

## Supplementary information:

# The long shadow of our chemical past - high DDT concentrations in fish near a former agrochemicals factory in England

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## Contents

### Figures

Figure S 1	Chemical structures and pathways of components of total DDTs .....	2
Figure S 2	Structure of pp' Dicofol .....	2
Figure S 3	Advertisement in Masseur (1946), demonstrating the production of DDT near the river Lee site in Wheathampstead.....	3
Figure S 4	Structure analyses plot from DNA microsatellite data.....	4

### Tables

Table S 1	Information about the catchment areas above the sampling sites .....	5
Table S 2	Physico-chemical properties of DDTs.....	6
Table S 3	Observed tissue concentration effect levels .....	6
Table S 4	$\sum$ DDTs: Pairwise P of ANOVA (Holm-Sidak method) for Log(total DDTs). .....	8
Table S 5	Chlordane: Pairwise P of ANOVA (Holm-Sidak method) for Log(chlordane). .....	9
Table S 6	Lindane ( $\gamma$ -HCH): Pairwise P of ANOVA (Holm-Sidak method) for Log( $\gamma$ -HCH). .....	10
Table S 7	Hexachlorobenzene (HCB): Pairwise P of ANOVA (Holm-Sidak method) for Log(HCB). ...	11
Table S 8	Copper: Pairwise P of ANOVA (Holm-Sidak method) for Log(total copper).....	12
Table S 9	$\sum$ 7PCBs: Pairwise P of ANOVA (Holm-Sidak method) for Log( $\sum$ 7PCBs).....	13
Table S 10	$\sum$ 6PBDEs: Pairwise P of ANOVA (Holm-Sidak method) for Log( $\sum$ 6PBDEs). .....	14

