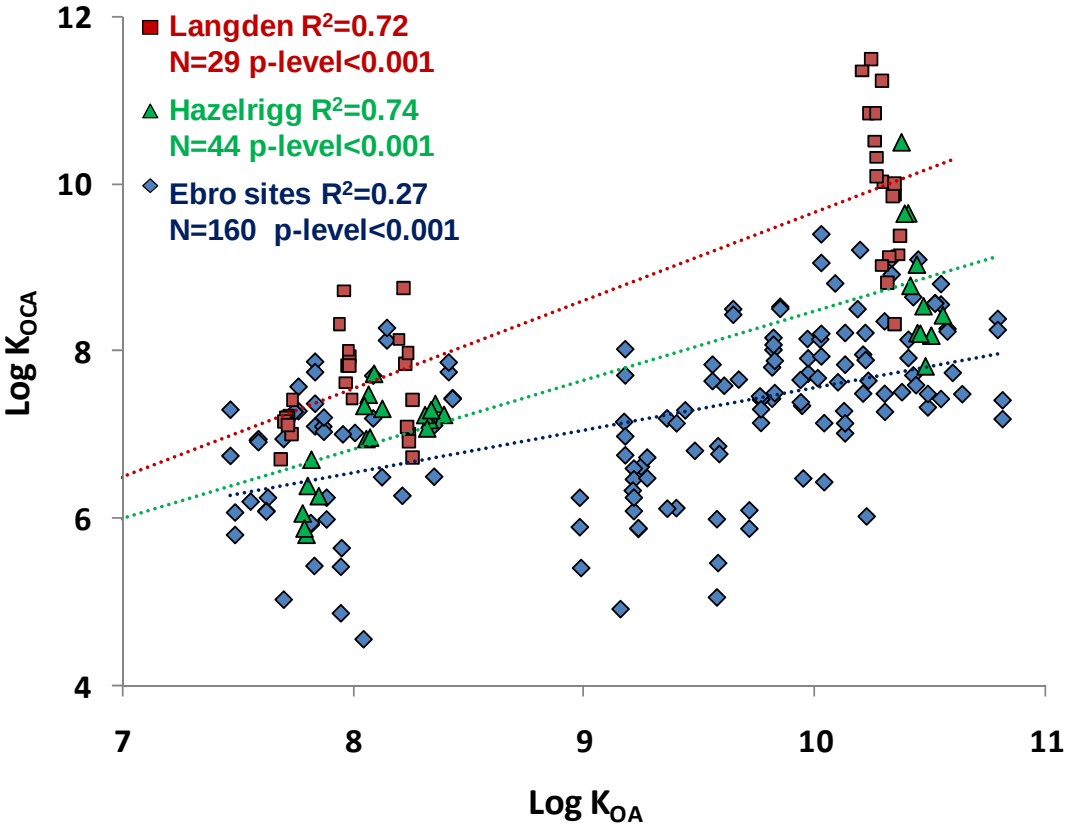

Figure S6. Log K_{OCA} regressed versus Log K_{OA} for different sampling sites in Spain (Ebro) and UK (Hazelrigg and Langden)

