



Supplement of

Pesticides in the atmosphere: a comparison of gas-particle partitioning and particle size distribution of legacy and current-use pesticides

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Quantification of OCPs and CUPs

OCPs:

OCPs were analyzed and quantified by gas chromatography coupled to a tandem mass spectrometer (GC-MS/MS). Samples were analyzed with three slightly different sets of parameters/instrument configurations due to changes in laboratory instrumentation during the dates of sample collection and processing.

High volume air samples (PUF and filter) collected from the January to June 2012 were analyzed on an Agilent 6890N GC coupled to Waters Quattro Micro GC. The column used for chromatographic separation was 60 m x 0.25 mm x 0.25 μm DB5-MSUI (Agilent, J&W). Injection volume was 1 μL . The temperature program was: 80°C, 15°C/min to 180°C, 5°C/min to 300°C. The inlet temperature was 280°C. The carrier gas was He with a flow rate of 1.5 mL/min. High volume air samples (PUF and filter) collected from the June 2012 to January 2014 were analyzed on an Agilent 7000B GC/MS Triple Quadrupole, with SGE HT-8 (60 m x 0.25 mm x 0.25 μm) column. The injection volume was 3 μL . The temperature program was: 80°C (1.5 min), 40°C/min to 200°C (18 min), 5°C/min to 305°C (21 min). The inlet temperature was 280°C. The carrier gas was He with a flow rate of 1.5 mL/min.

Cascade impactor samples were analyzed for OCPs by GC-MS/MS (Agilent 6890N coupled to Waters Quattro Micro GC) with a J&W Scientific DB-5-MS fused silica column (60 m x 0.25 mm x 0.25 μm). The injection volume was 1 μL . The temperature program was: 80°C (1 min), 15°C/min to 180°C, 5°C/min to 300°C. Injection was splitless at 280°C, interface held at 280°C, He constant flow 1.5 mL/min.

LOQs of samples collected from January to December 2012 were determined from calibration curves and were 1 ng/mL for each compound. This corresponds to approximately 0.22 $\text{pg}\cdot\text{m}^{-3}$ with our active sampling configuration. LOQs of samples collected from January to December 2013 were determined for each compound and for each batch of samples by the quantification software (TargetLynx®) and were defined as a peak with the signal/noise ratio of 9 in the respective chromatograms of the lowest point of the calibration curve (1 ng/mL). For those samples, LOQs ranged from 37.7 to 365 $\text{pg}\cdot\text{mL}^{-1}$ which corresponds to about 0.008 to 0.08 $\text{pg}\cdot\text{m}^{-3}$ with our sampling configuration. Finally, for cascade impactor samples, LOQs were also determined by TargetLynx® for each compound and each sample in the sample chromatograms. LOQs of individual samples ranged from 0.014 to 15 $\text{ng}\cdot\text{mL}^{-1}$ which corresponds to 0.0005 to 0.54 $\text{pg}\cdot\text{m}^{-3}$ for our active sampler configuration.

CUPs:

Separation of CUPs was performed by LC-MS using an Agilent 1290 HPLC (Agilent Technologies, Waldbronn, Germany) consisting of a vacuum degasser, a binary pump, a thermostated autosampler (10°C), and a thermostated column compartment kept at 30°C. The column was a Phenomenex Synergi Fusion C-18 endcapped (3 µm) 100 x 2.1 mm i.d., equipped with a Phenomenex SecureGuard C18 guard column (Phenomenex, Torrance, CA, USA). The mobile phase consisted of 5 mM ammonium acetate in water (A) and 5 mM ammonium acetate in methanol (B). The binary pump gradient was non-linear (increase from 20% B at 0 min to 80% B at 1 min, then increase to 90% B at 5 min, then 90% B for 8 min and 5 min column equilibration at initial conditions (20% B)); the flow rate was 0.25 mL/min. 10 µL of individual sample was injected for the analyses. CUPs were quantified using a mass spectrometer (AB Sciex Qtrap 5500, AB Sciex, Concord, ON, Canada) with electrospray ionization (ESI). Ions were detected in the positive mode. The ionization parameters were as follows: capillary voltage, 5.5 kV; desolvation temperature, 400°C; Curtain gas 15 psi, Gas1 40 psi, Gas2 30 psi. The m/z transitions presented in Table S5 were monitored in scheduled MRM mode (with corresponding values of declustering potential – DP (V), entrance potential – EP (V), collision cell exit potential – CXP(V) and collision energy – CE (V)). Values of method quantification limits (MQL) and retention times are also presented in Table S5. LODs and LOQs were estimated as the quantity of analyte with a signal to noise ratio of 3:1 and 10:1, respectively. The method was validated by quantifying spiked pre-cleaned PUFs and QFFs, for which the recoveries of individual CUPs ranged from 52.4% ± 21.4 for disulfoton to 115% ± 17.4 for alachlor (Table S9).

Temperature-dependence of OCPs and CUPs

The gas-phase behavior of SVOCs can be described by the Clausius-Clapeyron equation:

$$\ln P = (-\Delta H_v/R)(1/T) + \text{constant} \quad (\text{Eq. S1})$$

where P is partial pressure (Pa), T is temperature (K), ΔH_v is enthalpy of vaporization ($\text{kJ}\cdot\text{mol}^{-1}$) and R is the gas constant. The temperature dependence of atmospheric pesticide concentrations was expressed as the linear regressions of the natural logarithm of partial pressure versus the inverse of the temperature:

$$\ln P = m/T + b, \text{ where } m \text{ and } b \text{ are constants.} \quad (\text{Eq. S2})$$

Partial pressures of individual compounds were calculated for each sampling event using gas phase concentrations and the ideal gas law.

Gas-particle partitioning

Partitioning of organic compounds such as pesticides between the gas and particle phases is often described using the gas/particle partition coefficient, K_p (in $\text{m}^3 \cdot \mu\text{g}^{-1}$) defined by Harner and Bidleman (1998b):

$$K_p = (C_p/C_{\text{TSP}})/C_g \quad (\text{Eq. S3})$$

where C_p and C_g are the concentrations of the pesticides (in $\text{pg} \cdot \text{m}^{-3}$) in the particulate and gaseous phases, respectively and C_{TSP} is the concentration of the total suspended particles (TSP) in the air (in $\mu\text{g} \cdot \text{m}^{-3}$).

K_p can be predicted using the octanol-air partition coefficient (K_{OA}). This assumes that absorption into particulate organic matter of the particles determines the distribution process, while other types of molecular interaction are neglected (Harner and Bidleman, 1998b).

Then, K_p can be defined as:

$$K_p = \frac{f_{\text{OM}} \text{MW}_{\text{OCT}} \zeta_{\text{OCT}}}{\rho_{\text{OCT}} \text{MW}_{\text{OM}} \zeta_{\text{OM}} 10^{12}} K_{\text{OA}} \quad (\text{Eq. S4})$$

where f_{OM} is the fraction of organic matter (OM) phase on particles, MW_{OCT} and MW_{OM} are the mean molecular weights (in $\text{g} \cdot \text{mol}^{-1}$) of octanol and OM, respectively, ρ_{OCT} is the density of octanol ($0.820 \text{ kg} \cdot \text{L}^{-1}$), ζ_{OCT} is the activity coefficient of the absorbing compound in octanol and ζ_{OM} is the activity coefficient of the compound in the OM phase. With the assumptions that $\zeta_{\text{OCT}} / \zeta_{\text{OM}}$ and $\text{MW}_{\text{OCT}} / \text{MW}_{\text{OM}} = 1$, equation S4 can be written as:

$$\log K_p = \log K_{\text{OA}} + \log f_{\text{OM}} - 11.91 \quad (\text{Eq. S5})$$

Finally, predicted particulate fractions (θ_{pr}) can be calculated using:

$$\theta_{\text{pr}} = K_p C_{\text{TSP}} / (1 + K_p C_{\text{TSP}}) \quad (\text{Eq. S6})$$

In this study, a f_{OM} of 0.2 was used and determined to be reasonable for this background site, based on a research on particles at the European level (Putaud et al., 2004). As TSP values were not available at the sampling location, the hourly PM_{10} concentrations at this site from 2012 were used instead (data provided by the Czech Hydrometeorological Institute, <http://www.chmi.cz>)

It should be noted that the K_{OA} -based model might not give the best results for the more polar CUPs (dimethoate, metamitron, pyrazon and carbendazim) (Götz et al., 2007; Sadiki and Poissant, 2008) because this model may underestimate polar interactions and thus K_p (Götz et al., 2007). However, this method has previously produced reasonable results for CUPs (Sauret et al., 2008; Yao et al., 2008). Moreover, given

the limited available data for CUPs and the desire for comparability between OCPs and CUPs within this dataset, determination of K_p using K_{OA} was the best choice for the current study.

Temperature dependence of octanol-air partition coefficient (K_{OA})

Due to the large enthalpy change involved in octanol to air transfer, a strong temperature dependence of the octanol-air partition coefficient (K_{OA}) is expected (Harner and Mackay, 1995). When feasible, temperature dependence of K_{OA} of the studied pesticides was determined from published relationships, based on direct measurements (Odabasi and Cetin, 2012a; Shoeib and Harner, 2002a; Zhang et al., 2009). However for most CUPs, no information on the temperature dependence of K_{OA} was available. In lieu of this, temperature adjusted K_{OA} values were determined from an extrapolation of available experimental values determined for organochlorine pesticides (OCPs) (Odabasi and Cetin, 2012b; Shoeib and Harner, 2002b; Zhang et al., 2009), chlorobenzenes (CBs) (Harner and Mackay, 1995), polychlorinated biphenyls (PCBs) (Harner and Mackay, 1995), polycyclic aromatic hydrocarbons (PAHs) (Odabasi et al., 2006a), carbazole (CAR) (Odabasi et al., 2006b) and polychlorinated naphthalenes (PCNs) (Harner and Bidleman, 1998a). Except for prochloraz and temephos ($\log K_{OA}=13.6$ and 13.1 , respectively), the range of $\log K_{OA}$ of CUPs (7.49-11.2) is a sub-range of the range of $\log K_{OA}$ (4.36-12.6) spanned by these classes of compounds. To this end, the $\log K_{OA}$ values of OCPs, CBs, PCBs, PAHs, CAR and PCNs for each sampling temperature were plotted versus their corresponding values at the reference temperature (25°C). Examples of these plots are provided in Figure S3. Finally, using the linear regression relationship from these plots ($r^2 > 0.992$), $\log K_{OA}$ values of CUPs at the specific temperatures were calculated. $\log K_{OA}$ values of OCPs and CUPs at 25°C , T_{\min} (winter) and T_{\max} (summer) are shown in Table S15. We tested this method for chlorpyrifos and the differences in $\log K_{OA}$ values at different temperatures determined based on the experimental values and those extrapolated from the regression analysis were negligible (Table S15).

Table S1: Air sample volumes collected for OCP analysis from background site. Sample volumes were corrected for temperature and pressure.

Sampling start	Sampling end	Sampling volume (m ³)
11/01/2012	18/01/2012	4559.3
25/01/2012	01/02/2012	4576.5
08/02/2012	15/02/2012	8103.3
22/02/2012	29/02/2012	4532.5
07/03/2012	14/03/2012	4532.4
21/03/2012	28/03/2012	4471.1
04/04/2012	11/04/2012	4441.9
18/04/2012	25/04/2012	4394.2
02/05/2012	09/05/2012	4416.0
16/05/2012	23/05/2012	4405.2
30/05/2012	06/06/2012	4416.0
13/06/2012	20/06/2012	4385.3
27/06/2012	04/07/2012	3907.9
11/07/2012	18/07/2012	4419.2
25/07/2012	01/08/2012	4354.2
08/08/2012	15/08/2012	4423.9
22/08/2012	29/08/2012	4442.2
05/09/2012	12/09/2012	4405.4
19/09/2012	26/09/2012	4432.3
03/10/2012	10/10/2012	4452.7
17/10/2012	24/10/2012	4464.8
31/10/2012	07/11/2012	4466.7
14/11/2012	21/11/2012	3791.0
28/11/2012	05/12/2012	5055.8
12/12/2012	19/12/2012	4533.8
26/12/2012	02/01/2013	4550.3
09/01/2013	16/01/2013	4572.9
23/01/2013	30/01/2013	4478.6
06/02/2013	13/02/2013	4510.9
20/02/2013	27/02/2013	4550.5
06/03/2013	13/03/2013	4425.3
20/03/2013	27/03/2013	4561.0
03/04/2013	10/04/2013	4484.9
17/04/2013	24/04/2013	4435.5
01/05/2013	08/05/2013	4420.2
15/05/2013	22/05/2013	4355.1
29/05/2013	05/06/2013	4421.9
12/06/2013	19/06/2013	4364.2
26/06/2013	03/07/2013	1929.2
10/07/2013	17/07/2013	4393.7
24/07/2013	31/07/2013	3434.6
07/08/2013	14/08/2013	920.4
21/08/2013	28/08/2013	4435.9
04/09/2013	11/09/2013	3050.3
18/09/2013	25/09/2013	4453.2
02/10/2013	09/10/2013	4497.8
16/10/2013	23/10/2013	4416.5
30/10/2013	06/11/2013	4433.0
13/11/2013	20/11/2013	4515.6
27/11/2013	04/12/2013	4539.5
11/12/2013	18/12/2013	4562.4
25/12/2013	01/01/2014	4473.2

Table S2: Air sample volumes collected for CUPs analysis from background site. Sample volumes were corrected for temperature and pressure.