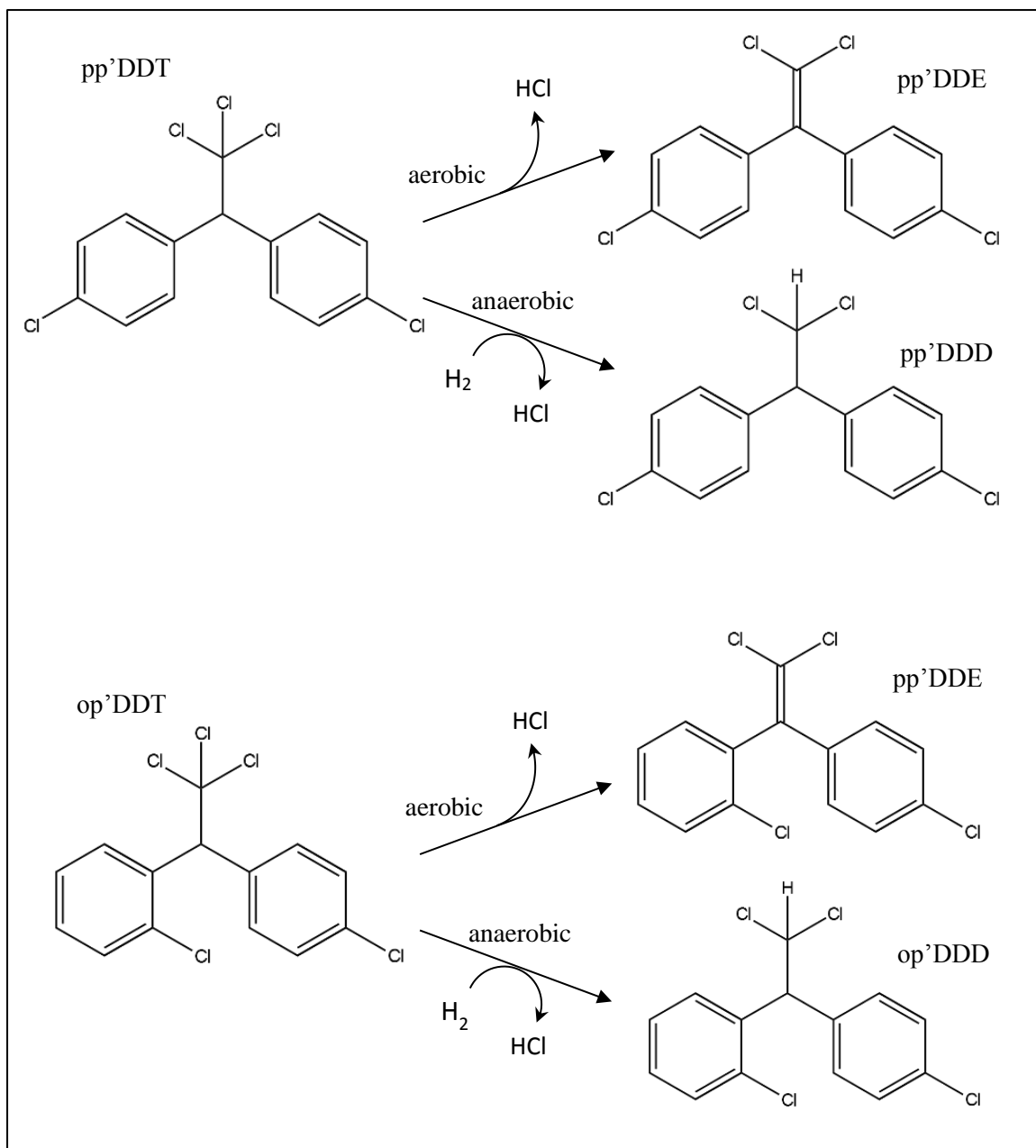


Figure S 1 Chemical structures and pathways of components of total DDTs

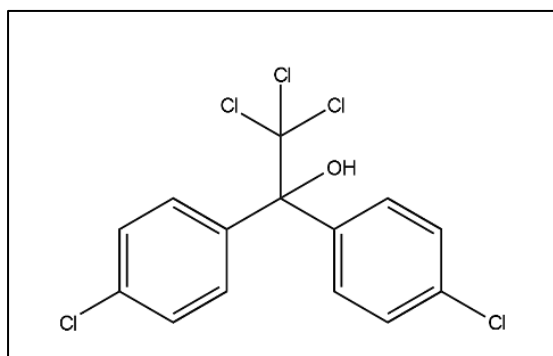


Figure S 2 Structure of *pp'* Dicofol

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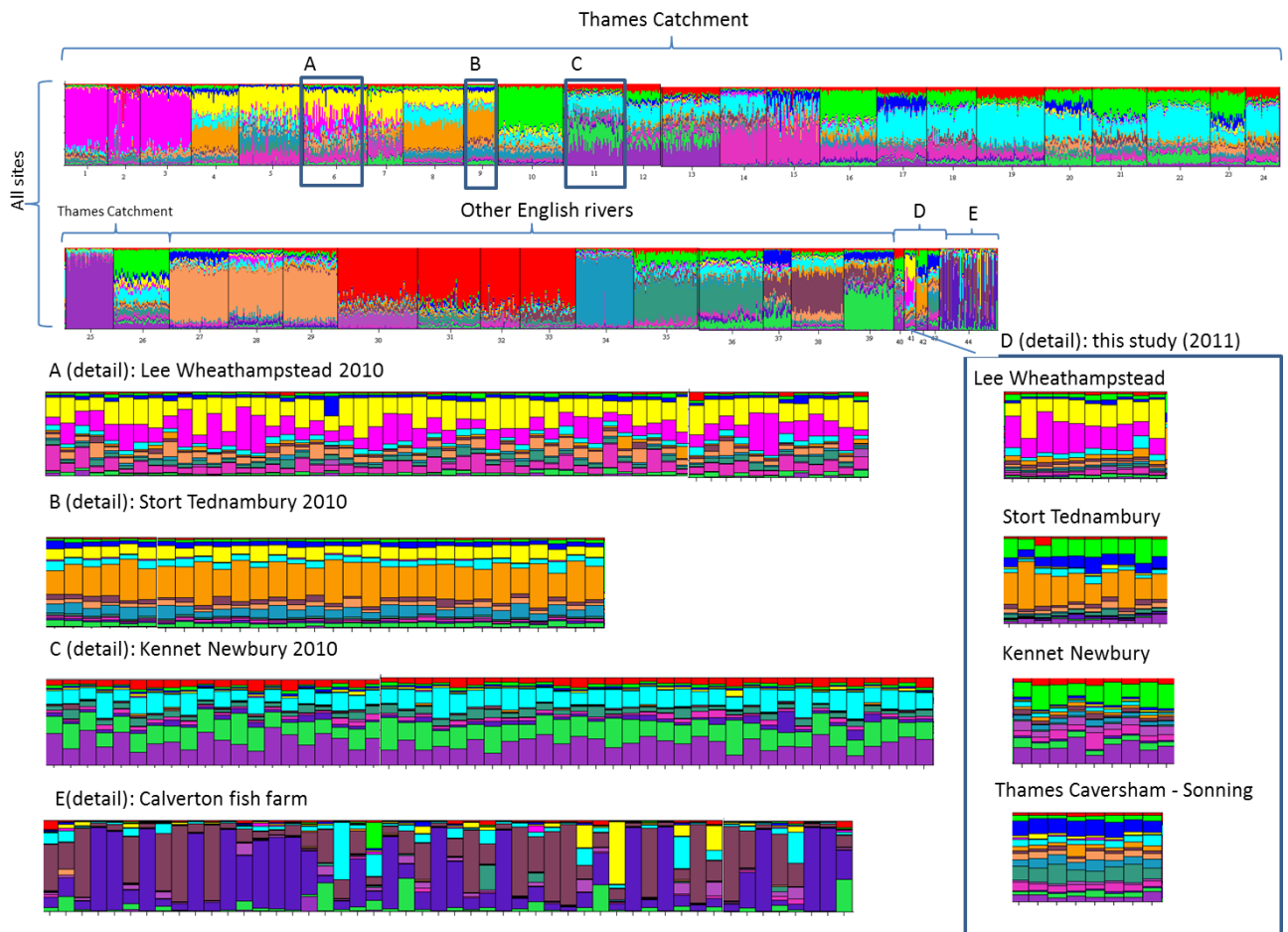
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Figure S 3 Advertisement in Masee (1946), demonstrating the production of DDT near the river Lee site in Wheathampstead



**Figure S 4** Structure analyses plot from DNA microsatellite data, using the locprior model at  $K = 16$ . Each individual is represented by a thin horizontal line, which is partitioned into  $K$  coloured segments representing an individual's estimated membership to fractions of  $K$  clusters. The dataset contains 1858 fish, including 1,769 roach from 32 river locations from Hamilton *et al.* (2014), 39 fish from 4 locations used for chemical analysis in this study (D) and 50 fish from Calverton fish farm, the source of fish used for restocking by the Environment Agency in England and Wales (E).