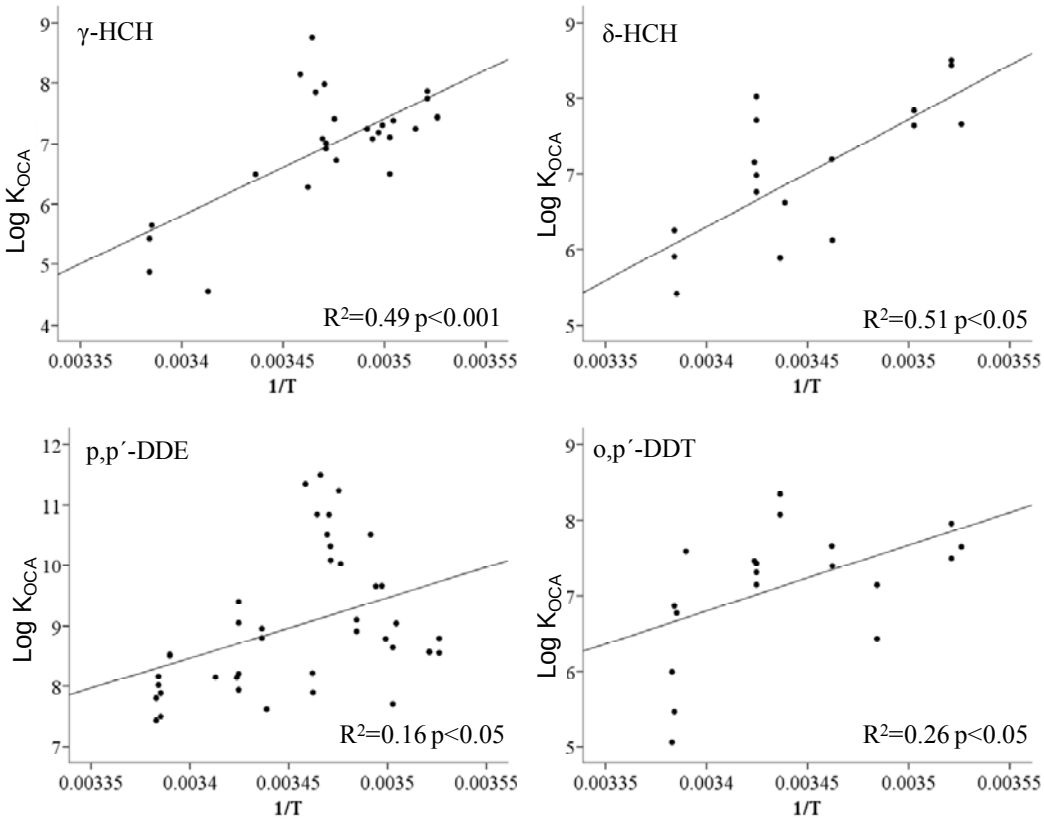

Figure S7. Examples of Log K_{OCA} regressed against 1/T

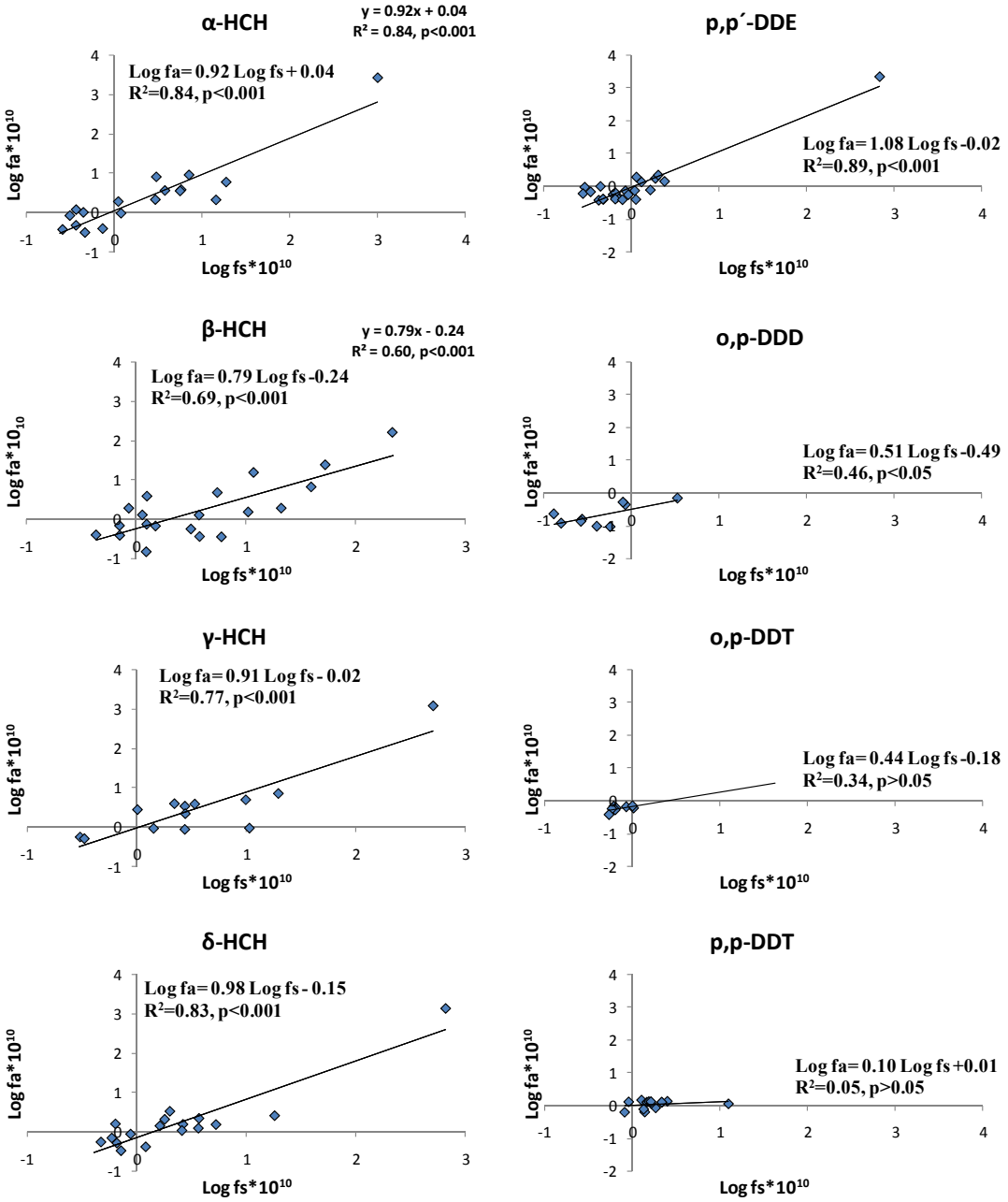

Figure S8a. Log fugacity in soil (fs) versus log fugacity in air (fa) for individual HCH isomers and DDT metabolites for sampling sites in Spain