Sampling start	Sampling end	Sampling volume (m ³)
04/01/2012	11/01/2012	4529.2
18/01/2012	25/01/2012	4540.0
01/02/2012	08/02/2012	4083.9
15/02/2012	22/02/2012	4550.7
29/02/2012	07/03/2012	4509.3
14/03/2012	21/03/2012	4487.2
28/03/2012	04/04/2012	4455.0
11/04/2012	18/04/2012	4423.1
25/04/2012	02/05/2012	4387.0
09/05/2012	16/05/2012	4430.8
23/05/2012	30/05/2012	4413.2
06/06/2012	13/06/2012	4405.9
20/06/2012	27/06/2012	4078.0
04/07/2012	11/07/2012	3210.0
18/07/2012	25/07/2012	3042.6
01/08/2012	08/08/2012	4373.7
15/08/2012	22/08/2012	4408.9
29/08/2012	05/09/2012	4409.5
12/09/2012	19/09/2012	4395.0
26/09/2012	03/10/2012	4436.4
10/10/2012	17/10/2012	4465.4
24/10/2012	31/10/2012	4503.7
07/11/2012	14/11/2012	4472.3
22/11/2012	28/11/2012	3801.2
05/12/2012	12/12/2012	4561.7
19/12/2012	26/12/2012	4541.7
02/01/2013	09/01/2013	4545.5
16/01/2013	23/01/2013	4504.0
30/01/2013	06/02/2013	4483.7
13/02/2013	20/02/2013	4541.4
27/02/2013	06/03/2013	4524.2
13/03/2013	20/03/2013	4505.0
27/03/2013	03/04/2013	4846.6
10/04/2013	17/04/2013	4448.4
24/04/2013	01/05/2013	4403.6
08/05/2013	15/05/2013	4411.9
22/05/2013	29/05/2013	4413.1
05/06/2013	12/06/2013	4379.3
19/06/2013	26/06/2013	4376.0
03/07/2013	10/07/2013	4388.4
17/07/2013	24/07/2013	4357.3
31/07/2013	07/08/2013	576.7
14/08/2013	21/08/2013	1886.4
28/08/2013	04/09/2013	4432.6
11/09/2013	18/09/2013	4452.3
25/09/2013	02/10/2013	4473.0
09/10/2013	16/10/2013	4467.6
23/10/2013	30/10/2013	4404.1
06/11/2013	13/11/2013	4446.2
20/11/2013	27/11/2013	4480.9
04/12/2013	11/12/2013	4515.9
18/12/2013	25/12/2013	4495.4

Table S3: Cascade impactor air sample volumes collected for OCPs analysis. Sample volumes were corrected for temperature and pressure.

		Start of sampling	End of sampling	Volume sampled (m ³)
Rural site	Winter	6/1/2010	13/1/2010	11014
		3/2/2010	9/2/2010	9604
		5/3/2010	12/3/2010	11424
	Spring	9/4/2010	16/4/2010	11162
		7/5/2010	10/5/2010	2397
		7/6/2010	14/6/2010	11103
	Summer	5/7/2010	11/7/2010	5957
		2/8/2010	9/8/2010	11425
		9/9/2010	15/9/2010	8313
	Autumn	30/10/2009	3/11/2009	6659
		20/11/2009	27/11/2009	11424
		8/10/2010	15/10/2010	7297
Urban site	Winter	6/1/2010	13/1/2010	11141
		3/2/2010	10/2/2010	11364
		3/3/2010	10/3/2010	11316
	Spring	9/4/2010	16/4/2010	10915
		7/5/2010	10/5/2010	4293
		14/6/2010	21/6/2010	11245
	Summer	12/7/2010	19/7/2010	11420
		16/8/2010	23/8/2010	10775
		15/9/2010	18/9/2010	4755
	Autumn	30/10/2009	2/11/2009	4909
		20/11/2009	27/11/2009	10964
		8/10/2010	15/10/2010	10863

Table S4: Cascade impactor air sample volumes collected for CUPs analysis. Sample volumes were corrected for temperature and pressure.

		Start of sampling	End of sampling	Volume sampled (m ³)
Rural site	Winter	27/11/2009	4/12/2009	11425
		13/1/2010	20/1/2010	11289
		10/2/2010	13/2/2010	4443
	Spring	12/3/2010	19/3/2010	11424
		16/4/2010	23/4/2010	11347
		13/5/2010	20/5/2010	11425
	Summer	14/6/2010	21/6/2010	11258
		12/7/2010	19/7/2010	10529
		16/8/2010	23/8/2010	9770
	Autumn	9/10/2009	16/10/2009	11353
		16/9/2010	23/9/2010	11425
	Urban site	Winter	27/11/2009	4/12/2009
13/1/2010			20/1/2010	11283
10/2/2010			17/2/2010	11411
Spring		10/3/2010	17/3/2010	11424
		16/4/2010	22/4/2010	9453
		21/5/2010	28/5/2010	11371
Summer		22/6/2010	25/6/2010	6049
		19/7/2010	26/7/2010	11117
		23/8/2010	26/8/2010	5005
Autumn		9/10/2009	16/10/2009	11181
		22/9/2010	29/9/2010	10700

Table S5: Selected LC-MS/MS experimental parameters, limits of detection (LODs) and limits of quantification (LOQs) (SRM indicates “Selected Reaction Monitoring” and R_t indicates “Retention time”).

Analyte	m/z	SRM 1	SRM 2	R _t (min)	LOD (ng/mL)	LOQ (ng/mL)
Acetochlor	270.1	224.2	148.1	4.2	0.1	0.25
Alachlor	270.1	238.1	162.1	4.2	0.5	1
Atrazine	216.1	174.2	68.0	3.5	0.05	0.1
Azinphos methyl	318.0	160.0	132.0	3.7	0.1	0.25
Carbendazim	192.0	160.0	131.9	2.9	0.03	0.05
Chlorpyrifos	349.9	96.9	197.9	6.1	0.25	0.5
Chlorotoluron	213.1	72.2	46.2	3.4	0.25	0.5
Diazinon	305.0	169.0	96.9	4.6	0.01	0.25
Dimethachlor	256.1	224.0	148.1	3.5	0.2	0.5
Dimethoate	230.0	198.9	124.9	2.8	0.01	0.03
Disulfoton	275.0	88.9	61.0	4.8	0.01	0.02
Diuron	232.9	71.8	46.1	3.5	0.1	0.25
Fenitrothion	278.0	124.9	108.9	4.0	5	20
Fenpropimorph	304.2	147.0	117.0	8.2	0.02	0.05
Fonofos	247.0	109.0	136.9	4.6	0.1	0.25
Isoproturon	207.2	72.1	46.1	3.5	0.06	0.12
Malathion	331.0	127.0	99.0	3.9	0.25	0.5
Metamitron	203.0	175.1	104.0	2.8	0.25	0.5
Metazachlor	278.2	134.2	210.2	3.8	0.05	0.1
Metribuzin	215.1	187.1	84.0	3.2	0.1	0.25
Prochloraz	376.0	308.0	70.0	4.7	0.01	0.03
Pyrazon	222.0	104.0	77.0	2.8	0.06	0.12
Simazine	202.0	132.0	124.1	3.2	0.05	0.1
S-metolachlor	284.1	252.2	176.1	4.2	0.1	0.25
Temephos	466.9	419.0	124.9	5.7	0.03	0.05
Terbufos	289.0	103.0	232.9	5.7	0.05	0.1
Terbuthylazine	230.1	174.0	67.9	3.9	0.05	0.1

Breakthrough

Breakthrough of gas-phase compounds was evaluated by quantifying separately each of the two PUFs placed in series for all the weekly air samples (sample volume = 3042-4561 m³ for CUPs and 3791-8103 m³ for OCPs) collected at the background site in 2012 (N=52). This covered a large range of meteorological conditions, and the results are considered applicable to 2013. Summary of the results of the breakthrough experiments is presented in Tables S5 and S6. Based on these results, the current sampling configuration with two PUFs in series is considered to be efficient for trapping the selected OCPs and CUPs in the gas phase.

Table S6: Results of breakthrough experiments for CUPs. N/D indicates compounds that were not detected.

Compound	Frequency of detection on upper PUF	Frequency of detection on lower PUF	% of compound mass found on the lower PUF			
			Min	Max	Average	Standard deviation
Acetochlor	50%	27%	0	34.5	10.4	13.6
Alachlor	3.8%	0%	0	0	0	N/D
Atrazine	3.8%	0%	0	0	0	N/D
Azinphos methyl	0%	0%	N/D	N/D	N/D	N/D
Carbendazim	0%	3.8%	100	100	100	N/D
Chlorpyrifos	92%	35%	0	22.9	2.86	5.95
Chlorotoluron	3.8%	0%	0	0	0	N/D
Diazinon	3.8%	0%	0	0	0	N/D
Dimethachlor	38%	15%	0	24.6	3.38	7.69
Dimethoate	0%	0%	N/D	N/D	N/D	N/D
Disulfoton	0%	0%	N/D	N/D	N/D	N/D
Diuron	0%	0%	N/D	N/D	N/D	N/D
Fenitrothion	0%	0%	N/D	N/D	N/D	N/D
Fenpropimorph	0%	3.8%	100	100	100	N/D
Fonofos	0%	0%	N/D	N/D	N/D	N/D
Isoproturon	54%	3.8%	0	50.2	3.59	13.4
Malathion	0%	0%	N/D	N/D	N/D	N/D
Metamitron	15%	3.8%	0	100	20.0	44.7
Metazachlor	69%	19%	0	21.6	1.48	5.14
Metribuzin	7.7%	3.8%	0	100	33.3	57.7
Prochloraz	0%	0%	N/D	N/D	N/D	N/D
Pyrazon	0%	0%	N/D	N/D	N/D	N/D
Simazine	0%	0%	N/D	N/D	N/D	N/D
S-metolachlor	69%	23%	0	11.0	1.16	2.71
Temephos	0%	0%	N/D	N/D	N/D	N/D
Terbufos	0%	0%	N/D	N/D	N/D	N/D
Terbutylazine	73%	23%	0	3.34	0.415	0.879

Table S7: Results of breakthrough experiments for OCPs.

Compound	Detection frequency on upper PUF	Detection frequency on lower PUF	% of compound mass found on the lower PUF			
			Min	Max	Average	Standard deviation
α -HCH	100%	100%	0.04	65.6	35.4	17.0
β -HCH	100%	88%	0	26.6	9.97	8.81
γ -HCH	100%	100%	1.03	49.3	21.9	18.0
δ -HCH	100%	96%	0	66.4	22.8	15.7
<i>o,p'</i> -DDE	100%	465	0	21.8	1.78	5.03
<i>p,p'</i> -DDE	100%	85%	0	22.0	2.01	5.03
<i>o,p'</i> -DDD	100%	35%	0	20.4	1.76	5.00
<i>p,p'</i> -DDD	100%	77%	0	18.6	2.22	4.58
<i>o,p'</i> -DDT	100%	73%	0	19.3	1.61	4.23
<i>p,p'</i> -DDT	100%	65%	0	16.1	1.40	3.50

Table S8: Recoveries (in %) and standard deviations (SD) of individual OCPs. Average recoveries were calculated from spike and recovery tests of PUFs (n=3).

Compound	Average recovery (%)	SD
α -HCH	94.2	6.31
β -HCH	91.2	6.10
γ -HCH	94.4	6.86
δ -HCH	99.1	7.83
<i>o,p'</i> -DDE	88.7	5.69
<i>p,p'</i> -DDE	87.2	6.26
<i>o,p'</i> -DDD	93.4	7.14
<i>p,p'</i> -DDD	91.4	8.28
<i>o,p'</i> -DDT	104.9	5.44
<i>p,p'</i> -DDT	113.8	6.10

Table S9: Recoveries (in %) and standard deviations (SD) of individual CUPs. Average recoveries were calculated from spike and recovery tests of air sampling media (filters and PUFs, n=10).

Compound	QFFs		PUFs	
	Average recovery (%)	SD	Average recovery (%)	SD
Acetochlor	97.2	13.2	93.2	17.3
Alachlor	81.4	6.05	115	17.4
Atrazine	90.5	9.67	82	15.8
Azinphos methyl	64.4	12.7	68.5	15.7
Carbendazim	58.1	4.45	83.3	9.6
Chlorotoluron	65.9	14.6	100	16.8
Chlorpyrifos	79.5	6.68	111	17.3
Diazinon	103	21.7	18.9	12.9
Dimethachlor	74.8	6.76	86.1	16
Dimethoate	77	8.44	98.1	15.8
Disulfoton	90.2	17.3	52.4	21.4
Diuron	59.8	14.4	102	13.9
Fenitrothion	81.2	36.5	56.3	19.5
Fenpropimorph	80.3	14.2	103.6	16.7
Fonofos	90.4	21.7	114	14.8
Isoproturon	113	23.7	109	17.8
Malathion	55.2	7.14	58	18.1
Metamitron	83.4	7.38	81.2	12.3
Metazachlor	58.9	9.48	107.1	18.7
Metribuzin	102	21.8	52.8	15.9
Prochloraz	81.5	12.2	78	16.7
Pyrazon	90.5	4.11	104	17.6
Simazine	80.7	8.3	81.3	15.5
S-metolachlor	109	10.1	109.8	19.2
Temephos	60.3	18.2	85.6	15.9
Terbufos	84.9	5.56	66.3	16.6
Terbutylazine	70.5	7.65	96	15.6

Table S10: Use of agricultural pesticides in the Czech Republic in 2012 (in kg) (SRS, 2013).

Compound	Total	Cereals	Maize	Legumes	Beets	Potatoes	Forage crops	Oil plants	Hops	Vegetables	Fruits	Grapes (vineyards)	Others
Acetochlor	173604	93	151815	3	0	0	0	21693	0	0	0	0	0
Carbendazim	42392	19018	0	6	855	4	0	22467	0	43	0	0	0
Chlorpyrifos	187245	41511	3124	3241	6779	1187	175	131217	0	0	8	0	3
Chlorotoluron	91315	89533	67	0	19	0	59	1637	0	0	0	0	0
Dimethachlor	54927	352	0	0	0	0	0	54574	0	0	0	0	0
Dimethoate	883	815	0	0	29	36	0	2	0	1	0	0	0
Fenpropimorph	66599	63116	0	0	3480	0	3	0	0	0	0	0	0
Isoproturon	141164	140976	9	0	33	0	2	145	0	0	0	0	0
Metamitron	94564	0	0	0	94123	0	8	426	0	5	3	0	0
Metazachlor	186977	444	0	0	0	3	0	185957	0	571	0	0	3
Metribuzin	4579	0	0	94	0	4324	49	0	0	111	0	0	0
Prochloraz	148446	117271	0	0	7789	0	0	23386	0	0	0	0	0
Pyrazon	40359	1	0	0	40358	0	0	0	0	0	0	0	0
S-Metolachlor	76871	244	72959	101	2077	348	0	1143	0	0	0	0	0
Terbutylazine	111639	228	111346	0	0	0	0	65	0	0	0	0	0
Total (selected pesticides)	1421563	473603	339320	3444	155541	5901	297	442712	0	730	11	0	6
Total (all pesticides)	5701697	2639442	599455	35410	318712	104919	43275	1416009	88792	27102	229389	169419	29773

Table S11: Use of agricultural pesticides in the Czech Republic in 2013 (in kg) (SRS, 2014).