**Table S 1 Information about the catchment areas above the sampling sites (Data from the National River Flow Archive (NRFA) and Landcover map 2000, summarized in Marsh and Hannaford (2008) and detailed in the IRN/RACQUEL program developed by CEH.**

Area	Anglian				Thames tributaries			Thames						
River	Glen	Nene			Kennet	Lee	Stort							
dominant geology	Oxford Clay, Great Oolite	Upper Lias, Inferior Oolite			Chalk		Chalk, London Clay	Great Oolite, Oxford Clay	Chalk, Great Oolite, Oxford Clay					
Site	Pinchbeck West	Cogenhoe	Thrapston	Oundle	Newbury	Wheat-hampstead	Tednam-bury	Castle Eaton	Caversham - Sonning	Temple - Marlow	Bray - Boveney	Old Windsor - Bell	Sunbury - Molesey	
Dist. from source <sup>a</sup> [km]	53	40	73	90	58	24	29	43	162-66	187-90	203-09	216-23	239-43	
Catchment area <sup>b</sup> [km <sup>2</sup> ]	187	612	1129	1314	543	88	135	547	5786	6700	7041	7169	9337	
Urban	1.2%	3.1%	3.3%	3.0%	0.6%	11.9%	1.9%	2.8%	1.9%	2.3%	2.5%	2.6%	3.2%	
Suburban/rural developed	3.0%	5.9%	5.9%	5.5%	2.7%	34.9%	8.1%	5.6%	5.5%	7.1%	7.6%	7.9%	9.9%	
Cereals	33.2%	20.8%	23.5%	24.9%	28.7%	12.4%	19.6%	15.2%	19.8%	18.8%	18.5%	18.3%	16.6%	
Arable (horticult. +non-rot.)	36.1%	29.9%	29.0%	28.7%	20.6%	15.0%	39.6%	22.5%	24.6%	22.9%	22.4%	22.1%	20.3%	
Improved grassland	7.5%	18.9%	15.9%	14.7%	26.0%	10.6%	5.2%	29.0%	24.0%	23.2%	23.0%	22.9%	20.8%	
Other grass or heathland	7.7%	10.5%	11.7%	12.0%	3.6%	8.3%	13.8%	11.8%	10.9%	11.2%	11.3%	11.3%	11.8%	
Woodland	10.6%	9.9%	9.5%	10.0%	14.4%	6.5%	10.9%	11.1%	11.5%	12.8%	13.1%	13.2%	15.6%	
Other	0.8%	1.1%	1.2%	1.1%	3.4%	0.5%	1.0%	2.1%	1.7%	1.7%	1.7%	1.7%	1.8%	
Modelled average treated sewage content in sampling stretch	1.0%	6.6%	29%	26%	3.0%	28%	43%	22%	13%	15%	16%	17%	16%	
annual rainfall [mm]	597	641	629	625	772	664	613	766	694	696	696	696	704	

<sup>a</sup> distance along the channel

<sup>b</sup> catchment area above a point approximately in the centre of the sampling stretch

**Table S 2 Physico-chemical properties of DDTs and dicofol (from <http://chem.sis.nlm.nih.gov/chemidplus>)**

Physical Property	T	Units	pp'DDT	op'DDT	pp'DDE	op'DDE	pp'DDD	op'DDD	dicofol
Melting Point		°C	108.5 (exp)		89 (exp)		109.5 (exp)	77 (exp)	77.5 (exp)
Boiling Point		°C			336 (exp)		350 (exp)		
Log P (Octanol-Water)		(none)	6.91 (exp)	6.79 (est)	6.51 (exp)	6.00 (est)	6.02 (exp)	5.87 (est)	5.02 (exp)
Water Solubility	25°C	mg/L	0.0055 (exp)	0.085 (exp)	0.04 (exp)	0.14 (exp)	0.09 (exp)	0.1 (exp)	0.8 (exp)
Vapour Pressure	variable	mm Hg	1.60E-07 at 20°C (exp)			6.20E-06 at 25°C (exp)	1.35E-06 at 25°C (exp)	1.94E-06 at 30°C (exp)	3.98E-07 at 25°C (exp)
Henry's Law Constant	25°C	atm-m <sup>3</sup> /mole	8.32E-06 (exp)	7.41E-06 (est)	4.16E-05 (exp)	1.85E-05 (est)	6.60E-06 (exp)	8.17E-06 (est)	2.42E-07 (est)
Atmospheric OH Reaction Rate Constant	25°C	cm <sup>3</sup> /mole-sec	3.44E-12 (est 25°C)		7.43E-12 (est)		4.34E-12 (est)		3.43E-12 (est)

**Table S 3 Observed tissue concentration effect levels (whole body LOECs) for exposed fish and estimated safe concentrations for fish and their consumers**