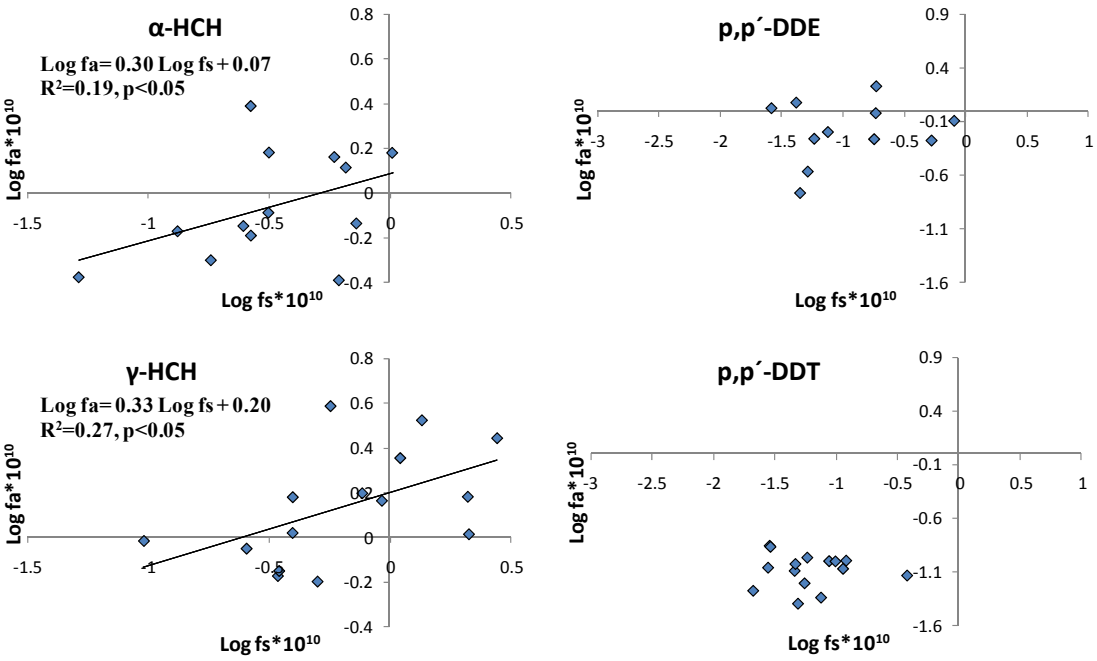

Figure S8b. Log fugacity in soil (fs) versus log fugacity in air (fa) for individual HCH isomers and DDT metabolites for sampling sites in UK

Annex 2. List of tables

	Lat	Long	Sampling site	Sampling period	Nº air samples	Nº soil samples
Borau	42˚38N	0˚35W	Rural	*13-15/06/2006 *17-19/11/2006 *16/09/2007	3 4 2	1 1 1
Alfaro	42˚11N	1˚47W	Rural	*19-21/06/2006 *14-16/11/2006 *17-19/09/2007	3 4 4	1 1 1
Tudela	42˚04N	1˚36W	Semi-rural	*19-21/06/2006 *14-16/11/2006 *17-19/09/2007	4 4 4	1 1 1
Sabiñanigo	42˚31N	0˚22W	Semi-rural	*16-18/06/2006 *17-19/11/2006 *14-16/09/2007	3 3 4	1 1 1
Najera	42˚25N	2˚45W	Rural	*22-24/06/2006	4	1
Uruñuela	42˚43N	2˚7W	Rural	*22-24/06/2006	4	1
Lasieso	42˚42N	0˚43W	Rural	*16-18/06/2006 *17-19/11/2006 *14-16/09/2007	4	1 1 1
Hazelrigg	54˚1N	2˚46W	Semi-rural	*12-17/08/2008 *18-29/08/2008	8 10	1 1
Langden	53˚95N	2˚56W	Rural	*17-24/09/2008	12	1
Total number of Samples					**84**	**20**

Table S1. Sampling sites and sampling details

	BORAU	SABIÑANIGO		TUDELA	ALFARO	NAJERA		URUÑUELA		LASIESO	
		Mean	SD			Mean	SD	Mean	SD	Mean	SD
HCB	Nd	nd		nd	nd	nd		nd		nd	
α-HCH	0.020	0.132	0.086	0.025	0.022	0.081	0.038	0.069	0.038	0.044	0.002
β-HCH	0.006	0.033	0.014	0.008	0.047	0.126	0.087	0.188	0.151	0.096	0.110
g-HCH	0.031	0.135	0.070	0.046	0.033	<loq		<loq		0.050	0.013
d-HCH	0.005	0.052	0.029	0.022	0.010	0.034	0.006	0.025	0.001	0.016	0.001
o,p´ -DDE	<loq	<loq		<loq	<loq	<loq		<loq		<loq	
p,p´-DDE	0.006	0.007	0.001	0.010	0.012	0.028	0.004	0.016	0.009	0.010	0.004
p,p´-DDD	<loq	<loq		0.009	0.010	0.033	0.014	0.023	0.012	<loq	<loq
o,p´-DDT	<loq	<loq		<loq	<loq	0.006	0.009	<loq		<loq	<loq
p,p´-DDT	<loq	<loq		<loq	0.009	0.005	0.008	0.008	0.012	<loq	<loq
ΣHCHs	0.062	0.352		0.102	0.113	0.242		0.282		0.206	
ΣDDTs	0.006	0.007		0.019	0.031	0.083		0.047		0.010	
Air T (K)	278.24	283.46		284.33	284.16	293.57	1.31	293.94	1.28	293.63	0.75
Precipitation (mm)	5.25	0.2		0.4	1.65	0	-	3.3	4.67	7.0	8.20

Table S2a. Ambient air concentration (mean±SD) in ng m^{-3}, ambient air temperature and precipitation for Ebro sampling sites in June 2006. <loq: data below quantification limit. Nd: not determined due to chromatographic problems.