Compound	Total	Cereals	Maize	Legumes	Beets	Potatoes	Forage crops	Oil plants	Hops	Vegetables	Fruits	Grapes (Vineyards)	Others
Acetochlor	101053	256	91002	0	0	0	0	9795	0	0	0	0	0
Carbendazim	31050	11186	16	0	701	0	0	19099	0	48	0	0	0
Chlorotoluron	107418	105063	45	0	0	0	37	2273	0	0	0	0	0
Chlorpyrifos	178363	29436	2111	3026	5525	1125	160	136915	0	65	0	0	0
Dimethachlor	44101	5	0	92	0	0	0	43783	0	191	0	0	30
Dimethoate	147	20	0	0	82	45	0	0	0	1	0	0	0
Fenpropimorph	79620	75795	2	0	3790	0	15	19	0	0	0	0	0
Isoproturon	94004	93858	26	0	0	0	80	40	0	0	0	0	0
Metamitron	94347	49	0	0	94158	0	0	0	0	45	0	0	96
Metazachlor	169986	393	0	0	35	0	0	168788	0	750	0	0	19
Metribuzin	4606	0	5	23	0	4488	23	10	0	42	0	0	16
Prochloraz	176504	143665	9	0	4636	0	0	28125	0	0	0	0	70
Pyrazon	43372	0	0	0	43229	0	0	0	0	34	0	0	109
S-Metolachlor	68577	1073	64090	33	1990	315	0	1076	0	1	0	0	0
Terbutylazine	113124	802	112102	0	0	0	0	195	0	1	0	0	24
Total (selected pesticides)	1306270	461600	269408	3174	154146	5973	316	410116	0	1176	0	0	362
Total (all pesticides)	5511032	2480955	560947	30239	299283	107638	36680	1402589	81403	28488	252425	208568	21820

Table S12: Concentration of individual CUPs (in $\mu\text{g}\cdot\text{m}^{-3}$) in the gaseous phase at the background site. N/D indicates compounds that were not detected, N/A those that were not analyzed, and <LOQ indicates samples that were below the limit of quantification (in Table S7).

Sampling start	Sampling end	Acetochlor	Alachlor	Atrazine	Azinphos methyl	Carbendazim	Chlorotoluron	Chlorpyrifos	Diazinon	Dimethachlor	Dimethoate	Disulfoton	Diuron	Fenitrothion	Fenpropimorph
4.1.2012	11.1.2012	N/D	N/D	N/D	N/D	N/D	N/D	0.119	N/D	N/D	N/D	N/D	N/D	N/D	N/D
18.1.2012	25.1.2012	N/D	N/D	N/D	N/D	N/D	N/D	0.260	N/D	N/D	N/D	N/D	N/D	N/D	N/D
1.2.2012	8.2.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
15.2.2012	22.2.2012	N/D	N/D	N/D	N/D	N/D	N/D	0.128	N/D	N/D	N/D	N/D	N/D	N/D	N/D
29.2.2012	7.3.2012	N/D	N/D	N/D	N/D	N/D	N/D	0.517	N/D	N/D	N/D	N/D	N/D	N/D	N/D
14.3.2012	21.3.2012	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	N/D	N/D	N/D	N/D
28.3.2012	4.4.2012	N/D	N/D	N/D	N/D	N/D	N/D	18.635	N/D	N/D	N/D	N/D	N/D	N/D	N/D
11.4.2012	18.4.2012	10.197	N/D	N/D	N/D	N/D	N/D	31.200	N/D	N/D	N/D	N/D	N/D	N/D	N/D
25.4.2012	2.5.2012	30.134	N/D	N/D	N/D	N/D	N/D	6.218	N/D	0.944	N/D	N/D	N/D	N/D	N/D
9.5.2012	16.5.2012	157.828	<LOQ	N/D	N/D	N/D	N/D	2.753	N/D	0.422	N/D	N/D	N/D	N/D	N/D
23.5.2012	30.5.2012	24.379	0.231	0.757	N/D	N/D	N/D	2.148	N/D	0.134	N/D	N/D	N/D	N/D	N/D
6.6.2012	13.6.2012	33.367	<LOQ	N/D	N/D	0.223	N/D	3.155	N/D	<LOQ	N/D	N/D	N/D	N/D	<LOQ
20.6.2012	27.6.2012	8.646	N/D	N/D	N/D	N/D	0.390	7.540	N/D	0.220	N/D	N/D	N/D	N/D	N/D
4.7.2012	11.7.2012	3.003	N/D	N/D	N/D	N/D	N/D	6.732	N/D	N/D	N/D	N/D	N/D	N/D	1.271
18.7.2012	25.7.2012	0.670	N/D	N/D	N/D	N/D	N/D	7.418	N/D	N/D	N/D	N/D	N/D	N/D	N/D
1.8.2012	8.8.2012	0.474	N/D	N/D	N/D	N/D	N/D	1.877	N/D	N/D	N/D	N/D	N/D	N/D	N/D
15.8.2012	22.8.2012	0.152	N/D	N/D	N/D	N/D	N/D	0.148	N/D	47.699	N/D	N/D	N/D	N/D	N/D
29.8.2012	5.9.2012	0.730	N/D	N/D	N/D	N/D	N/D	6.047	0.177	70.805	N/D	N/D	N/D	N/D	N/D
12.9.2012	19.9.2012	N/D	N/D	N/D	N/D	N/D	N/D	1.515	N/D	6.497	N/D	N/D	N/D	N/D	N/D
26.9.2012	3.10.2012	0.651	<LOQ	N/D	N/D	N/D	N/D	11.094	N/D	1.501	N/D	N/D	N/D	N/D	N/D
10.10.2012	17.10.2012	0.157	N/D	N/D	N/D	N/D	N/D	22.842	N/D	0.425	N/D	N/D	N/D	N/D	N/D
24.10.2012	31.10.2012	N/D	N/D	N/D	N/D	N/D	N/D	64.355	N/D	0.133	N/D	N/D	N/D	N/D	N/D
7.11.2012	14.11.2012	N/D	N/D	N/D	N/D	N/D	N/D	17.105	N/D	N/D	N/D	N/D	N/D	N/D	N/D

22.11.2012	28.11.2012	N/D	N/D	N/D	N/D	N/D	N/D	6.814	N/D	N/D	N/D	N/D	N/D	N/D	N/D
5.12.2012	12.12.2012	N/D	N/D	N/D	N/D	N/D	N/D	0.212	N/D	N/D	N/D	N/D	N/D	N/D	N/D
19.12.2012	26.12.2012	N/D	N/D	N/D	N/D	N/D	N/D	1.519	N/D	N/D	N/D	N/D	N/D	N/D	N/D
2.1.2013	9.1.2013	N/D	N/D	N/D	N/D	N/D	N/D	0.814	N/D	N/D	N/D	N/D	N/D	N/D	N/D
16.1.2013	23.1.2013	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	N/D	N/D	N/D	N/D
30.1.2013	6.2.2013	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
13.2.2013	20.2.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
27.2.2013	6.3.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
13.3.2013	20.3.2013	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	N/D	N/D	N/D	N/D
27.3.2013	3.4.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.042
10.4.2013	17.4.2013	N/D	N/D	N/D	N/D	N/D	N/D	1.538	N/D	N/D	N/D	N/D	N/D	N/D	0.103
24.4.2013	1.5.2013	40.649	N/D	N/D	N/D	N/D	N/D	157.827	N/D	0.534	N/D	N/D	N/D	N/D	N/D
8.5.2013	15.5.2013	118.997	N/D	N/D	N/D	N/D	N/D	36.039	N/D	N/D	N/D	N/D	N/D	N/D	N/D
22.5.2013	29.5.2013	17.040	N/D	N/D	N/D	N/D	N/D	3.240	N/D	0.442	N/D	N/D	N/D	N/D	N/D
5.6.2013	12.6.2013	2.466	N/D	N/D	N/D	N/D	N/D	3.791	N/D	N/D	N/D	N/D	N/D	N/D	N/D
19.6.2013	26.6.2013	10.626	N/D	N/D	N/D	N/D	N/D	14.671	N/D	N/D	N/D	N/D	N/D	N/D	N/D
3.7.2013	10.7.2013	1.199	N/D	0.206	N/D	N/D	N/D	17.982	N/D	N/D	N/D	N/D	N/D	N/D	N/D
17.7.2013	24.7.2013	N/D	N/D	N/D	N/D	N/D	N/D	17.043	N/D	N/D	N/D	N/D	N/D	N/D	0.019
31.7.2013	7.8.2013	N/D	N/D	N/D	N/D	N/D	N/D	2.746	N/D	N/D	N/D	N/D	N/D	N/D	N/D
14.8.2013	21.8.2013	0.981	N/D	N/D	N/D	N/D	N/D	1.280	N/D	6.467	N/D	N/D	N/D	N/D	N/D
28.8.2013	4.9.2013	0.575	N/D	0.084	N/D	N/D	N/D	2.449	N/D	60.233	N/D	N/D	N/D	N/D	N/D
11.9.2013	18.9.2013	N/D	N/D	0.023	N/D	N/D	N/D	1.831	N/D	5.664	N/D	N/D	N/D	N/D	N/D
25.9.2013	2.10.2013	0.984	N/D	N/D	N/D	N/D	N/D	10.126	N/D	3.957	N/D	N/D	N/D	N/D	N/D
9.10.2013	16.10.2013	N/D	N/D	N/D	N/D	N/D	0.427	36.491	N/D	0.898	N/D	N/D	N/D	N/D	N/D
23.10.2013	30.10.2013	0.284	N/D	0.096	N/D	N/D	0.483	64.530	N/D	0.602	N/D	N/D	N/D	N/D	N/D
6.11.2013	13.11.2013	N/D	N/D	N/D	N/D	N/D	N/D	25.223	N/D	N/D	N/D	N/D	N/D	N/D	N/D
20.11.2013	27.11.2013	N/D	N/D	N/D	N/D	N/D	N/D	14.913	N/D	N/D	N/D	N/D	N/D	N/D	N/D
4.12.2013	11.12.2013	N/D	N/D	N/D	N/D	N/D	N/D	2.946	N/D	N/D	N/D	N/D	N/D	N/D	N/D
18.12.2013	25.12.2013	N/D	N/D	N/D	N/D	N/D	N/D	2.897	N/D	N/D	N/D	N/D	N/D	N/D	N/D

Table S12 (continued): Concentration of individual CUPs (in pg.m⁻³) in the gaseous phase at the background site.

Sampling start	Sampling end	Fonofos	Isoproturon	Malathion	Metamitron	Metazachlor	Metribuzin	Prochloraz	Pyrazon	Simazine	S-Metolachlor	Temephos	Terbufos	Terbutylazine
4.1.2012	11.1.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
18.1.2012	25.1.2012	N/D	N/D	N/D	7.401	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
1.2.2012	8.2.2012	N/D	0.039	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
15.2.2012	22.2.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ
29.2.2012	7.3.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
14.3.2012	21.3.2012	N/D	N/D	N/D	2.674	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.040
28.3.2012	4.4.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
11.4.2012	18.4.2012	N/D	<LOQ	N/D	N/D	0.111	N/D	N/D	N/D	N/D	1.598	N/D	N/D	0.190
25.4.2012	2.5.2012	N/D	0.641	N/D	N/D	0.074	N/D	N/D	N/D	N/D	309.479	N/D	N/D	17.641
9.5.2012	16.5.2012	N/D	0.046	N/D	N/D	0.060	5.462	N/D	N/D	N/D	160.401	N/D	N/D	22.208
23.5.2012	30.5.2012	N/D	0.074	N/D	N/D	0.261	1.137	N/D	N/D	N/D	27.739	N/D	N/D	10.310
6.6.2012	13.6.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	37.223	N/D	N/D	13.278
20.6.2012	27.6.2012	N/D	0.136	N/D	N/D	0.299	N/D	N/D	N/D	N/D	47.040	<LOQ	N/D	25.588
4.7.2012	11.7.2012	N/D	0.063	N/D	N/D	0.183	N/D	N/D	N/D	N/D	11.830	N/D	N/D	9.971
18.7.2012	25.7.2012	N/D	N/D	N/D	N/D	0.176	N/D	N/D	N/D	N/D	3.780	N/D	N/D	3.706
1.8.2012	8.8.2012	N/D	N/D	N/D	N/D	0.306	N/D	N/D	N/D	N/D	1.338	N/D	N/D	1.873
15.8.2012	22.8.2012	N/D	N/D	N/D	N/D	50.614	N/D	N/D	N/D	N/D	0.370	N/D	N/D	0.367
29.8.2012	5.9.2012	N/D	N/D	N/D	N/D	80.598	0.937	N/D	N/D	N/D	0.540	N/D	N/D	0.710
12.9.2012	19.9.2012	N/D	0.106	N/D	N/D	15.246	N/D	N/D	N/D	N/D	0.571	N/D	N/D	1.106
26.9.2012	3.10.2012	N/D	0.044	N/D	N/D	3.855	N/D	N/D	N/D	N/D	0.861	N/D	N/D	1.287
10.10.2012	17.10.2012	N/D	0.095	N/D	N/D	0.690	N/D	N/D	N/D	N/D	0.204	N/D	N/D	0.437
24.10.2012	31.10.2012	N/D	0.406	N/D	N/D	0.480	N/D	N/D	N/D	N/D	0.116	N/D	N/D	0.286
7.11.2012	14.11.2012	N/D	0.152	N/D	10.129	0.326	N/D	N/D	N/D	N/D	0.101	N/D	N/D	0.221