species	effect	exposure	tissue conc. [µg/kg]	Reference
<b>Early Life Stages (ELS), whole body effect levels (LOECs)</b>				
Japanese Medaka	intersex and altered gene expression	∑op'DDTs	58	(Sun <i>et al.</i> 2016)
Atlantic Croaker	behaviour	op'DDT, food of parent	70 in eggs	(Faulk <i>et al.</i> 1999)
Japanese Medaka	intersex, increased plasma estradiol, altered gene expression	pp'DDE	271	(Sun <i>et al.</i> 2016)
Brook Trout	survival	technical DDT, food	890	(Macek 1968b) <sup>a</sup>
Coho salmon	survival	pp'DDT maternal transfer	1,090	(Johnson 1996) <sup>a</sup>
Coho salmon	behaviour	DDT maternal transfer	1,100	(Johnson 1996) <sup>a</sup>
Winter flounder	survival	technical DDT aqueous	1,110	(Smith 1973) <sup>a</sup>
Rainbow trout	survival	DDT in food of parent	1,150	(Cuerrier 1967) <sup>a</sup>
Rainbow trout	survival	DDT maternal transfer	1,270	(Hopkins 1969) <sup>a</sup>
Spotted sea trout	survival	DDT maternal transfer	1,500	(Butler 1972) <sup>a</sup>
Lake trout	survival	DDT maternal transfer	2,930	(Burdick 1964) <sup>a</sup>
Brook Trout	survival	technical DDT, food	11,920	(Burdick 1972) <sup>a</sup>
Fathead minnow	survival	technical DDT aqueous	24,000	(Jarvinen 1976,1977) <sup>a</sup>
Summer flounder (males)	vitellogenin ↑ gonadosomatic index ↓	op'DDT, total dose injected	60,000	(Mills <i>et al.</i> 2001)
Rainbow trout	vitellogenin ↑ Hepatic estrogen binding sites ↑	op'DDT liver conc. after 8 weeks total dose injected: op'DDT+ op'DDE 45+90 µg/g	320,000 135,000	(Donohoe and Curtis 1996)
<b>Adults, whole body effect levels (LOECs)</b>				
Lake trout	survival	DDE aqueous+food	290	(Berlin <i>et al.</i> 1981) <sup>a</sup>
Pinfish	survival	pp'DDT food	550	(Butler 1969) <sup>a</sup>
Cutthroat salmon	survival	pp'DDT aqueous	1,100	(Allison <i>et al.</i> 1963,1964) <sup>a</sup>
Chinook salmon	survival	technical DDT, food	3,650	(Buhler 1969) <sup>a</sup>
Coho Salmon	survival	technical DDT, food	33,800	(Buhler 1969) <sup>a</sup>
Fathead minnow	survival	technical DDT, aqueous	112,700	(Jarvinen 1976,1977) <sup>a</sup>
Goldfish	behaviour	pp'DDT aqueous	1,650	(Davy <i>et al.</i> 1972) <sup>a</sup>
Brook trout	reproduction	technical DDT, food	7,600	(Macek 1968b) <sup>a</sup>
Brook trout	growth	technical DDT, food	11,200	(Macek 1968a) <sup>a</sup>
Tilapia	osmoregulation	pp'DDT, water	ca. 20,000	(Mills <i>et al.</i> 2001)

species	effect	exposure	tissue conc. [µg/kg]	Reference
<b>Estimated safe tissue concentrations for fish themselves</b>				
Adult fish	provisional protective level	∑DDTs	600	(Beckvar <i>et al.</i> 2005)
Fish early life stages	provisional protective level	∑DDTs	700	(Beckvar <i>et al.</i> 2005)
<b>Estimated safe tissue concentrations for consumers of fish</b>				
Birds	based on egg shell thinning in ducks <sup>bc</sup>	∑DDTs	14	(Canadian Council of Ministers of the Environment 1999)
Kingfisher	based on effects on pelicans (see below) with food intake for kingfisher and factor NOEL=LOEL/3	∑DDTs	20	(Lazorchak <i>et al.</i> 2003)
Humans	risk based threshold for cancer (from 4 fish meals/month)	∑DDT	69	(Stahl <i>et al.</i> 2009)
Mammals	calculation uses effects on rats and food intake rate for mink <sup>c</sup>	∑DDT	94	(Canadian Council of Ministers of the Environment 1999)
Osprey	dietary effects concentration	pp'DDE	90-190	(Hinck <i>et al.</i> 2009a)
Bald Eagle	dietary effects concentration	pp'DDE	130	(Hinck <i>et al.</i> 2009a)
Pelican	reduced fledgling rate when declining conc. in fish were still 0.15 µg/g	∑DDTs	< 150 (LOEC)	(US EPA 1995)
Bald Eagle	no effects hazard concentration	∑DDTs	270	(Hinck <i>et al.</i> 2009b)
Mink	calculation uses effects on rats and food intake rate for mink with factor 10 for interspecies difference	∑DDTs	360	(Lazorchak <i>et al.</i> 2003)
Otter	calculation uses effects on rats and food intake rate for otter with factor 10 for interspecies difference	∑DDTs	490	(Lazorchak <i>et al.</i> 2003)
Humans	food standard for meat <sup>d</sup>	∑DDT	1,000	(European Union 2005)

<sup>a</sup> Quoted in Beckvar *et al.* (2005)

<sup>b</sup> Most sensitive endpoint (egg shell thinning in ducks) and highest food intake rate for fish eating birds (Wilson's storm petrel), includes uncertainty factor 10 to account for other species and a factor 2.4 to extrapolate from LOEC to threshold level.

<sup>c</sup> Experimental data was for pp'DDT in food but results are applied to ∑DDTs

<sup>d</sup> To our knowledge no standard has been set for fish

## Statistical analysis

Statistical tests were carried out in the SigmaPlot program (SigmaPlot for Windows, version 12.5, Systat Software Inc.)