	BORAU		SABIÑANIGO	TUDELA		ALFARO	
	Mean	SD		Mean	SD	Mean	SD
HCB	0.018	0.022	0.022	ND		0.065	0.024
α-HCH	0.004	0.001	0.054	0.026	0.020	0.013	0.002
β-HCH	0.005	0.000	0.006	0.040	0.023	0.018	0.002
g-HCH	0.007	0.000	0.058	ND		0.031	0.006
d-HCH	0.003	0.004	0.019	0.036	0.008	0.032	0.018
o,p´ -DDE	<loq		<loq	<loq		<loq	
p,p´-DDE	0.007	0.003	0.009	0.007	0.002	0.013	0.007
p,p´-DDD	0.003	0.004	<loq	<loq		0.003	0.004
o,p´-DDT	<loq		<loq	<loq		0.012	0.001
p,p´-DDT	0.005	0.007	<loq	0.018	0.012	0.017	0.006
ΣHCHs	0.019		0.137	0.101		0.093	
ΣDDTs	0.015		0.009	0.024		0.045	
Air T (K)	278.24	1.60	283.46	284.33	2.56	284.16	3.82
Precipitation (mm)	5.25	0.21	0.2	0.4	0.57	1.65	2.33

Table S2b. Ambient air concentration (mean±SD) in ng m^{-3}, ambient air temperature and precipitation for Ebro sampling sites in November 2006. <loq: data below quantification limit

	BORAU	SABIÑANIGO		ALFARO		TUDELA	
	Mean	Mean	SD	Mean	SD	Mean	SD
HCB	0.054	0.055	0.003	0.057	0.018	0.066	0.023
α-HCH	0.004	0.038	0.019	0.010		<loq	
β-HCH	0.007	0.010	0.008	0.031	0.038	0.009	0.000
g-HCH	0.010	<loq		<loq		0.010	0.002
d-HCH	0.018	0.007	0.002	0.004	0.001	0.016	0.004
o,p´ -DDE	<loq	<loq		<loq		<loq	
p,p´-DDE	0.005	0.005	0.000	0.019	0.013	0.012	0.007
p,p´-DDD	0.002	0.001	0.000	0.002	0.000	0.005	0.003
o,p´-DDT	0.008	0.009	0.000	0.009	0.001	0.008	0.003
p,p´-DDT	0.018	0.019	0.000	0.018	0.001	0.015	0.006
ΣHCHs	0.035	0.055		0.046		0.034	
ΣDDTs	0.111	0.035		0.048		0.041	
Air T (K)	288.83	293.50	1.52	289.60	2.29	289.83	2.10
Precipitation (mm)	na	0.3	0.42	0.4	0.57	0.75	1.06

Table S2c. Ambient air concentration (mean±SD) in ng m^{-3}, ambient air temperature and precipitation for Ebro sampling sites in September 2007.<loq: data below quantification limit, na= non-available data

	Hazelrigg		Langden	
	Mean	SD	Mean	SD
HCB	0.037	0.013	0.034	0.010
α-HCH	0.014	0.008	0.009	0.004
β-HCH	0.041		<loq	
g-HCH	0.024	0.012	0.014	0.010
d-HCH	<loq		<loq	
o,p´ -DDE	0.002	0.001	0.001	0.000
p,p´-DDE	0.011	0.007	0.009	0.001
o,p´-DDD	0.001	0.000	0.001	0.000
p,p´-DDD	0.001	0.000	0.001	0.000
o,p´-DDT	<loq		<loq	
p,p´-DDT	0.001	0.000	0.001	0.000
ΣHCHs	0.078		0.023	
ΣDDTs	0.016		0.012	
Air T (K)	288.22	0.48	285.70	0.64
Precipitation (mm)	0.41	0.54	0.03	0.00

Table S2d. Ambient air concentration (mean±SD) in ng m^{-3}, ambient air temperature and precipitation for UK sampling sites.