22.11.2012	28.11.2012	N/D	0.471	N/D	14.706	0.347	N/D	N/D	N/D	N/D	0.195	N/D	N/D	0.261
5.12.2012	12.12.2012	N/D	0.051	N/D	N/D	0.057	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
19.12.2012	26.12.2012	N/D	0.033	N/D	16.470	0.081	N/D	N/D	N/D	N/D	0.060	N/D	N/D	0.068
2.1.2013	9.1.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
16.1.2013	23.1.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ
30.1.2013	6.2.2013	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
13.2.2013	20.2.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
27.2.2013	6.3.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
13.3.2013	20.3.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
27.3.2013	3.4.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
10.4.2013	17.4.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ
24.4.2013	1.5.2013	N/D	122.401	N/D	N/D	1.789	N/D	N/D	N/D	N/D	9.969	N/D	N/D	4.860
8.5.2013	15.5.2013	N/D	7.684	N/D	N/D	0.764	N/D	N/D	N/D	N/D	69.358	N/D	N/D	15.345
22.5.2013	29.5.2013	N/D	0.256	N/D	N/D	0.132	N/D	N/D	N/D	N/D	196.914	N/D	N/D	14.570
5.6.2013	12.6.2013	N/D	0.313	N/D	N/D	<LOQ	0.738	N/D	N/D	N/D	5.138	N/D	N/D	1.482
19.6.2013	26.6.2013	N/D	1.616	N/D	N/D	0.272	N/D	N/D	N/D	N/D	20.498	N/D	N/D	33.821
3.7.2013	10.7.2013	N/D	0.447	N/D	N/D	N/D	N/D	N/D	N/D	N/D	22.346	N/D	N/D	10.232
17.7.2013	24.7.2013	N/D	0.159	0.298	N/D	N/D	N/D	N/D	N/D	N/D	1.357	N/D	N/D	3.509
31.7.2013	7.8.2013	N/D	N/D	N/D	N/D	0.696	N/D	N/D	N/D	N/D	0.145	N/D	N/D	N/D
14.8.2013	21.8.2013	N/D	N/D	N/D	7.178	0.552	N/D	N/D	N/D	N/D	0.378	N/D	N/D	0.191
28.8.2013	4.9.2013	N/D	0.061	N/D	1.845	262.115	N/D	N/D	0.801	N/D	1.386	N/D	N/D	0.706
11.9.2013	18.9.2013	N/D	N/D	N/D	0.238	28.069	N/D	N/D	N/D	N/D	0.331	N/D	N/D	0.247
25.9.2013	2.10.2013	N/D	0.056	N/D	1.028	25.424	N/D	N/D	N/D	N/D	0.531	N/D	N/D	0.259
9.10.2013	16.10.2013	N/D	0.475	N/D	1.072	12.096	N/D	N/D	N/D	N/D	0.449	N/D	N/D	0.235
23.10.2013	30.10.2013	N/D	0.309	N/D	N/D	7.480	0.691	N/D	N/D	N/D	0.307	N/D	N/D	0.402
6.11.2013	13.11.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.207	N/D	N/D
20.11.2013	27.11.2013	N/D	N/D	N/D	N/D	0.404	0.240	N/D	N/D	N/D	0.130	N/D	N/D	0.022
4.12.2013	11.12.2013	N/D	N/D	N/D	N/D	N/D	1.199	N/D	N/D	N/D	N/D	N/D	N/D	N/D
18.12.2013	25.12.2013	N/D	N/D	N/D	N/D	0.467	N/D	N/D	N/D	N/D	0.061	N/D	N/D	0.022

Table S13: Concentration of individual CUPs (in pg.m^{-3}) in the particulate phase at the background site. N/D indicates compounds that were not detected and <LOQ indicates samples that were below the limit of quantification (in Table S7).

Sampling start	Sampling end	Acetochlor	Alachlor	Atrazine	Azinphos methyl	Carbendazim	Chlorotoluron	Chlorpyrifos	Diazinon	Dimethachlor	Dimethoate	Disulfoton	Diuron	Fenitrothion	Fenpropimorph
4.1.2012	11.1.2012	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	N/D	N/D	N/D	N/D
18.1.2012	25.1.2012	N/D	N/D	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
1.2.2012	8.2.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
15.2.2012	22.2.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
29.2.2012	7.3.2012	0.532	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
14.3.2012	21.3.2012	0.566	N/D	N/D	N/D	N/D	0.368	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
28.3.2012	4.4.2012	N/D	N/D	N/D	N/D	N/D	0.505	9.428	N/D	N/D	N/D	N/D	N/D	N/D	0.160
11.4.2012	18.4.2012	N/D	N/D	N/D	N/D	N/D	1.411	3.301	N/D	N/D	N/D	N/D	N/D	N/D	0.171
25.4.2012	2.5.2012	1.712	0.442	N/D	N/D	1.974	3.693	0.451	N/D	N/D	N/D	N/D	N/D	N/D	0.745
9.5.2012	16.5.2012	23.247	0.819	N/D	N/D	2.618	0.655	0.280	N/D	N/D	N/D	N/D	N/D	N/D	73.802
23.5.2012	30.5.2012	0.528	0.279	0.487	N/D	1.983	0.135	0.117	N/D	N/D	N/D	N/D	N/D	N/D	10.129
6.6.2012	13.6.2012	0.577	N/D	N/D	N/D	0.252	N/D	0.760	N/D	N/D	N/D	N/D	N/D	N/D	5.561
20.6.2012	27.6.2012	N/D	N/D	N/D	N/D	N/D	N/D	0.267	N/D	N/D	0.027	N/D	N/D	N/D	0.392
4.7.2012	11.7.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.156	N/D	1.168
18.7.2012	25.7.2012	N/D	N/D	N/D	N/D	0.355	N/D	0.250	N/D	N/D	N/D	N/D	0.265	N/D	0.467
1.8.2012	8.8.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.240	N/D	0.206
15.8.2012	22.8.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	1.851	N/D	N/D	0.205	N/D	0.151
29.8.2012	5.9.2012	N/D	N/D	N/D	N/D	N/D	N/D	0.143	N/D	0.519	N/D	N/D	N/D	N/D	1.506
12.9.2012	19.9.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.136	N/D	0.026
26.9.2012	3.10.2012	N/D	N/D	N/D	N/D	N/D	2.434	0.166	N/D	N/D	N/D	N/D	0.211	N/D	0.013
10.10.2012	17.10.2012	N/D	N/D	N/D	N/D	N/D	7.681	0.553	N/D	N/D	N/D	N/D	0.098	N/D	N/D
24.10.2012	31.10.2012	N/D	N/D	N/D	N/D	N/D	12.900	0.631	N/D	N/D	N/D	N/D	0.351	N/D	N/D
7.11.2012	14.11.2012	N/D	N/D	N/D	N/D	N/D	2.728	0.398	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ
22.11.2012	28.11.2012	N/D	N/D	N/D	N/D	N/D	0.802	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
5.12.2012	12.12.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
19.12.2012	26.12.2012	N/D	N/D	N/D	N/D	N/D	0.185	N/D	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ

2.1.2013	9.1.2013	N/D	N/D	N/D	N/D	N/D	N/D	0.121	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ
16.1.2013	23.1.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
30.1.2013	6.2.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	<LOQ
13.2.2013	20.2.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
27.2.2013	6.3.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
13.3.2013	20.3.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	1.232	N/D	<LOQ
27.3.2013	3.4.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
10.4.2013	17.4.2013	1.155	N/D	N/D	N/D	N/D	2.675	0.142	N/D	N/D	N/D	N/D	0.353	N/D	N/D
24.4.2013	1.5.2013	11.854	N/D	N/D	N/D	N/D	6.063	1.512	N/D	N/D	N/D	N/D	N/D	N/D	0.544
8.5.2013	15.5.2013	5.123	N/D	N/D	N/D	1.281	0.324	0.231	N/D	N/D	N/D	N/D	N/D	N/D	4.253
22.5.2013	29.5.2013	2.810	N/D	N/D	N/D	1.353	0.428	0.166	N/D	N/D	N/D	N/D	N/D	N/D	29.631
5.6.2013	12.6.2013	N/D	N/D	N/D	N/D	5.937	0.136	N/D	N/D	N/D	N/D	N/D	N/D	N/D	48.356
19.6.2013	26.6.2013	N/D	N/D	N/D	N/D	1.529	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	5.386
3.7.2013	10.7.2013	N/D	N/D	N/D	N/D	5.965	N/D	0.117	N/D	N/D	N/D	N/D	0.080	N/D	19.138
17.7.2013	24.7.2013	N/D	N/D	0.063	N/D	8.289	<LOQ	0.416	<LOQ	N/D	N/D	N/D	0.145	N/D	9.612
31.7.2013	7.8.2013	N/D	N/D	0.247	N/D	2.161	N/D	0.423	N/D	0.485	N/D	N/D	0.746	N/D	3.217
14.8.2013	21.8.2013	N/D	N/D	N/D	N/D	1.286	N/D	<LOQ	N/D	9.357	N/D	N/D	0.117	N/D	0.936
28.8.2013	4.9.2013	N/D	N/D	0.082	N/D	2.528	N/D	N/D	N/D	1.290	N/D	N/D	<LOQ	N/D	15.632
11.9.2013	18.9.2013	N/D	N/D	N/D	N/D	5.172	N/D	N/D	N/D	0.611	0.081	N/D	<LOQ	N/D	1.118
25.9.2013	2.10.2013	N/D	N/D	0.097	N/D	12.525	2.114	N/D	N/D	0.230	N/D	N/D	0.074	N/D	0.469
9.10.2013	16.10.2013	N/D	N/D	N/D	N/D	11.419	2.513	1.646	N/D	N/D	N/D	N/D	N/D	N/D	1.291
23.10.2013	30.10.2013	N/D	N/D	N/D	N/D	8.242	24.665	0.112	<LOQ	N/D	N/D	2.216	0.098	N/D	0.922
6.11.2013	13.11.2013	N/D	N/D	0.140	N/D	7.554	5.739	0.131	N/D	0.335	N/D	N/D	0.072	N/D	0.358
20.11.2013	27.11.2013	N/D	N/D	0.041	N/D	8.460	1.106	N/D	N/D	0.123	N/D	N/D	<LOQ	N/D	0.279
4.12.2013	11.12.2013	N/D	N/D	0.038	N/D	1.388	0.252	0.156	N/D	<LOQ	N/D	N/D	N/D	N/D	0.542
18.12.2013	25.12.2013	N/D	N/D	N/D	N/D	12.107	0.266	0.250	<LOQ	N/D	N/D	N/D	N/D	N/D	0.698

Table S13 (continued): Concentration of individual CUPs (in $\text{pg}\cdot\text{m}^{-3}$) in the particulate phase at the background site.

Sampling start	Sampling end	Fonofos	Isoproturon	Malathion	Metamitron	Metazachlor	Metribuzin	Prochloraz	Pyrazon	Simazine	S-Metolachlor	Temephos	Terbufos	Terbutylazine
4.1.2012	11.1.2012	N/D	0.213	N/D	N/D	0.371	N/D	N/D	N/D	N/D	0.146	N/D	N/D	0.155
18.1.2012	25.1.2012	N/D	0.094	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
1.2.2012	8.2.2012	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
15.2.2012	22.2.2012	N/D	0.061	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
29.2.2012	7.3.2012	N/D	0.224	N/D	N/D	0.088	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
14.3.2012	21.3.2012	N/D	1.836	N/D	N/D	0.135	N/D	0.354	N/D	N/D	N/D	N/D	N/D	0.100
28.3.2012	4.4.2012	N/D	5.612	N/D	N/D	0.198	N/D	0.212	N/D	N/D	N/D	N/D	N/D	0.258
11.4.2012	18.4.2012	N/D	5.494	N/D	N/D	0.087	N/D	0.132	N/D	N/D	0.158	N/D	N/D	0.260
25.4.2012	2.5.2012	N/D	20.241	N/D	3.784	0.262	N/D	1.493	1.049	N/D	19.603	N/D	N/D	10.235
9.5.2012	16.5.2012	N/D	5.033	N/D	N/D	0.876	N/D	1.659	N/D	N/D	90.955	N/D	N/D	31.597
23.5.2012	30.5.2012	N/D	0.938	N/D	2.357	0.252	N/D	0.446	0.897	N/D	4.962	N/D	N/D	3.761
6.6.2012	13.6.2012	N/D	0.161	N/D	N/D	0.071	N/D	0.169	N/D	N/D	5.515	N/D	N/D	11.394
20.6.2012	27.6.2012	N/D	0.038	N/D	N/D	<LOQ	N/D	0.128	N/D	N/D	0.103	N/D	N/D	0.136
4.7.2012	11.7.2012	N/D	<LOQ	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.138	N/D	N/D	0.704
18.7.2012	25.7.2012	N/D	<LOQ	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.156	N/D	N/D	0.483
1.8.2012	8.8.2012	N/D	N/D	N/D	N/D	0.919	N/D	N/D	N/D	N/D	0.157	N/D	N/D	0.206
15.8.2012	22.8.2012	N/D	<LOQ	N/D	N/D	30.166	N/D	0.055	N/D	N/D	<LOQ	N/D	N/D	0.176
29.8.2012	5.9.2012	N/D	N/D	N/D	N/D	6.894	N/D	0.098	N/D	N/D	N/D	N/D	N/D	0.245
12.9.2012	19.9.2012	N/D	0.051	N/D	N/D	2.159	N/D	0.136	N/D	N/D	N/D	N/D	N/D	0.138
26.9.2012	3.10.2012	N/D	11.564	N/D	N/D	3.787	N/D	0.809	N/D	N/D	0.092	N/D	N/D	0.205
10.10.2012	17.10.2012	N/D	27.769	N/D	N/D	0.634	N/D	0.282	N/D	N/D	N/D	N/D	N/D	0.212
24.10.2012	31.10.2012	N/D	56.175	N/D	N/D	0.302	N/D	0.104	N/D	N/D	N/D	N/D	N/D	0.000
7.11.2012	14.11.2012	N/D	2.817	N/D	N/D	0.140	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.190