**Table S 4**  $\Sigma$ DDTs: Pairwise P of ANOVA (Holm-Sidak method) for Log(total DDTs). Pink shaded cells indicate that the difference between these groups was statistically significant at  $\alpha=0.05$ . Normality Test (Shapiro-Wilk) passed, equal variance test passed, both at  $\alpha=0.05$

$\Sigma$ DDTs		GL 53 km	NE 40 km	NE 73 km	NE 90 km	KE 58 km	LE 24 km	ST 29 km	TH 43 km	TH 162- 166 km	TH 187- 190 km	TH 203- 209 km	TH 216- 223 km	TH 239- 243 km
number analysed		4	5	5	5	9	10	10	10	8	4	2	5	4
average conc. [ $\mu\text{g}/\text{kg}$ ]		12	5.4	7.9	3.7	1.8	88	5.9	2.4	3.3	7.5	3.9	6.1	3.4
std dev [ $\mu\text{g}/\text{kg}$ ]		1.9	1.3	2.1	1.4	0.5	70	2.2	0.9	1.3	3.3	2.2	2.8	0.5
GL Pinch- beck West	GL 53 km		0.187	0.953	0.003	<0.001	<0.001	0.084	<0.001	<0.001	0.894	0.076	0.249	0.003
NE Cogenhoe	NE 40 km	0.187		0.971	0.959	<0.001	<0.001	0.995	0.036	0.699	0.991	0.993	1.000	0.963
NE Thrapston	NE 73 km	0.953	0.971		0.221	<0.001	<0.001	0.965	<0.001	0.022	1.000	0.747	0.986	0.218
NE Oundle	NE 90 km	0.003	0.959	0.221		0.138	<0.001	0.910	0.906	1.000	0.538	0.955	0.951	1.000
KE Newbury	KE 58 km	<0.001	<0.001	<0.001	0.138		<0.001	<0.001	0.969	0.214	<0.001	0.641	<0.001	0.336
LE Wheat- hampstead	LE 24 km	<0.001	<0.001	<0.001	<0.001	<0.001		<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
ST Tednambury	ST 29 km	0.084	0.995	0.965	0.910	<0.001	<0.001		0.002	0.299	0.990	0.986	1.000	0.899
TH Castle Eaton	TH 43 km	<0.001	0.036	<0.001	0.906	0.969	<0.001	0.002		0.961	0.002	0.969	0.021	0.959
TH Cav.- Sonning	TH 162- 166 km	<0.001	0.699	0.022	1.000	0.214	<0.001	0.299	0.961		0.123	1.000	0.565	1.000
TH Temp- Marlow	TH 187- 190 km	0.894	0.991	1.000	0.538	<0.001	<0.001	0.990	0.002	0.123		0.913	0.996	0.507
TH Bray- Boveney	TH 203- 209 km	0.076	0.993	0.747	0.955	0.641	<0.001	0.986	0.969	1.000	0.913		0.986	1.000
TH Old W.- Bell	TH 216- 223 km	0.249	1.000	0.986	0.951	<0.001	<0.001	1.000	0.021	0.565	0.996	0.986		0.935
TH Sunbury- Molesey	TH 239- 243 km	0.003	0.963	0.218	1.000	0.336	<0.001	0.899	0.959	1.000	0.507	1.000	0.935	
significantly different to number of other groups		6/12	3/12	4/12	2/12	7/12	12/12	3/12	7/12	3/12	3/12	1/12	3/12	2/12



**Table S 5 Chlordane: Pairwise P of ANOVA (Holm-Sidak method) for Log(chlordane). Pink shaded cells indicate that the difference between these groups was statistically significant at  $\alpha=0.05$ . Normality Test (Shapiro-Wilk) passed at  $\alpha=0.05$ , equal variance failed passed ( $p=0.012$ ). Although the equal variance test had failed a standard ANOVA was carried out to allow comparison with other chemicals.**

Chlordane		GL 53 km	NE 40 km	NE 73 km	NE 90 km	KE 58 km	LE 24 km	ST 29 km	TH 43 km	TH 162- 166 km	TH 203- 209 km	TH 216- 223 km	TH 239- 243 km
number analysed		4	5	5	5	9	10	9	10	8	2	5	4
average conc. [ $\mu\text{g}/\text{kg}$ ]		0.07	0.23	0.40	0.13	0.05	0.66	0.17	0.16	0.29	0.08	0.98	0.18
std dev [ $\mu\text{g}/\text{kg}$ ]		0.01	0.11	0.15	0.07	0.01	0.24	0.03	0.03	0.27	0.02	0.47	0.05
GL Pinch- beck West	GL 53 km		0.016	<0.001	0.658	0.891	<0.001	0.035	0.067	0.003	0.989	<0.001	0.086
NE Cogenhoe	NE 40 km	0.016		0.469	0.758	<0.001	<0.001	0.993	0.966	0.842	0.318	<0.001	0.996
NE Thrapston	NE 73 km	<0.001	0.469		0.004	<0.001	0.654	0.043	0.014	0.438	0.003	0.283	0.104
NE Oundle	NE 90 km	0.658	0.758	0.004		0.005	<0.001	0.946	0.961	0.524	0.970	<0.001	0.944
KE Newbury	KE 58 km	0.891	<0.001	<0.001	0.005		<0.001	<0.001	<0.001	<0.001	0.852	<0.001	<0.001
LE Wheat- hampstead	LE 24 km	<0.001	<0.001	0.654	<0.001	<0.001		<0.001	<0.001	<0.001	<0.001	0.906	<0.001
ST Tednambury	ST 29 km	0.035	0.993	0.043	0.946	<0.001	<0.001		0.976	0.973	0.548	<0.001	0.971
TH Castle Eaton	TH 43 km	0.067	0.966	0.014	0.961	<0.001	<0.001	0.976		0.917	0.672	<0.001	0.997
TH Cav.- Sonning	TH 162- 166 km	0.003	0.842	0.438	0.524	<0.001	<0.001	0.973	0.917		0.170	<0.001	0.994
TH Bray- Boveney	TH 203- 209 km	0.989	0.318	0.003	0.970	0.852	<0.001	0.548	0.672	0.170		<0.001	0.587
TH Old W.- Bell	TH 216- 223 km	<0.001	<0.001	0.283	<0.001	<0.001	0.906	<0.001	<0.001	<0.001	<0.001		<0.001
TH Sunbury- Molesey	TH 239- 243 km	0.086	0.996	0.104	0.944	<0.001	<0.001	0.971	0.997	0.994	0.587	<0.001	
significantly different to number of other groups		6/11	4/11	6/11	4/11	9/11	9/11	5/11	4/11	4/11	3/11	9/11	3/11