<loq: data below quantification limit

	BORAU			SABIÑANIGO			LASIESO			ALFARO			TUDELA			URUÑUELA	NAJERA
	Jun-06	Nov-06	Sep-07	Jun-06	Nov-06	Sep-07	Jun-06	Nov-06	Sep-07	Jun-06	Nov-06	Sep-07	Jun-06	Nov-06	Sep-07	Jun-06	Jun-06
HCB	0.0439	0.0447	0.0397	0.0400	0.0443	0.0989	0.0036	0.0089	0.0086	0.0508	0.0474	0.1559	0.1697	0.1503	0.4853	<loq	0.1174
α-HCH	<loq	<loq	<loq	<loq	0.2442	0.0037	<loq	<loq	<loq	<loq	0.0324	0.0277	<loq	<loq	0.0105	0.0249	<loq
β-HCH	0.0002	0.0002	0.0277	0.0013	<loq	0.0077	<loq	n	0.0039	<loq	<loq	0.0081	<loq	<loq	0.0108	0.0554	<loq
γ-HCH	0.0022	0.0051	0.0032	0.0005	0.1149	<loq	0.0001	0.0001	0.0011	<loq	0.0151	0.0029	<loq	<loq	0.0120	0.0271	0.0136
δ-HCH	0.0038	0.0062	0.0044	0.0009	0.1570	0.0090	0.0004	0.0002	0.0041	<loq	0.0767	<loq	<loq	<loq	0.0054	0.0672	0.0281
o,p´-DDE	<loq	<loq	0.0014	0.0019	0.0449	0.0016	0.0003	<loq	0.0002	<loq	0.1015	0.0001	0.0042	0.0049	0.0279	0.3618	0.2232
p,p´-DDE	0.0324	0.0792	0.0607	0.0747	0.0690	0.1126	0.0099	0.0150	0.0106	0.0161	0.2320	0.2274	0.6494	0.7226	2.4911	0.8141	0.0195
p,p´-DDD	0.0015	0.0076	0.0052	<loq	0.0795	<loq	0.0001	<loq	<loq	<loq	0.1049	<loq	<loq	0.0118	0.0948	0.1147	0.2409
o,p´-DDT	0.0072	0.0162	0.0122	0.0195	0.0303	0.0347	0.0022	0.0036	0.0063	0.0045	0.0000	0.0173	0.0598	0.0568	0.4004	0.0994	0.0729
p,p´-DDT	0.0197	0.0305	0.0315	0.0279	0.1302	0.0557	0.0040	<loq	0.0145	0.0043	0.0829	0.0327	0.1531	0.1817	1.7154	0.3691	0.6607
ΣHCHs	0.0062	0.0114	0.0354	0.0026	0.5161	0.0204	0.0005	0.0003	0.0091	<loq	0.1242	0.0387	<loq	<loq	0.0387	0.1745	0.0417
ΣDDTs	0.0608	0.1334	0.1110	0.1240	0.3538	0.2046	0.0166	0.0186	0.0316	0.0249	0.5212	0.2775	0.8665	0.9778	4.7297	1.7591	1.2173
%TOC	4.78	5.75	5.21	5.97	6.54	7.88	1.20	1.26	0.48	6.66	14.07	9.88	8.92	9.5	22.53	2.92	3.06
%N	0.49	0.58	0.51	0.44	0.47	0.57	0.12	na	na	0.66	1.33	0.93	0.55	0.60	1.00	0.27	0.22
Soil T	295.4	283.6	294.8	293.0	284	287.7	295.5	285.4	299.3	295.6	285.5	292.0	295 .0	287 .0	291.0	292.0	290.8
pH	7.96	7.87	8.2	7.81	7.66	8.00	8.38	8.18	8.60	7.83	8.13	8.30	7.93	8.14	8.20	8.20	8.03
Redox	250	298	305	242	266	nd	214	298	298	253	240	240	231	254	261	239	238
SWC %	3.36	30.06	4.29	14.62	28.87	5.97	3.38	16.50	5.57	19.92	27.87	17.53	9.16	15.07	22.29	18.85	16.89
Altitude	910	910	910	790	790	790	720	720	720	302	302	302	269	269	269	499	265