22.11.2012	28.11.2012	N/D	4.183	N/D	N/D	0.271	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
5.12.2012	12.12.2012	N/D	0.226	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
19.12.2012	26.12.2012	N/D	0.410	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
2.1.2013	9.1.2013	N/D	0.323	N/D	N/D	0.306	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.112
16.1.2013	23.1.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
30.1.2013	6.2.2013	N/D	0.156	N/D	N/D	0.368	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
13.2.2013	20.2.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
27.2.2013	6.3.2013	N/D	0.071	N/D	N/D	0.158	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
13.3.2013	20.3.2013	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
27.3.2013	3.4.2013	N/D	0.048	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D
10.4.2013	17.4.2013	N/D	4.653	N/D	N/D	0.888	N/D	0.147	N/D	N/D	0.877	N/D	N/D	0.490
24.4.2013	1.5.2013	N/D	290.673	N/D	N/D	0.806	N/D	1.637	N/D	N/D	3.520	N/D	N/D	6.813
8.5.2013	15.5.2013	N/D	13.804	N/D	6.414	0.069	N/D	0.258	0.415	N/D	1.927	N/D	N/D	5.734
22.5.2013	29.5.2013	N/D	8.860	N/D	N/D	0.258	N/D	0.847	N/D	N/D	41.694	N/D	N/D	28.551
5.6.2013	12.6.2013	N/D	1.206	N/D	N/D	0.169	N/D	1.948	N/D	N/D	5.161	N/D	N/D	22.492
19.6.2013	26.6.2013	N/D	0.233	N/D	N/D	N/D	N/D	0.119	1.090	N/D	0.132	N/D	N/D	0.526
3.7.2013	10.7.2013	N/D	0.265	N/D	N/D	0.158	N/D	0.872	N/D	N/D	0.482	N/D	N/D	2.288
17.7.2013	24.7.2013	N/D	0.209	N/D	N/D	0.154	N/D	0.355	2.254	0.087	0.198	N/D	N/D	0.750
31.7.2013	7.8.2013	8.028	1.305	N/D	N/D	0.332	N/D	0.251	1.023	N/D	2.659	N/D	N/D	7.352
14.8.2013	21.8.2013	1.081	0.420	N/D	N/D	275.496	N/D	0.045	0.371	N/D	1.158	N/D	N/D	1.198
28.8.2013	4.9.2013	0.650	0.163	N/D	N/D	82.106	N/D	N/D	0.280	N/D	<LOQ	N/D	N/D	0.165
11.9.2013	18.9.2013	N/D	0.259	N/D	N/D	123.587	N/D	0.264	N/D	N/D	0.295	N/D	N/D	0.254
25.9.2013	2.10.2013	N/D	2.451	<LOQ	N/D	13.128	N/D	1.298	N/D	N/D	<LOQ	N/D	N/D	0.101
9.10.2013	16.10.2013	N/D	5.894	N/D	N/D	5.457	1.825	1.131	N/D	N/D	<LOQ	0.076	N/D	0.074
23.10.2013	30.10.2013	N/D	33.710	0.132	N/D	30.236	1.795	0.126	N/D	N/D	0.171	0.105	N/D	0.259
6.11.2013	13.11.2013	N/D	3.862	N/D	N/D	7.474	N/D	N/D	N/D	N/D	0.253	N/D	N/D	0.335
20.11.2013	27.11.2013	N/D	1.529	N/D	N/D	1.618	N/D	N/D	N/D	N/D	0.070	N/D	0.614	0.036
4.12.2013	11.12.2013	N/D	0.226	N/D	N/D	0.567	N/D	0.030	N/D	N/D	0.061	N/D	N/D	0.027
18.12.2013	25.12.2013	N/D	0.279	N/D	N/D	0.977	N/D	N/D	N/D	N/D	<LOQ	N/D	N/D	0.049

Table S14: Concentrations of individual OCPs (in $\text{pg}\cdot\text{m}^{-3}$) in the particulate phase. <LOQ indicates samples that were below the limit of quantification (in Table S7).

Sampling time	α -HCH	β -HCH	γ -HCH	δ -HCH	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>o,p'</i> -DDD	<i>p,p'</i> -DDD	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT
11-18/01/12	<LOQ	0.011	<LOQ	0.008	0.007	0.169	0.068	0.096	0.040	0.149
25/01-01/02/12	0.031	0.030	0.025	0.029	0.051	0.900	0.109	0.399	0.099	0.288
08-15/02/12	0.012	0.018	0.018	0.020	0.054	0.526	0.076	0.345	0.114	0.462
22-29/02/12	<LOQ	0.020	0.000	0.027	0.007	0.209	0.047	0.098	0.037	0.106
07-14/03/12	0.022	0.020	0.042	0.025	0.011	0.388	0.052	0.141	0.074	0.189
21-28/03/12	0.027	0.020	0.043	0.064	0.016	0.583	0.063	0.250	0.113	0.317
04-11/04/12	0.007	0.012	0.013	0.065	0.014	0.306	0.035	0.123	0.051	0.143
18-25/04/12	<LOQ	0.008	<LOQ	0.000	0.004	0.190	0.022	0.070	0.034	0.098
02-09/05/12	0.016	0.074	0.019	0.035	0.002	0.133	0.029	0.041	0.022	0.052
16-23/05/12	0.005	0.005	0.014	0.008	0.004	0.086	0.015	0.031	0.021	0.049
30/05-06/06/12	0.016	0.011	<LOQ	0.004	0.002	0.067	0.023	0.021	0.022	0.044
13-20/06/12	0.011	<LOQ	0.014	<LOQ	<LOQ	0.057	0.005	0.006	0.011	0.042
27/06-04/07/12	0.008	<LOQ	0.014	<LOQ	<LOQ	0.048	<LOQ	0.006	0.010	0.036
11-18/07/12	0.004	<LOQ	0.010	<LOQ	<LOQ	0.042	<LOQ	0.005	0.009	0.040
25/07-01/08/12	0.003	<LOQ	0.007	<LOQ	<LOQ	0.060	<LOQ	0.004	0.006	0.023
08-15/08/12	0.003	<LOQ	0.006	<LOQ	<LOQ	0.057	<LOQ	0.007	0.014	0.048
22-29/08/12	0.003	<LOQ	0.005	<LOQ	<LOQ	0.035	<LOQ	0.004	0.005	0.018
05-12/09/12	0.004	<LOQ	0.010	<LOQ	<LOQ	0.079	<LOQ	0.007	0.015	0.046
19-26/09/12	0.004	<LOQ	0.010	<LOQ	<LOQ	0.158	<LOQ	0.012	0.027	0.100
03-10/10/12	0.004	<LOQ	0.011	<LOQ	<LOQ	0.271	<LOQ	0.027	0.054	0.204
17-24/10/12	0.009	0.007	0.016	<LOQ	<LOQ	0.397	<LOQ	0.034	0.064	0.238
31/11-07/11/12	0.006	0.006	0.010	<LOQ	<LOQ	0.273	<LOQ	0.026	0.044	0.191
14-21/11/12	0.008	0.014	0.013	<LOQ	<LOQ	0.635	<LOQ	0.044	0.097	0.356
28/11-05/12/12	0.006	0.004	0.016	<LOQ	<LOQ	0.501	0.014	0.076	0.093	0.419
12-19/12/12	0.010	<LOQ	0.024	<LOQ	<LOQ	0.466	<LOQ	0.076	0.102	0.390
26/12/12-02/01/13	0.005	<LOQ	0.018	<LOQ	<LOQ	0.437	0.012	0.057	0.083	0.353
09-16/01/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.410	<LOQ	<LOQ	<LOQ	0.303
23-30/01/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.499	<LOQ	<LOQ	<LOQ	0.504
06-13/02/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.472	<LOQ	<LOQ	<LOQ	0.339
20-27/02/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.656	<LOQ	<LOQ	<LOQ	0.348
06-13/03/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.963	<LOQ	<LOQ	<LOQ	0.444
20-27/03/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.370	<LOQ	<LOQ	<LOQ	0.298
03-10/04/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.330	<LOQ	<LOQ	<LOQ	<LOQ
17-24/04/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.243	<LOQ	<LOQ	<LOQ	0.169
01-08/05/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.162	<LOQ	<LOQ	<LOQ	<LOQ
15-22/05/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.111	<LOQ	<LOQ	<LOQ	<LOQ
29/05-05/06/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.072	<LOQ	<LOQ	<LOQ	<LOQ
12-19/06/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.066	<LOQ	<LOQ	<LOQ	<LOQ
26/06-03/07/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
10-17/07/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
24-31/07/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
07-14/08/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
21-26/08/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.148	<LOQ	<LOQ	<LOQ	<LOQ
04-11/09/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
18-25/09/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.153	<LOQ	<LOQ	<LOQ	<LOQ
02-09/10/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.547	<LOQ	<LOQ	<LOQ	<LOQ
16-23/10/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.220	<LOQ	<LOQ	<LOQ	<LOQ
30/10-06/11/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
13-20/11/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.944	<LOQ	<LOQ	<LOQ	<LOQ
27/11-04/12/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.551	<LOQ	<LOQ	<LOQ	<LOQ
11-18/12/13	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.922	<LOQ	<LOQ	<LOQ	<LOQ
25/12/13-01/01/14	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.658	<LOQ	<LOQ	<LOQ	<LOQ

Table S15: Concentrations of individual OCPs (in pg.m⁻³) in the gaseous phase. <LOQ indicates samples that were below the limit of quantification (in Table S7).

Sampling time	α -HCH	β -HCH	γ -HCH	δ -HCH	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>o,p'</i> -DDD	<i>p,p'</i> -DDD	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT
11-18/01/12	1.419	0.191	1.279	0.133	0.141	5.282	0.169	0.250	0.354	0.265
25/01-01/02/12	2.028	0.190	1.136	0.095	0.130	2.812	0.066	0.141	0.258	0.281
08-15/02/12	1.078	0.150	0.470	0.098	0.073	0.612	0.024	0.118	0.119	0.130
22-29/02/12	2.352	0.206	3.663	0.167	0.272	10.366	0.355	0.548	0.761	0.537
07-14/03/12	1.684	0.198	2.208	0.108	0.292	11.552	0.405	0.665	0.818	0.578
21-28/03/12	1.759	0.189	3.357	0.123	0.479	19.750	0.623	1.081	1.619	1.399
04-11/04/12	2.480	0.159	3.180	0.164	0.352	14.747	0.414	0.688	1.053	0.905
18-25/04/12	1.740	0.098	2.518	0.103	0.267	12.826	0.361	0.612	1.065	0.936
02-09/05/12	1.788	0.256	7.522	0.220	0.737	33.746	1.276	2.565	3.463	3.307
16-23/05/12	3.145	0.255	5.562	0.100	0.348	12.285	0.444	0.708	1.416	1.180
30/05-06/06/12	1.832	0.182	7.994	0.169	0.445	19.230	0.657	1.070	2.547	2.307
13-20/06/12	2.574	0.343	9.468	0.260	0.647	27.765	0.299	0.442	4.558	5.556
27/06-04/07/12	2.243	0.461	8.816	0.420	0.739	31.242	0.326	0.453	5.041	6.598
11-18/07/12	3.004	0.182	10.049	0.185	0.245	11.689	0.118	0.187	1.606	1.942
25/07-01/08/12	2.149	0.462	9.214	0.338	0.617	29.743	0.324	0.419	5.674	7.025
08-15/08/12	2.235	0.210	5.112	0.180	0.378	16.873	0.169	0.254	2.381	2.802
22-29/08/12	1.886	0.226	7.798	0.249	0.453	25.550	0.207	0.313	3.208	4.299
05-12/09/12	1.392	0.243	5.456	0.179	0.682	37.298	0.343	0.524	4.757	5.910
19-26/09/12	3.331	0.150	6.585	0.175	0.667	40.370	0.260	0.339	4.236	4.771
03-10/10/12	4.161	0.194	8.211	0.179	0.566	36.428	0.243	0.363	3.429	4.245
17-24/10/12	6.090	0.190	7.632	0.263	0.649	32.436	0.178	0.250	2.992	3.366
31/11-07/11/12	4.103	0.092	2.990	0.103	0.314	15.834	0.095	0.154	1.451	1.621
14-21/11/12	9.779	0.322	8.351	0.214	0.748	27.029	0.159	0.244	2.342	2.330
28/11-05/12/12	4.185	0.103	3.706	0.105	0.385	15.392	0.107	0.164	1.394	1.324
12-19/12/12	3.040	0.069	2.957	0.086	0.210	5.647	0.085	0.142	0.578	0.552
26/12/12-02/01/13	3.918	0.085	4.316	0.108	0.260	10.985	0.104	0.161	0.980	0.920
09-16/01/13	3.575	<LOQ	3.177	0.315	0.168	5.729	<LOQ	<LOQ	<LOQ	0.329
23-30/01/13	2.291	<LOQ	2.031	<LOQ	0.223	2.925	<LOQ	<LOQ	<LOQ	0.485
06-13/02/13	1.767	<LOQ	1.451	<LOQ	0.141	4.238	<LOQ	<LOQ	<LOQ	0.261
20-27/02/13	3.686	<LOQ	1.784	<LOQ	0.402	6.506	<LOQ	<LOQ	<LOQ	0.563
06-13/03/13	4.203	<LOQ	4.277	<LOQ	0.576	19.574	<LOQ	<LOQ	1.543	1.383
20-27/03/13	2.297	<LOQ	1.192	<LOQ	0.152	4.132	<LOQ	<LOQ	0.318	0.299
03-10/04/13	3.849	<LOQ	2.206	<LOQ	0.359	10.195	0.114	<LOQ	0.871	0.899
17-24/04/13	3.253	0.158	7.594	0.205	0.704	29.800	0.249	<LOQ	2.845	3.220
01-08/05/13	3.440	0.218	6.218	0.165	0.833	36.576	0.294	0.473	3.355	3.484
15-22/05/13	1.658	0.126	5.701	<LOQ	0.397	19.756	0.154	0.251	1.681	1.857
29/05-05/06/13	4.692	0.126	6.712	<LOQ	0.434	18.004	0.204	0.253	1.959	1.751
12-19/06/13	1.477	0.264	4.797	0.267	0.626	25.908	0.311	0.397	3.409	3.777
26/06-03/07/13	4.100	<LOQ	5.223	<LOQ	0.303	15.104	<LOQ	0.248	1.383	1.498
10-17/07/13	1.776	0.336	6.741	<LOQ	<LOQ	16.669	0.272	0.354	3.637	4.033
24-31/07/13	2.206	0.592	9.186	<LOQ	0.950	38.013	0.488	0.441	6.696	8.037
07-14/08/13	7.074	<LOQ	21.775	<LOQ	1.420	71.437	<LOQ	0.892	9.177	9.985
21-26/08/13	3.556	0.293	6.445	<LOQ	0.977	49.387	0.394	0.322	5.705	5.813
04-11/09/13	4.306	<LOQ	8.588	<LOQ	0.724	40.787	0.281	0.253	4.137	4.279
18-25/09/13	3.067	<LOQ	7.702	<LOQ	0.576	34.714	0.305	0.270	3.515	3.912
02-09/10/13	3.539	<LOQ	3.285	<LOQ	0.391	15.309	<LOQ	<LOQ	1.386	1.435
16-23/10/13	2.951	0.229	6.366	<LOQ	<LOQ	31.749	0.202	0.178	2.962	2.892
30/10-06/11/13	3.031	<LOQ	4.464	<LOQ	0.280	14.546	<LOQ	<LOQ	1.216	1.014
13-20/11/13	4.845	0.216	4.077	<LOQ	0.517	19.105	<LOQ	0.188	1.543	1.365
27/11-04/12/13	3.052	<LOQ	2.299	<LOQ	0.238	8.752	<LOQ	<LOQ	0.573	0.506
11-18/12/13	3.675	<LOQ	3.161	<LOQ	0.314	12.571	<LOQ	<LOQ	0.692	0.461
25/12/13-01/01/14	3.604	<LOQ	4.193	<LOQ	0.376	17.425	<LOQ	<LOQ	0.784	0.490

Table S16: Results of Clausius-Clapeyron plots showing slopes (m), constants (b) and coefficient of determination (r^2) for OCPs. Compounds in bold are those where the regression parameters suggest local revolatilization as the main control on gas-phase air concentrations.