**Table S 6 Lindane ( $\gamma$ -HCH): Pairwise P of ANOVA (Holm-Sidak method) for Log( $\gamma$ -HCH). Pink shaded cells indicate that the difference between these groups was statistically significant at  $\alpha=0.05$ . The standard method for DDTs was here also used for  $\gamma$ -HCH. This is less sensitive for HCHs than the method used in Jürgens *et al.* (2015) and suffered from problems with peaks corresponding to  $\gamma$ -HCH found in all procedural blanks. The values were not blank corrected, as that would have led to negative concentrations for some samples. Therefore, the lindane measurements and their associated statistics are to be seen as only semi-quantitative, but nevertheless show some differences between groups.**

$\gamma$ -HCH		NE 90 km	KE 58 km	LE 24 km	ST 29 km	TH 43 km
number analysed		5	9	10	10	10
average conc. [ $\mu\text{g}/\text{kg}$ ]		1.12	1.54	8.11	2.22	0.95
std dev [ $\mu\text{g}/\text{kg}$ ]		0.36	1.10	3.57	1.79	0.50
NE Oundle	NE 90 km		0.704	<0.001	0.431	0.732
KE Newbury	KE 58 km	0.704		<0.001	0.454	0.484
LE Wheat-hampstead	LE 24 km	<0.001	<0.001		<0.001	<0.001
ST Tednambury	ST 29 km	0.431	0.454	<0.001		0.037
TH Castle Eaton	TH 43 km	0.732	0.484	<0.001	0.037	
significantly different to number of other groups		1/4	1/4	4/4	2/4	2/4

**Table S 7 Hexachlorobenzene (HCB): Pairwise P of ANOVA (Holm-Sidak method) for Log(HCB). Pink shaded cells indicate that the difference between these groups was statistically significant at  $\alpha=0.05$ . Normality Test (Shapiro-Wilk) failed ( $p<0.001$ ), equal variance test passed at  $\alpha=0.05$ . Although the normality test had failed, a standard ANOVA was carried out to allow comparison with other chemicals.**

HCB		GL 53 km	NE 40 km	NE 73 km	NE 90 km	KE 58 km	LE 24 km	ST 29 km	TH 43 km	TH 162- 166 km	TH 203- 209 km	TH 216- 223 km	TH 239- 243 km
number analysed		4	5	5	5	9	10	10	10	8	2	5	4
average conc. [ $\mu\text{g}/\text{kg}$ ]		0.21	0.92	0.91	0.21	0.24	0.38	0.32	0.97	0.52	0.14	0.33	0.30
std dev [ $\mu\text{g}/\text{kg}$ ]		0.02	0.36	0.17	0.11	0.07	0.21	0.15	0.38	0.48	0.03	0.19	0.12
GL Pinch- beck West	GL 53 km		0.105	0.073	1.000	1.000	1.000	1.000	0.023	0.996	1.000	1.000	1.000
NE Cogenhoe	NE 40 km	0.105		1.000	0.019	0.040	0.148	0.092	1.000	0.751	0.061	0.245	0.446
NE Thrapston	NE 73 km	0.073	1.000		0.012	0.023	0.094	0.059	0.999	0.610	0.045	0.169	0.338
NE Oundle	NE 90 km	1.000	0.019	0.012		1.000	0.999	1.000	0.002	0.829	1.000	1.000	1.000
KE Newbury	KE 58 km	1.000	0.040	0.023	1.000		1.000	1.000	0.002	0.994	1.000	1.000	1.000
LE Wheat- hampstead	LE 24 km	1.000	0.148	0.094	0.999	1.000		1.000	0.014	1.000	0.995	1.000	1.000
ST Tednambury	ST 29 km	1.000	0.092	0.059	1.000	1.000	1.000		0.007	1.000	0.999	0.978	1.000
TH Castle Eaton	TH 43 km	0.023	1.000	0.999	0.002	0.002	0.014	0.007		0.282	0.022	0.060	0.168
TH Cav.- Sonning	TH 162- 166 km	0.996	0.751	0.610	0.829	0.994	1.000	1.000	0.282		0.838	1.000	1.000
TH Bray- Boveney	TH 203- 209 km	1.000	0.061	0.045	1.000	1.000	0.995	0.999	0.022	0.838		0.999	0.999
TH Old W.- Bell	TH 216- 223 km	1.000	0.245	0.169	1.000	1.000	1.000	0.978	0.060	1.000	0.999		1.000
TH Sunbury- Molesey	TH 239- 243 km	1.000	0.446	0.338	1.000	1.000	1.000	1.000	0.168	1.000	0.999	1.000	
significantly different to number of other groups		1/11	2/11	3/11	3/11	3/11	1/11	1/11	6/11	0/11	2/11	0/11	0/11