Table S3a. Soil concentration in (ng gdw^{-1}) and soil physico-chemical properties (%TOC: soil organic carbon, %N: soil nitrogen, Soil T: Soil temperature, soil pH, soil redox, soil water content (SWC) and site altitude for selected sites along the Ebro river basin. <loq: data below quantification limit, Nd: not measured data due to sensor problems. Na: non-available data

	Hazelrigg	Hazelrigg	Langden
HCB	0.0792	0.1044	0.4127
α-HCH	0.0014	0.0064	0.0320
β-HCH	<loq	<loq	<loq
g-HCH	0.0007	0.0128	0.0349
d-HCH	0.0040	0.0124	0.0307
o,p´ -DDE	0.0083	0.0187	0.0560
p,p´-DDE	0.0391	3.3920	0.7008
o,p´-DDD	<loq		
o,p´-DDT	0.0083	0.0726	0.0483
p,p´-DDT	0.0165	0.0226	0.0533
ΣHCHs	0.0061	0.0316	0.0975
ΣDDTs	0.0722	3.5058	0.8584
%TOC	4.48	5.54	48.33
%N	0.20	0.30	1
Soil T	289.10	289.7	286.4
pH	5.87	5.94	4.5
Redox	427	400	407
SWC %	47.45	56.20	81.80
Altitude	94.2	94.2	190

Table S3b. Soil concentration in (ng g dw^{-1}) and soil physico-chemical properties (%TOC: soil organic carbon, %N: soil nitrogen, Soil T: Soil temperature, soil pH, soil redox, soil water content (SWC) and site altitude in UK s sites. <loq: data below quantification limit.

	HCB	ΣDDT	ΣHCH	ΣPCBs	ΣPAHs
HCB	1.00				
ΣDDT	0.67**	1.00			
ΣHCH	0.42*	0.54*	1.00		
ΣPCBs	0.71**	0.64**	0.24*	1.00	
ΣPAHs	0.57**	0.48*	0.30*	0.49**	1.00

*p<0.05, **p<0.001, the data were log-transformed before analysis.

Table S4. Correlation coefficients for all soils

	R^2	A	p	b	p	N
HCB			>0.05		>0.05	
α-HCH	0.14	-0.80	<0.05	-0.72	<0.05	39
β-HCH	0.12	-0.46	>0.05	-0.86	0.09	25
g-HCH	0.15	-0.51	>0.05	-0.66	>0.05	34
d-HCH			>0.05		>0.05	
o,p´ -DDE			>0.05		>0.05	
p,p´-DDE			>0.05		>0.05	
p,p´-DDD			>0.05		>0.05	
o,p´-DDT	0.19	-0.63	>0.05	-0.86	<0.05	20
p,p´-DDT	0.12	-1.12	>0.05	-0.70	<0.05	35

Table S5a. Soil fugacity regressed against f_{OC}; a, b are the fitting parameters in the equation Log fs=a +b(log f_{OC}) p the level of significance and N the number of samples considered

	R^2	A	p	b	p	N
HCB			>0.05		>0.05	
α-HCH	0.16	1.65	<0.05	-1.37	<0.05	39
β-HCH			>0.05		>0.05	
g-HCH	0.21	1.91	<0.05	-1.38	<0.05	34
d-HCH			>0.05		>0.05	
o,p´ -DDE	0.26	0.23	>0.05	-1.56	<0.05	15
p,p´-DDE	0.32	2.26	<0.05	-2.25	<0.001	41
p,p´-DDD	0.42	2.08	<0.05	-2.35	<0.001	26
o,p´-DDT			>0.05		>0.05	
p,p´-DDT	0.59	3.12	<0.001	-2.79	<0.001	35