	N	m	b	r^2	p
α -HCH	52	-141.6	24.00	<0.01	<0.819
β-HCH	36	-3148	16.03	0.40	<0.01
γ -HCH	52	-6097	-2.373	0.73	<0.01
δ -HCH	30	-2792	-17.42	0.43	<0.01
<i>o,p'</i> -DDE	50	-4797	-9.484	0.62	<0.01
<i>p,p'</i> -DDE	52	-7225	2.823	0.68	<0.01
<i>o,p'</i> -DDD	38	-5060	-9.217	0.44	<0.01
<i>p,p'</i> -DDD	39	-3353	-0.2346	0.23	<0.01
<i>o,p'</i> -DDT	48	-8729	5.774	0.81	<0.01
<i>p,p'</i> -DDT	52	-9802	9.570	0.84	<0.01

Table S17: Results of Clausius-Clapeyron plots showing slopes (m), constants (b) and coefficient of determination (r^2) for CUPs. Compounds in bold are those where regression parameters suggest local revolatilization as the main control on gas-phase air concentrations.

	N	m	b	r^2	p
Metazachlor	31	-6855	1.381	0.06	<0.182
Isoproturon	25	-5313	-7.936	0.09	<0.154
Chlorpyrifos	44	-4922	-6.857	0.06	<0.109
Terbutylazine	34	-20647	47.26	0.48	<0.01
S-Metolachlor	34	-15791	30.44	0.22	<0.01
Acetochlor	23	4408	-39.56	0.01	<0.632
Dimethachlor	18	-15721	30.19	0.19	<0.075

Table S18: Octanol-air partitioning coefficients, K_{OA} , of OCPs and CUPs at 25°C, T_{min} (-10.3°C for OCPs and -13.3°C for CUPs) and T_{max} (22.7°C for OCPs and 22.8°C for CUPs). Extrapolation: see text.

Compound	$\log K_{OA}(T_{ref})$	Reference	Temperature-corrected $\log K_{OA}(T)$		
			T_{min}	T_{max}	Reference
Acetochlor	9.07	(Coscollà et al., 2013b)	11.2	9.16	Extrapolated
Alachlor	9.98	(Coscollà et al., 2013b)	12.3	10.1	Extrapolated
Atrazine	9.62	(Götz et al., 2007)	11.9	9.72	Extrapolated
Azinphos Methyl	8.76	(Coscollà et al., 2013a)	10.8	8.85	Extrapolated
Carbendazim	10.6	(Coscollà et al., 2013a)	13.1	10.7	Extrapolated
Chlorotoluron	10.6	(Götz et al., 2007)	13.1	10.7	Extrapolated
Chlorpyrifos	8.41	(Odabasi and Cetin, 2012b)	10.3	8.50	(Odabasi and Cetin, 2012b)
Diazinon	9.14	(Coscollà et al., 2013b)	11.3	9.24	Extrapolated
Dimethachlor	9.34	(U.S. EPA, 2012)	11.5	9.44	Extrapolated
Dimethoate	9.15	(Coscollà et al., 2013a)	11.3	9.25	Extrapolated
Disulfoton	8.07	(U.S. EPA, 2012)	10.0	8.16	Extrapolated
Diuron	10.4	(Coscollà et al., 2013a)	12.8	10.5	Extrapolated
Fenitrothion	7.72	(U.S. EPA, 2012)	9.58	7.80	Extrapolated
Fenpropimorph	8.93	(Coscollà et al., 2013b)	11.0	9.02	Extrapolated
Fonofos	7.48	(U.S. EPA, 2012)	9.30	7.56	Extrapolated
Isoproturon	11.2	(Götz et al., 2007)	13.8	11.3	Extrapolated
Malathion	9.06	(Coscollà et al., 2013b)	11.2	9.15	Extrapolated
Metamitron	11.2	(U.S. EPA, 2012)	13.8	11.4	Extrapolated
Metazachlor	9.76	(Coscollà et al., 2013b)	12.0	9.86	Extrapolated
Metribuzin	10.0	(U.S. EPA, 2012)	12.3	10.1	Extrapolated
Prochloraz	13.6	(U.S. EPA, 2012)	16.6	13.7	Extrapolated
Pyrazon	9.01	(U.S. EPA, 2012)	11.1	9.10	Extrapolated
Simazine	9.59	(Götz et al., 2007)	11.8	9.69	Extrapolated
S-Metolachlor	9.33	(U.S. EPA, 2012)	11.5	9.43	Extrapolated
Temephos	13.1	(U.S. EPA, 2012)	16.0	13.2	Extrapolated
Terbufos	7.49	(U.S. EPA, 2012)	9.30	7.57	Extrapolated
Terbutylazine	9.03	(Coscollà et al., 2013a)	11.2	9.12	Extrapolated
α -HCH	7.61	(Shoeib and Harner, 2002b)	9.06	7.69	(Shoeib and Harner, 2002b)
β -HCH	8.88	(Shoeib and Harner, 2002b)	11.1	9.00	(Shoeib and Harner, 2002b)
γ -HCH	7.85	(Shoeib and Harner, 2002b)	9.38	7.93	(Shoeib and Harner, 2002b)
δ -HCH	8.84	(Shoeib and Harner, 2002b)	11.0	8.97	(Shoeib and Harner, 2002b)
<i>o,p'</i> -DDE	9.26	(Zhang et al., 2009)	10.9	9.12	(Zhang et al., 2009)
<i>p,p'</i> -DDE	9.68	(Shoeib and Harner, 2002b)	12.0	9.80	(Shoeib and Harner, 2002b)
<i>o,p'</i> -DDD	9.57	(Zhang et al., 2009)	11.3	9.42	(Zhang et al., 2009)
<i>p,p'</i> -DDD	10.1	(Shoeib and Harner, 2002b)	12.0	10.2	(Shoeib and Harner, 2002b)
<i>o,p'</i> -DDT	9.45	(Shoeib and Harner, 2002b)	11.5	9.57	(Shoeib and Harner, 2002b)
<i>p,p'</i> -DDT	9.82	(Shoeib and Harner, 2002b)	11.9	9.93	(Shoeib and Harner, 2002b)

Table S19: Particulate concentrations of individual OCPs (in $\text{pg}\cdot\text{m}^{-3}$) according to particle size (in μm) at the rural and the urban sites. N/D indicates that a compound was not detected. Numbers in bolds represent the particle size fraction with the highest concentration.

	Size fraction (μm)	α -HCH	β -HCH	γ -HCH	δ -HCH	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>o,p'</i> -DDD	<i>p,p'</i> -DDD	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	ΣOCPs	
Rural site	Winter	7.2-10	0.01	0.05	0.17	0.04	0.05	0.67	0.03	0.19	0.31	1.47	2.99
		3.0-7.2	0.16	0.02	0.33	0.08	0.05	0.91	0.06	0.37	0.25	1.38	3.61
		1.5-3.0	0.18	0.13	0.33	0.04	0.08	1.01	0.06	0.43	0.24	1.51	4.01
		0.95-1.5	0.09	0.09	0.23	0.10	0.17	2.71	0.27	1.73	0.48	3.29	9.16
		0.49-0.95	N/D	0.06	0.17	0.07	0.24	4.00	0.32	2.24	0.55	4.04	11.69
		<0.49	0.44	0.18	0.56	0.36	0.18	3.67	0.27	2.04	1.02	9.88	18.6
	Spring	7.2-10	0.22	0.04	0.49	0.24	0.05	1.25	0.04	0.11	0.20	0.54	3.18
		3.0-7.2	0.11	N/D	0.37	0.16	0.02	0.92	0.02	0.12	0.14	0.48	2.34
		1.5-3.0	0.19	N/D	0.48	0.19	0.03	1.17	0.03	0.15	0.16	0.69	3.09
		0.95-1.5	0.33	0.02	0.68	0.24	0.04	1.65	0.09	0.25	0.30	1.71	5.31
		0.49-0.95	0.05	0.06	0.26	0.03	0.05	2.43	0.08	0.46	0.41	2.59	6.42
		<0.49	0.04	1.27	0.32	0.17	0.06	1.94	0.16	0.23	0.69	4.69	9.57
	Summer	7.2-10	0.05	N/D	0.25	N/D	0.03	1.17	0.13	0.42	0.06	N/D	2.11
		3.0-7.2	0.07	N/D	0.26	N/D	0.03	1.01	0.13	0.50	0.06	0.09	2.15
		1.5-3.0	0.26	0.01	0.09	N/D	0.02	0.73	0.08	0.37	N/D	N/D	1.56
		0.95-1.5	0.10	N/D	0.15	0.05	0.01	0.65	0.08	0.35	N/D	0.02	1.41
		0.49-0.95	0.22	0.14	0.22	0.10	0.02	1.11	0.13	0.49	0.04	0.23	2.7
		<0.49	0.09	0.40	0.17	0.06	0.07	2.26	0.15	0.77	0.15	0.35	4.47
	Autumn	7.2-10	0.01	0.03	0.04	0.00	0.01	0.65	0.07	0.26	0.00	N/D	1.07
		3.0-7.2	0.01	N/D	0.03	N/D	0.03	1.57	0.13	0.70	0.01	N/D	2.48
		1.5-3.0	N/D	0.20	0.06	0.04	0.04	2.18	0.18	1.03	N/D	0.00	3.73
		0.95-1.5	0.13	0.16	0.15	0.11	0.10	5.31	0.41	2.63	0.07	0.18	9.25
		0.49-0.95	0.02	0.12	0.06	0.04	0.13	8.17	0.60	4.21	0.18	0.52	14.05
		<0.49	0.05	0.09	0.03	N/D	0.10	6.11	0.47	3.53	0.35	1.39	12.12
Urban site	Winter	7.2-10	N/D	0.89	0.11	N/D	0.07	0.64	0.13	0.40	0.03	N/D	2.27
		3.0-7.2	N/D	0.53	0.07	N/D	0.08	0.75	0.15	0.39	0.05	0.08	2.10
		1.5-3.0	N/D	0.24	0.12	N/D	0.12	1.12	0.19	0.58	0.06	0.11	2.54
		0.95-1.5	N/D	0.20	0.11	0.02	0.16	1.75	0.29	1.02	0.13	0.13	3.81
		0.49-0.95	0.01	0.12	0.12	N/D	0.19	1.92	0.35	1.37	0.08	0.15	4.31
		<0.49	0.22	0.17	0.33	0.18	0.19	2.29	0.49	1.85	0.41	0.40	6.53
	Spring	7.2-10	0.02	0.56	0.09	0.03	0.05	0.68	0.16	0.58	0.06	0.06	2.29
		3.0-7.2	0.10	0.75	0.15	0.06	0.07	0.93	0.20	0.71	0.06	0.07	3.10
		1.5-3.0	N/D	N/D	0.03	N/D	0.04	0.68	0.16	0.56	0.04	0.03	1.54
		0.95-1.5	N/D	0.08	0.03	N/D	0.05	0.77	0.18	0.65	0.05	0.06	1.87
		0.49-0.95	0.01	N/D	0.08	N/D	0.08	1.32	0.31	1.20	0.09	0.14	3.23
		<0.49	0.07	0.52	0.32	N/D	0.13	2.05	0.46	1.89	0.25	0.46	6.15
	Summer	7.2-10	N/D	0.48	0.04	N/D	0.05	0.68	0.17	0.57	0.07	0.06	2.12
		3.0-7.2	0.05	0.04	0.08	N/D	0.07	1.11	0.27	1.07	0.13	0.21	3.03
		1.5-3.0	0.14	0.16	0.11	N/D	0.05	0.69	0.18	0.66	0.12	0.25	2.36
		0.95-1.5	0.11	0.07	0.09	N/D	0.06	0.80	0.22	0.80	0.09	0.12	2.36
		0.49-0.95	0.23	0.98	0.26	0.14	0.07	0.94	0.24	0.99	0.13	0.25	4.23
		<0.49	0.19	0.41	0.38	0.23	0.14	1.95	0.43	1.56	0.30	0.39	5.98
	Autumn	7.2-10	0.02	0.49	0.17	0.04	0.10	2.07	0.28	1.26	0.17	0.33	4.93
		3.0-7.2	N/D	0.47	0.07	N/D	0.12	2.08	0.32	1.15	0.19	0.24	4.64
		1.5-3.0	N/D	0.35	0.07	N/D	0.10	1.81	0.33	1.24	0.12	0.28	4.30
		0.95-1.5	N/D	0.02	0.03	N/D	0.15	2.77	0.45	2.16	0.20	0.50	6.28
		0.49-0.95	N/D	0.03	0.07	N/D	0.22	4.06	0.68	3.49	0.46	1.14	10.15
		<0.49	N/D	0.02	0.18	0.06	0.22	4.77	0.92	3.92	0.54	1.27	11.9

Table S20: Particulate concentrations of individual CUPs (in $\text{pg}\cdot\text{m}^{-3}$) according to particle size (in μm) at the rural and urban sites. Disulfoton was not included in this dataset. N/D indicates compounds that were not detected, <LOD indicates compounds that were below the limit of detection and <LOQ indicates samples that were below the limit of quantification. LODs and LOQs are found in Table S7. Numbers in bolds represent the particle size fraction with the highest concentration.