**Table S 8 Copper: Pairwise P of ANOVA (Holm-Sidak method) for Log(total copper). Pink shaded cells indicate that the difference between these groups was statistically significant at  $\alpha=0.05$ . Normality Test (Shapiro-Wilk) failed ( $p=0.002$ ), equal variance test passed at  $\alpha=0.05$ . Although the normality test had failed, a standard ANOVA was carried out to allow comparison with other chemicals.**

Copper		GL 53 km	NE 40 km	NE 73 km	NE 90 km	KE 58 km	LE 24 km	ST 29 km	TH 43 km	TH 162- 166 km	TH 187- 190 km	TH 203- 209 km	TH 216- 223 km	TH 243- 248 km
number analysed		5	9	10	9	9	10	10	10	10	5	8	5	10
average conc. [ $\mu\text{g}/\text{kg}$ ]		1397	1047	893	699	602	1619	904	618	717	587	801	632	901
std dev [ $\mu\text{g}/\text{kg}$ ]		471	569	433	158	127	448	624	178	111	266	247	209	334
GL Pinch- beck West	GL 53 km		0.973	0.549	0.050	0.004	1.000	0.189	0.004	0.075	0.007	0.321	0.034	0.656
NE Cogenhoe	NE 40 km	0.973		1.000	0.919	0.264	0.126	0.999	0.283	0.974	0.300	1.000	0.738	1.000
NE Thrapston	NE 73 km	0.549	1.000		1.000	0.822	0.007	1.000	0.853	1.000	0.797	1.000	0.989	1.000
NE Oundle	NE 90 km	0.050	0.919	1.000		1.000	<0.001	1.000	1.000	1.000	1.000	1.000	1.000	0.999
KE Newbury	KE 58 km	0.004	0.264	0.822	1.000		<0.001	0.995	0.993	1.000	1.000	0.992	1.000	0.732
LE Wheat- hampstead	LE 24 km	1.000	0.126	0.007	<0.001	<0.001		<0.001	<0.001	<0.001	<0.001	0.003	<0.001	0.011
ST Tednambury	ST 29 km	0.189	0.999	1.000	1.000	0.995	<0.001		0.997	1.000	0.985	0.999	1.000	1.000
TH Castle Eaton	TH 43 km	0.004	0.283	0.853	1.000	0.993	<0.001	0.997		1.000	1.000	0.995	0.949	0.762
TH Cav.- Sonning	TH 162- 166 km	0.075	0.974	1.000	1.000	1.000	<0.001	1.000	1.000		0.999	1.000	1.000	1.000
TH Temp- Marlow	TH 187- 190 km	0.007	0.300	0.797	1.000	1.000	<0.001	0.985	1.000	0.999		0.979	1.000	0.722
TH Bray- Boveney	TH 203- 209 km	0.321	1.000	1.000	1.000	0.992	0.003	0.999	0.995	1.000	0.979		1.000	1.000
TH Old W.- Bell	TH 216- 223 km	0.034	0.738	0.989	1.000	1.000	<0.001	1.000	0.949	1.000	1.000	1.000		0.976
TH Molesey- Kingston	TH 243- 248 km	0.656	1.000	1.000	0.999	0.732	0.011	1.000	0.762	1.000	0.722	1.000	0.976	
significantly different to number of other groups		4/12	0/12	1/12	1/12	2/12	10/12	1/12	2/12	1/12	2/12	1/12	2/12	1/12

**Table S 9**  $\Sigma$ 7PCBs: Pairwise P of ANOVA (Holm-Sidak method) for Log( $\Sigma$ 7PCBs). Pink shaded cells indicate that the difference between these groups was statistically significant at  $\alpha=0.05$ . Normality Test (Shapiro-Wilk) failed ( $p<0.001$ ), equal variance test passed at  $\alpha=0.05$ . Although the normality test had failed, a standard ANOVA was carried out to allow comparison with other chemicals.