Table S5b. Soil fugacity regressed against f_{OC}/f_N; a, b are the fitting parameters in the equation Log fs=a +b(log f_{OC}/f_N) p the level of significance and N the number of samples considered

	R^2	A	p	b	p	N
HCB			>0.05		>0.05	
α-HCH	0.19	-1.77	<0.05	0.25	<0.05	39
β-HCH			>0.05		>0.05	
g-HCH	0.30	-1.67	<0.05	0.27	<0.05	34
d-HCH			>0.05		>0.05	
o,p´ -DDE	0.47	-4.46	<0.001	0.45	<0.05	15
p,p´-DDE	0.50	-3.70	<0.001	0.45	<0.001	41
p,p´-DDD	0.67	-4.10	<0.001	0.47	<0.001	26
o,p´-DDT			>0.05			
p,p´-DDT	0.80	-3.92	>0.001	0.51	<0.001	35

Table S5c. Soil fugacity regressed against pH; a, b are the fitting parameters in the equation Log fs=a +b(pH) p the level of significance and N the number of samples considered

	R^2	A	p	b	p	N
HCB	0.16	8.38	<0.001	-0.75	<0.05	35
α-HCH	0.17	9.37	<0.001	-0.78	<0.05	26
β-HCH	ne					
g-HCH	0.26	9.94	<0.001	-1.23	<0.05	27
d-HCH			>0.05		>0.05	
o,p´ -DDE	0.57	14.22	<0.001	-2.22	<0.05	15
p,p´-DDE	0.60	15.25	<0.001	-2.52	<0.001	41
p,p´-DDD	ne					
o,p´-DDT			<0.05		<0.05	
p,p´-DDT	0.55	12.68	<0.001	-1.84	<0.001	36

Table S6a. K_{OCA} regressed against altitude; a, b are the fitting parameters in the equation Log K_{OCA} =a +b(log altitude), p the level of significance, ne (not enough data for statistic analysis) and N the number of samples considered

	R^2	A	p	b	p	N
HCB			>0.05		>0.05	
α-HCH			>0.05		>0.05	
β-HCH	Ne					
g-HCH			>0.05		>0.05	
d-HCH			>0.05		>0.05	
o,p´ -DDE	Ne					
p,p´-DDE	0.29	5.61	<0.001	2.81	<0.001	41
p,p´-DDD	Ne					
o,p´-DDT	0.31	2.73	<0.001	4.00	<0.05	20
p,p´-DDT	0.19	6.21	<0.001	1.61	<0.05	36

Table S6b. K_{OCA} regressed against f_{OC}/f_N; a, b are the fitting parameters in the equation Log K_{OCA} =a +b(log f_{OC}/f_N), p the level of significance, ne (not enough data for statistic analysis) and N the number of samples considered.

	R^2	A	p	b	p	N
HCB			>0.05		>0.05	
α-HCH			>0.05		>0.05	
β-HCH	Ne					
g-HCH	0.23	9.04	<0.05	-0.31	<0.05	27
d-HCH	0.30	20.86	<0.05	-2.45	<0.05	18
o,p´ -DDE	Ne					
p,p´-DDE	0.48	13.21	<0.001	-0.60	<0.001	41
p,p´-DDD	Ne					
o,p´-DDT			>0.05		>0.05	
p,p´-DDT	0.26	10.27	<0.001	-0.30	<0.05	36

Table S6c. K_{OC} regressed against soil pH; a, b are the fitting parameters in the equation Log K_{OC} =a +b (pH), p the level of significance, ne (not enough data for statistic analysis) and N the number of samples considered.