	Size fraction (μm)	Acetochlor	Alachlor	Atrazine	Azinphos methyl	Carbendazim	Chlorotoluron	Chlorpyrifos	Diazinon	Dimethachlor	Dimethoate	Diuron	Fenitrothion	
Rural site	Winter	7.2-10	0.05	<LOD	0.21	N/D	N/D	<LOQ	0.94	N/D	N/D	<LOQ	<LOD	
		3.0-7.2	0.07	0.16	0.31	N/D	N/D	N/D	<LOQ	0.76	N/D	N/D	0.06	<LOD
		1.5-3.0	0.06	0.31	0.24	N/D	N/D	N/D	N/D	0.27	N/D	N/D	<LOQ	<LOD
		0.95-1.5	<LOQ	<LOD	0.96	N/D	N/D	N/D	<LOQ	0.32	N/D	N/D	0.21	<LOD
		0.49-0.95	0.09	<LOQ	0.56	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.29	<LOD
		<0.49	0.26	<LOD	N/D	N/D	N/D	N/D	0.15	N/D	N/D	N/D	0.96	<LOD
	Spring	7.2-10	20.91	<LOQ	N/D	0.05	0.44	0.71	0.27	1.20	N/D	N/D	0.11	<LOD
		3.0-7.2	32.96	<LOQ	0.04	0.06	1.38	1.63	0.15	1.87	N/D	N/D	0.24	<LOD
		1.5-3.0	31.67	0.12	0.04	0.06	0.59	0.83	0.28	0.71	N/D	0.01	0.37	<LOD
		0.95-1.5	17.81	<LOD	N/D	N/D	0.19	N/D	0.20	0.30	N/D	0.09	0.34	<LOD
		0.49-0.95	26.33	0.48	0.04	N/D	N/D	N/D	0.24	0.25	N/D	0.26	0.52	<LOD
		<0.49	144.51	<LOD	0.34	N/D	<LOQ	N/D	0.67	0.95	N/D	0.53	2.29	<LOD
	Summer	7.2-10	1.99	0.23	0.05	N/D	0.82	N/D	<LOQ	1.57	N/D	N/D	1.05	<LOD
		3.0-7.2	2.91	0.12	0.13	0.07	2.04	N/D	<LOQ	0.80	N/D	N/D	1.73	<LOD
		1.5-3.0	4.26	0.18	0.10	N/D	0.87	N/D	<LOQ	1.31	N/D	N/D	2.40	<LOD
		0.95-1.5	3.84	0.27	0.03	N/D	0.40	N/D	<LOQ	1.75	N/D	N/D	2.53	<LOD
		0.49-0.95	1.26	0.30	N/D	N/D	0.40	N/D	<LOQ	0.88	N/D	N/D	3.40	<LOD
		<0.49	1.10	0.79	N/D	N/D	0.63	N/D	0.11	0.29	N/D	N/D	16.72	<LOD
Autumn	7.2-10	0.40	N/D	<LOQ	N/D	0.12	N/D	<LOQ	0.69	N/D	N/D	0.16	N/D	
	3.0-7.2	0.39	N/D	<LOD	N/D	0.01	N/D	N/D	0.51	N/D	N/D	0.25	N/D	
	1.5-3.0	0.57	<LOD	N/D	0.06	1.48	0.33	<LOQ	0.95	0.11	N/D	0.36	N/D	
	0.95-1.5	0.26	N/D	N/D	N/D	N/D	N/D	<LOQ	1.16	N/D	N/D	0.76	<LOD	
	0.49-0.95	0.21	1.47	N/D	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	1.41	N/D	
	<0.49	0.64	N/D	N/D	N/D	0.25	N/D	<LOQ	3.29	0.43	N/D	5.11	N/D	

Urban site	Winter	7.2-10	0.03	0.50	0.25	0.05	N/D	N/D	N/D	0.90	N/D	0.02	N/D	N/D
		3.0-7.2	0.04	0.21	0.06	N/D	N/D	N/D	N/D	0.07	N/D	N/D	0.03	N/D
		1.5-3.0	0.06	N/D	0.40	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.07	N/D
		0.95-1.5	0.08	N/D	0.27	N/D	N/D	N/D	N/D	0.23	N/D	N/D	<LOQ	N/D
		0.49-0.95	0.21	N/D	0.56	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	0.06	6.22
		<0.49	0.39	10.47	1.03	N/D	N/D	N/D	0.66	N/D	N/D	N/D	0.20	N/D
	Spring	7.2-10	2.17	0.17	0.35	N/D	0.71	0.63	<LOQ	0.61	N/D	N/D	0.09	N/D
		3.0-7.2	1.59	0.25	0.04	N/D	1.05	1.06	N/D	0.05	N/D	N/D	0.14	N/D
		1.5-3.0	3.67	0.21	0.08	N/D	0.62	0.08	0.11	1.18	N/D	N/D	0.13	N/D
		0.95-1.5	2.61	N/D	N/D	N/D	N/D	N/D	0.31	0.44	N/D	N/D	0.10	N/D
		0.49-0.95	1.14	N/D	0.06	N/D	N/D	N/D	0.17	N/D	N/D	N/D	0.16	N/D
		<0.49	6.95	N/D	0.36	N/D	0.15	N/D	0.30	N/D	N/D	N/D	1.39	N/D
	Summer	7.2-10	0.61	N/D	N/D	N/D	1.21	N/D	<LOQ	1.40	0.35	N/D	0.06	N/D
		3.0-7.2	0.44	<LOQ	<LOQ	N/D	0.98	N/D	0.11	0.96	0.51	N/D	0.17	N/D
		1.5-3.0	1.76	N/D	<LOD	N/D	0.70	N/D	0.12	1.18	0.40	N/D	1.16	N/D
		0.95-1.5	0.62	N/D	0.02	N/D	0.20	N/D	0.06	0.55	N/D	N/D	0.21	N/D
		0.49-0.95	0.68	N/D	N/D	N/D	1.66	N/D	1.06	0.82	0.29	N/D	0.67	N/D
		<0.49	3.53	0.61	0.14	N/D	N/D	N/D	0.31	2.14	7.32	N/D	3.51	N/D
Autumn	7.2-10	0.34	0.25	N/D	N/D	0.06	N/D	<LOQ	1.01	N/D	N/D	0.05	N/D	
	3.0-7.2	0.47	0.37	0.11	N/D	0.36	0.19	N/D	N/D	<LOQ	N/D	0.15	N/D	
	1.5-3.0	0.36	N/D	N/D	N/D	0.21	N/D	N/D	1.47	0.10	N/D	N/D	N/D	
	0.95-1.5	0.24	0.53	N/D	N/D	N/D	N/D	N/D	1.35	<LOD	N/D	<LOQ	<LOD	
	0.49-0.95	0.09	0.88	0.10	N/D	0.01	N/D	N/D	N/D	N/D	N/D	0.26	N/D	
	<0.49	0.22	0.61	0.11	N/D	N/D	N/D	<LOQ	0.79	0.09	N/D	0.29	N/D	

Table S20 (continued): Particulate concentrations of individual CUPs (in pg.m⁻³) according to particle size (in μm) at the rural and urban sites.

	Size fraction (μm)	Fenpropimorph	Fonofos	Isoproturon	Malathion	Metamitron	Metazachlor	Metribuzin	Prochloraz	Pyrazon	Simazine	S-metolachlor	Temephos	Terbufos	Terbuthylazine	ΣCUPs	
Rural site	Winter	7.2-10	0.03	0.04	0.03	<LOQ	<LOD	N/D	<LOD	N/D	N/D	0.19	N/D	0.03	N/D	N/D	1.52
		3.0-7.2	0.03	0.05	0.11	0.08	<LOD	N/D	<LOD	0.08	N/D	0.06	N/D	N/D	0.09	N/D	1.86
		1.5-3.0	0.05	0.06	N/D	0.15	0.16	N/D	<LOQ	N/D	N/D	0.08	N/D	N/D	N/D	N/D	1.38
		0.95-1.5	0.19	0.08	N/D	<LOQ	<LOD	N/D	<LOD	N/D	N/D	0.09	N/D	N/D	N/D	N/D	1.85
		0.49-0.95	0.18	0.06	N/D	<LOQ	<LOD	N/D	0.10	N/D	N/D	0.87	N/D	N/D	N/D	N/D	2.15
		<0.49	0.37	0.07	0.05	<LOQ	<LOD	N/D	0.07	N/D	N/D	0.09	N/D	N/D	0.15	0.03	2.2
	Spring	7.2-10	0.09	0.03	3.79	<LOQ	0.79	N/D	<LOD	0.41	0.53	0.04	1.13	N/D	N/D	1.23	31.73
		3.0-7.2	0.36	0.03	6.52	<LOQ	1.96	N/D	<LOD	0.92	0.98	0.07	1.93	N/D	0.07	2.70	53.87
		1.5-3.0	0.72	0.03	4.71	<LOQ	<LOD	N/D	<LOD	0.36	0.22	0.41	2.27	N/D	0.07	2.06	45.53
		0.95-1.5	1.88	0.04	1.96	<LOQ	<LOD	N/D	<LOD	0.19	0.21	0.33	1.46	N/D	0.09	0.93	26.02
		0.49-0.95	7.47	0.08	0.54	<LOQ	N/D	N/D	<LOD	0.14	0.31	0.06	2.33	N/D	N/D	0.49	39.54
		<0.49	36.05	0.06	2.45	<LOQ	<LOD	N/D	N/D	0.15	N/D	0.61	22.05	N/D	0.12	1.26	212.0
	Summer	7.2-10	1.00	0.04	N/D	<LOQ	<LOD	0.32	<LOD	1.84	0.30	0.04	0.87	N/D	N/D	1.18	11.3
		3.0-7.2	2.30	0.04	0.03	<LOQ	0.23	0.63	<LOD	3.93	0.53	0.25	1.24	N/D	0.08	2.08	19.14
		1.5-3.0	5.06	0.04	N/D	<LOQ	0.13	0.18	<LOD	1.46	0.22	0.05	1.83	N/D	0.08	1.69	19.86
		0.95-1.5	20.98	0.04	0.04	<LOQ	<LOD	0.07	<LOD	0.48	0.11	0.04	1.68	N/D	0.08	1.08	33.42
		0.49-0.95	58.37	0.06	N/D	<LOQ	N/D	0.06	<LOD	0.22	0.16	0.05	0.78	N/D	0.08	0.41	66.43
		<0.49	110.9	0.05	0.11	<LOQ	N/D	1.61	<LOQ	0.32	N/D	0.06	0.84	N/D	0.10	0.36	134.0
Autumn	7.2-10	0.27	<LOD	0.65	N/D	N/D	1.56	N/D	0.11	N/D	0.22	N/D	N/D	N/D	<LOD	4.18	
	3.0-7.2	0.05	N/D	0.33	N/D	N/D	0.59	N/D	0.13	N/D	0.15	N/D	N/D	N/D	<LOD	2.41	
	1.5-3.0	0.22	N/D	1.81	N/D	N/D	2.61	N/D	0.17	N/D	N/D	<LOQ	N/D	0.05	0.02	8.74	
	0.95-1.5	0.23	<LOQ	0.25	N/D	N/D	1.46	N/D	N/D	N/D	0.45	N/D	N/D	N/D	N/D	4.57	
	0.49-0.95	0.53	N/D	0.05	N/D	N/D	1.35	N/D	N/D	N/D	0.49	N/D	0.05	0.08	N/D	5.64	
	<0.49	0.65	0.15	0.34	N/D	N/D	11.44	N/D	N/D	N/D	0.87	N/D	N/D	0.17	<LOQ	23.34	

Urban site	Winter	7.2-10	0.04	N/D	0.04	0.10	N/D	N/D	N/D	0.05	N/D	0.25	N/D	0.01	N/D	N/D	2.24
		3.0-7.2	0.02	0.10	0.11	N/D	N/D	N/D	N/D	N/D	N/D	0.36	N/D	N/D	N/D	N/D	1
		1.5-3.0	0.19	N/D	N/D	<LOD	N/D	N/D	N/D	N/D	N/D	0.95	N/D	0.02	0.03	N/D	1.72
		0.95-1.5	0.09	N/D	<LOD	N/D	N/D	N/D	N/D	N/D	N/D	0.31	N/D	N/D	N/D	N/D	0.98
		0.49-0.95	0.20	0.39	<LOD	N/D	N/D	N/D	0.43	0.04	N/D	0.80	N/D	N/D	N/D	N/D	8.91
		<0.49	0.38	N/D	0.04	<LOQ	N/D	1.05	N/D	0.11	N/D	0.83	N/D	0.04	0.12	0.12	15.44
	Spring	7.2-10	0.16	N/D	4.84	N/D	N/D	N/D	N/D	0.20	0.65	0.28	0.57	N/D	N/D	1.74	13.17
		3.0-7.2	0.22	N/D	6.26	N/D	0.23	N/D	N/D	0.22	0.87	0.30	0.49	0.03	N/D	2.09	14.89
		1.5-3.0	0.40	N/D	2.39	0.07	N/D	N/D	N/D	0.19	N/D	0.17	1.30	N/D	N/D	1.35	11.95
		0.95-1.5	1.67	N/D	0.95	N/D	N/D	N/D	N/D	0.15	N/D	0.29	1.01	N/D	N/D	0.59	8.12
		0.49-0.95	3.63	N/D	0.28	N/D	N/D	N/D	N/D	0.09	N/D	0.49	0.21	<LOQ	N/D	0.09	6.32
		<0.49	6.59	N/D	1.79	N/D	N/D	0.03	N/D	N/D	N/D	0.85	2.31	N/D	N/D	0.38	21.1
	Summer	7.2-10	0.24	0.08	N/D	N/D	N/D	2.48	N/D	0.99	0.43	0.33	0.53	0.03	N/D	0.39	9.13
		3.0-7.2	0.43	N/D	N/D	N/D	N/D	0.97	N/D	1.65	0.17	0.38	0.66	N/D	<LOQ	0.54	7.97
		1.5-3.0	1.67	N/D	N/D	N/D	N/D	2.55	N/D	0.73	N/D	0.31	0.88	N/D	N/D	0.93	12.39
		0.95-1.5	1.91	N/D	N/D	N/D	N/D	0.65	0.56	0.15	N/D	0.34	0.41	N/D	<LOQ	0.44	6.12
		0.49-0.95	10.85	N/D	N/D	N/D	N/D	0.91	N/D	0.18	1.70	0.38	0.64	N/D	N/D	0.50	20.34
		<0.49	22.58	N/D	0.11	N/D	N/D	8.43	N/D	0.16	N/D	0.91	4.67	N/D	N/D	2.02	56.44
Autumn	7.2-10	0.04	N/D	1.09	N/D	N/D	1.17	N/D	0.13	N/D	0.24	<LOQ	N/D	0.03	0.04	4.45	
	3.0-7.2	N/D	N/D	1.77	N/D	N/D	1.39	N/D	0.28	N/D	0.43	N/D	N/D	<LOQ	<LOQ	5.52	
	1.5-3.0	0.02	N/D	0.71	N/D	N/D	0.69	N/D	0.15	N/D	0.23	0.10	N/D	<LOQ	<LOD	4.04	
	0.95-1.5	0.07	N/D	0.32	N/D	N/D	0.46	N/D	0.15	N/D	0.42	<LOQ	N/D	0.04	0.06	3.64	
	0.49-0.95	0.11	N/D	0.02	N/D	N/D	5.02	N/D	N/D	N/D	0.38	<LOD	N/D	0.02	N/D	6.89	
	<0.49	0.18	N/D	0.37	N/D	N/D	2.68	N/D	N/D	N/D	1.06	<LOD	N/D	0.05	0.03	6.48	

Table S21: Particle-phase concentrations of individual OCPs (in ng.g(PM)⁻¹) according to particle size (in µm) at the rural and the urban sites. N/D indicates compounds that were not detected.