$\Sigma$ 7 PCBs		GL 53 km	NE 40 km	NE 73 km	NE 90 km	KE 58 km	LE 24 km	ST 29 km	TH 43 km	TH 162- 166 km	TH 187- 190 km	TH 203- 209 km	TH 216- 223 km	TH 239- 243 km
number analysed		4	5	5	5	9	10	10	10	8	4	2	5	4
average conc. [ $\mu\text{g}/\text{kg}$ ]		3.1	12.5	9.4	6.3	4.8	17.5	17.9	11.9	15.4	16.0	10.7	11.8	13.8
std dev [ $\mu\text{g}/\text{kg}$ ]		0.7	2.2	2.7	1.9	1.1	5.7	7.2	2.7	4.9	7.4	4.9	6.0	1.2
GL Pinch- beck West	GL 53 km		<0.001	0.004	0.384	0.932	<0.001	<0.001	<0.001	<0.001	<0.001	0.033	<0.001	<0.001
NE Cogenhoe	NE 40 km	<0.001		0.999	0.202	0.002	0.998	1.000	1.000	1.000	1.000	1.000	1.000	1.000
NE Thrapston	NE 73 km	0.004	0.999		0.982	0.152	0.261	0.458	1.000	0.805	0.944	1.000	1.000	0.991
NE Oundle	NE 90 km	0.384	0.202	0.982		1.000	<0.001	0.002	0.143	0.009	0.052	0.991	0.757	0.118
KE Newbury	KE 58 km	0.932	0.002	0.152	1.000		<0.001	<0.001	<0.001	<0.001	<0.001	0.492	0.027	0.001
LE Wheat- hampstead	LE 24 km	<0.001	0.998	0.261	<0.001	<0.001		1.000	0.848	1.000	1.000	0.982	0.752	1.000
ST Tednambury	ST 29 km	<0.001	1.000	0.458	0.002	<0.001	1.000		0.974	1.000	0.998	0.995	0.912	1.000
TH Castle Eaton	TH 43 km	<0.001	1.000	1.000	0.143	<0.001	0.848	0.974		0.999	1.000	1.000	1.000	1.000
TH Cav.- Sonning	TH 162- 166 km	<0.001	1.000	0.805	0.009	<0.001	1.000	1.000	0.999		0.968	1.000	0.991	1.000
TH Temp- Marlow	TH 187- 190 km	<0.001	1.000	0.944	0.052	<0.001	1.000	0.998	1.000	0.968		1.000	0.998	0.991
TH Bray- Boveney	TH 203- 209 km	0.033	1.000	1.000	0.991	0.492	0.982	0.995	1.000	1.000	1.000		0.995	1.000
TH Old W.- Bell	TH 216- 223 km	<0.001	1.000	1.000	0.757	0.027	0.752	0.912	1.000	0.991	0.998	0.995		1.000
TH Sunbury- Molesey	TH 239- 243 km	<0.001	1.000	0.991	0.118	0.001	1.000	1.000	1.000	1.000	0.991	1.000	1.000	
significantly different to number of other groups		10/12	2/12	1/12	3/12	8/12	3/12	3/12	2/12	3/12	2/12	1/12	2/12	2/12

**Table S 10**  $\Sigma$ 6PBDEs: Pairwise P of ANOVA (Holm-Sidak method) for  $\text{Log}\Sigma(6\text{PBDEs})$ . Pink shaded cells indicate that the difference between these groups was statistically significant at  $\alpha=0.05$ . Normality Test (Shapiro-Wilk) failed ( $p<0.001$ ), equal variance test passed at  $\alpha=0.05$ . Although the normality test had failed, a standard ANOVA was carried out to allow comparison with other chemicals.

$\Sigma$ 6 PBDEs		GL 53 km	NE 40 km	NE 73 km	NE 90 km	KE 58 km	LE 24 km	ST 29 km	TH 43 km	TH 162- 166 km	TH 187- 190 km	TH 203- 209 km	TH 216- 223 km	TH 239- 243 km
number analysed		5	5	5	5	9	10	10	10	8	4	2	5	4
average conc. [ $\mu\text{g}/\text{kg}$ ]		3.0	22.8	29.9	13.5	3.6	25.2	16.6	16.5	7.1	9.3	14.5	6.3	10.1
std dev [ $\mu\text{g}/\text{kg}$ ]		1.0	15.0	9.1	4.6	1.0	7.8	6.2	4.8	2.3	3.3	7.9	1.2	1.7
GL Pinch- beck West	GL 53 km		<0.001	<0.001	<0.001	0.998	<0.001	<0.001	<0.001	0.017	0.005	0.001	0.123	<0.001
NE Cogenhoe	NE 40 km	<0.001		0.751	0.992	<0.001	0.959	0.998	0.998	0.006	0.400	0.995	0.007	0.722
NE Thrapston	NE 73 km	<0.001	0.751		0.088	<0.001	0.997	0.192	0.255	<0.001	0.002	0.555	<0.001	0.010
NE Oundle	NE 90 km	<0.001	0.992	0.088		<0.001	0.197	0.996	0.998	0.243	0.978	0.891	0.225	0.998
KE Newbury	KE 58 km	0.998	<0.001	<0.001	<0.001		<0.001	<0.001	<0.001	0.048	0.012	0.003	0.335	0.002
LE Wheat- hampstead	LE 24 km	<0.001	0.959	0.997	0.197	<0.001		0.375	0.501	<0.001	0.004	0.807	<0.001	0.020
ST Tednambury	ST 29 km	<0.001	0.998	0.192	0.996	<0.001	0.375		0.979	0.004	0.554	0.990	0.007	0.871
TH Castle Eaton	TH 43 km	<0.001	0.998	0.255	0.998	<0.001	0.501	0.979		0.002	0.453	0.999	0.004	0.804
TH Cav.- Sonning	TH 162- 166 km	0.017	0.006	<0.001	0.243	0.048	<0.001	0.004	0.002		0.995	0.649	0.980	0.936
TH Temp- Marlow	TH 187- 190 km	0.005	0.400	0.002	0.978	0.012	0.004	0.554	0.453	0.995		0.990	0.987	0.996
TH Bray- Boveney	TH 203- 209 km	0.001	0.995	0.555	0.891	0.003	0.807	0.990	0.999	0.649	0.990		0.555	0.998
TH Old W.- Bell	TH 216- 223 km	0.123	0.007	<0.001	0.225	0.335	<0.001	0.007	0.004	0.980	0.987	0.555		0.873
TH Sunbury- Molesey	TH 239- 243 km	<0.001	0.722	0.010	0.998	0.002	0.020	0.871	0.804	0.936	0.996	0.998	0.873	
significantly different to number of other groups		10/12	4/12	6/12	2/12	10/12	6/12	4/12	4/12	7/12	4/12	2/12	5/12	4/12

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