		Size fraction (in µm)	α-HCH	β-HCH	γ-HCH	δ-HCH	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>o,p'</i> -DDD	<i>p,p'</i> -DDD	<i>o,p'</i> -DDT	<i>p,p'</i> -DDT	ΣOCPs
Rural site	Winter	7.2-2010	0.39	1.31	4.68	0.98	1.33	18.24	0.92	5.30	8.38	40.09	81.6
		3.0-7.2	2.20	0.24	4.48	1.06	0.72	12.38	0.76	5.03	3.40	18.76	49.0
		1.5-3.0	1.96	1.45	3.62	0.38	0.82	11.02	0.67	4.66	2.65	16.39	43.6
		0.95-1.5	0.38	0.39	0.97	0.40	0.69	11.18	1.10	7.16	2.00	13.59	37.9
		0.49-0.95	N/D	0.17	0.47	0.20	0.66	11.14	0.90	6.26	1.54	11.27	32.6
		<0.49	1.40	0.57	1.77	1.14	0.57	11.59	0.84	6.44	3.21	31.22	58.8
	Spring	7.2-2010	5.77	1.11	12.72	6.27	1.34	32.54	1.00	2.74	5.19	14.00	82.7
		3.0-7.2	0.94	N/D	3.28	1.43	0.21	8.21	0.20	1.07	1.21	4.31	20.9
		1.5-3.0	1.74	N/D	4.43	1.79	0.25	10.88	0.31	1.44	1.50	6.44	28.8
		0.95-1.5	2.27	0.15	4.67	1.63	0.25	11.40	0.62	1.70	2.06	11.77	36.5
		0.49-0.95	0.22	0.25	1.19	0.13	0.22	11.12	0.39	2.10	1.88	11.85	29.4
		<0.49	0.07	2.19	0.55	0.29	0.10	3.35	0.27	0.39	1.19	8.09	16.5
	Summer	7.2-2010	2.90	N/D	15.10	N/D	1.59	69.95	7.63	25.04	3.58	N/D	125.8
		3.0-7.2	0.74	N/D	2.62	N/D	0.30	10.38	1.33	5.14	0.57	0.88	22.0
		1.5-3.0	6.48	0.36	2.34	N/D	0.43	18.50	2.08	9.37	N/D	N/D	39.6
		0.95-1.5	1.22	N/D	1.83	0.59	0.12	7.93	0.94	4.24	N/D	0.19	17.1
		0.49-0.95	1.38	0.90	1.40	0.65	0.15	6.95	0.82	3.05	0.23	1.44	17.0
		<0.49	0.36	1.69	0.71	0.24	0.28	9.45	0.64	3.21	0.62	1.47	18.7
Autumn	7.2-2010	0.71	2.03	2.86	0.26	1.01	43.97	4.88	17.42	0.29	N/D	73.4	
	3.0-7.2	0.10	N/D	0.31	N/D	0.41	19.28	1.66	8.60	0.11	N/D	30.5	
	1.5-3.0	N/D	2.48	0.73	0.50	0.57	27.53	2.30	12.95	N/D	0.02	47.1	
	0.95-1.5	0.61	0.74	0.68	0.50	0.47	24.72	1.90	12.25	0.32	0.85	43.0	
	0.49-0.95	0.05	0.34	0.17	0.11	0.39	24.24	1.78	12.50	0.52	1.54	41.6	
	<0.49	0.14	0.27	0.07	N/D	0.29	17.57	1.36	10.15	1.00	4.00	34.9	

Urban site	Winter	7.2.2010	N/D	7.27	0.93	N/D	0.57	5.20	1.02	3.29	0.21	N/D	18.5
		3.0-7.2	N/D	2.86	0.40	N/D	0.45	4.02	0.79	2.10	0.25	0.44	11.3
		1.5-3.0	N/D	1.89	0.98	N/D	0.95	8.99	1.53	4.62	0.50	0.85	20.3
		0.95-1.5	N/D	1.17	0.62	0.11	0.92	10.17	1.71	5.95	0.73	0.74	22.1
		0.49-0.95	0.03	0.36	0.37	N/D	0.56	5.65	1.03	4.05	0.24	0.44	12.7
		<0.49	0.45	0.35	0.68	0.38	0.40	4.79	1.03	3.87	0.87	0.83	13.7
	Spring	7.2.2010	0.16	6.03	1.00	0.38	0.55	7.42	1.76	6.27	0.65	0.61	24.8
		3.0-7.2	0.63	4.54	0.93	0.34	0.40	5.60	1.21	4.31	0.38	0.44	18.8
		1.5-3.0	N/D	N/D	0.42	N/D	0.62	9.36	2.15	7.69	0.50	0.39	21.1
		0.95-1.5	N/D	0.84	0.31	N/D	0.58	8.17	1.92	6.91	0.56	0.62	19.9
		0.49-0.95	0.03	N/D	0.45	N/D	0.46	7.36	1.71	6.73	0.53	0.78	18.1
		<0.49	0.21	1.61	0.97	N/D	0.41	6.32	1.41	5.84	0.78	1.42	19.0
	Summer	7.2.2010	N/D	4.63	0.38	N/D	0.51	6.53	1.65	5.48	0.63	0.58	20.4
		3.0-7.2	0.32	0.26	0.49	N/D	0.47	6.95	1.72	6.74	0.81	1.29	19.1
		1.5-3.0	2.01	2.33	1.61	N/D	0.74	9.70	2.55	9.41	1.63	3.50	33.5
		0.95-1.5	1.60	0.94	1.29	N/D	0.91	11.52	3.14	11.48	1.29	1.79	34.0
		0.49-0.95	1.46	6.29	1.65	0.92	0.45	6.03	1.51	6.37	0.86	1.63	27.2
		<0.49	0.66	1.38	1.29	0.79	0.48	6.58	1.45	5.27	1.03	1.33	20.3
Autumn	7.2.2010	0.15	3.44	1.21	0.30	0.72	14.45	1.97	8.77	1.19	2.32	34.5	
	3.0-7.2	N/D	1.78	0.26	N/D	0.46	7.94	1.21	4.41	0.72	0.92	17.7	
	1.5-3.0	N/D	2.55	0.48	N/D	0.75	13.00	2.34	8.89	0.84	2.00	30.9	
	0.95-1.5	N/D	0.03	0.04	N/D	0.19	3.56	0.58	2.78	0.26	0.64	8.1	
	0.49-0.95	N/D	0.07	0.17	N/D	0.55	10.32	1.72	8.89	1.17	2.90	25.8	
	<0.49	N/D	0.04	0.35	0.11	0.43	9.15	1.76	7.53	1.04	2.43	22.8	

Table S22: Particulate concentrations of individual CUPs (in $\mu\text{g.g}(\text{PM})^{-1}$) according to particle size (in μm) at the rural and urban sites. N/D indicates compounds that were not detected, <LOD indicates compounds that were below the limit of detection and <LOQ indicates samples that were below the limit of quantification. LODs and LOQs are found in Table S7.

	Size fraction (μm)	Acetochlor	Alachlor	Atrazine	Azinphos methyl	Carbendazim	Chlorotoluron	Chlorpyrifos	Diazinon	Dimethachlor	Dimethoate	Diuron	Fenitrothion	Fenproimorph	
Rural site	Winter	7.2-10	0.097	<LOD	0.376	N/D	N/D	<LOQ	1.706	N/D	N/D	<LOQ	<LOD	0.046	
		3.0-7.2	0.032	0.068	0.134	N/D	N/D	N/D	<LOQ	0.330	N/D	N/D	0.027	<LOD	0.015
		1.5-3.0	0.015	0.080	0.063	N/D	N/D	N/D	N/D	0.071	N/D	N/D	<LOQ	<LOD	0.014
		0.95-1.5	<LOQ	<LOD	0.096	N/D	N/D	N/D	<LOQ	0.032	N/D	N/D	0.021	<LOD	0.019
		0.49-0.95	0.007	<LOQ	0.046	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.024	<LOD	0.014
		<0.49	0.031	<LOD	N/D	N/D	N/D	N/D	0.018	N/D	N/D	N/D	0.115	<LOD	0.044
	Spring	7.2-10	33.891	<LOQ	N/D	0.086	0.717	1.156	0.430	1.937	N/D	N/D	0.172	<LOD	0.138
		3.0-7.2	15.897	<LOQ	0.019	0.031	0.666	0.784	0.071	0.904	N/D	N/D	0.116	<LOD	0.173
		1.5-3.0	21.749	0.085	0.028	0.038	0.405	0.572	0.190	0.487	N/D	0.008	0.256	<LOD	0.496
		0.95-1.5	6.634	<LOD	N/D	N/D	0.072	N/D	0.074	0.112	N/D	0.032	0.125	<LOD	0.701
		0.49-0.95	4.939	0.089	0.007	N/D	N/D	N/D	0.045	0.047	N/D	0.049	0.098	<LOD	1.401
		<0.49	30.076	<LOD	0.071	N/D	<LOQ	N/D	0.139	0.199	N/D	0.109	0.477	<LOD	7.504
	Summer	7.2-10	3.225	0.373	0.077	N/D	1.327	N/D	<LOQ	2.546	N/D	N/D	1.697	<LOD	1.624
		3.0-7.2	1.091	0.046	0.049	0.026	0.763	N/D	<LOQ	0.299	N/D	N/D	0.648	<LOD	0.863
		1.5-3.0	3.568	0.151	0.085	N/D	0.728	N/D	<LOQ	1.099	N/D	N/D	2.007	<LOD	4.238
		0.95-1.5	3.175	0.227	0.025	N/D	0.331	N/D	<LOQ	1.443	N/D	N/D	2.089	<LOD	17.329
		0.49-0.95	0.427	0.101	N/D	N/D	0.135	N/D	<LOQ	0.299	N/D	N/D	1.154	<LOD	19.805
		<0.49	0.191	0.138	N/D	N/D	0.109	N/D	0.020	0.051	N/D	N/D	2.916	<LOD	19.361
	Autumn	7.2-10	0.189	N/D	<LOQ	N/D	0.056	N/D	<LOQ	0.327	N/D	N/D	0.075	N/D	0.126
		3.0-7.2	0.145	N/D	<LOD	N/D	0.005	N/D	N/D	0.188	N/D	N/D	0.093	N/D	0.017
		1.5-3.0	0.220	<LOD	N/D	0.023	0.569	0.126	<LOQ	0.366	0.044	N/D	0.140	N/D	0.083
		0.95-1.5	0.079	N/D	N/D	N/D	N/D	N/D	<LOQ	0.348	N/D	N/D	0.227	<LOD	0.070

Urban site		0.49-0.95	0.043	0.297	N/D	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	0.284	N/D	0.107
		<0.49	0.143	N/D	N/D	N/D	0.057	N/D	<LOQ	0.737	0.096	N/D	1.144	N/D	0.145
	Winter	7.2-10	0.010	0.152	0.075	0.016	N/D	N/D	N/D	0.276	N/D	0.005	N/D	N/D	0.012
		3.0-7.2	0.006	0.036	0.011	N/D	N/D	N/D	N/D	0.012	N/D	N/D	0.006	N/D	0.003
		1.5-3.0	0.017	N/D	0.106	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.020	N/D	0.050
		0.95-1.5	0.010	N/D	0.033	N/D	N/D	N/D	N/D	0.028	N/D	N/D	<LOQ	N/D	0.011
		0.49-0.95	0.018	N/D	0.049	N/D	N/D	N/D	<LOQ	N/D	N/D	N/D	0.005	0.545	0.018
		<0.49	0.031	0.841	0.083	N/D	N/D	N/D	0.053	N/D	N/D	N/D	0.016	N/D	0.031
	Spring	7.2-10	0.456	0.035	0.073	N/D	0.149	0.133	<LOQ	0.129	N/D	N/D	0.018	N/D	0.035
		3.0-7.2	0.221	0.035	0.005	N/D	0.146	0.146	N/D	0.007	N/D	N/D	0.019	N/D	0.030
		1.5-3.0	1.068	0.061	0.023	N/D	0.181	0.023	0.032	0.344	N/D	N/D	0.039	N/D	0.115
		0.95-1.5	0.630	N/D	N/D	N/D	N/D	N/D	0.075	0.107	N/D	N/D	0.025	N/D	0.403
		0.49-0.95	0.160	N/D	0.008	N/D	N/D	N/D	0.023	0.000	N/D	N/D	0.023	N/D	0.511
		<0.49	0.628	N/D	0.033	N/D	0.014	N/D	0.027	0.000	N/D	N/D	0.126	N/D	0.595
	Summer	7.2-10	0.291	N/D	N/D	N/D	0.578	N/D	0.019	0.668	0.168	N/D	0.030	N/D	0.116
		3.0-7.2	0.125	<LOQ	<LOQ	N/D	0.278	N/D	0.032	0.273	0.146	N/D	0.048	N/D	0.123
		1.5-3.0	0.790	N/D	<LOD	N/D	0.314	N/D	0.055	0.529	0.178	N/D	0.521	N/D	0.750
		0.95-1.5	0.375	N/D	0.014	N/D	0.119	N/D	0.038	0.334	N/D	N/D	0.127	N/D	1.159
		0.49-0.95	0.181	N/D	N/D	N/D	0.439	N/D	0.280	0.218	0.077	N/D	0.176	N/D	2.868
		<0.49	0.408	0.071	0.016	N/D	N/D	N/D	0.036	0.248	0.848	N/D	0.406	N/D	2.613
	Autumn	7.2-10	0.150	0.112	N/D	N/D	0.028	N/D	<LOQ	0.451	N/D	N/D	0.022	N/D	0.018
		3.0-7.2	0.131	0.105	0.031	N/D	0.102	0.053	N/D	N/D	<LOQ	N/D	0.043	N/D	N/D
		1.5-3.0	0.199	N/D	N/D	N/D	0.117	N/D	N/D	0.806	0.058	N/D	N/D	N/D	0.010
		0.95-1.5	0.105	0.231	N/D	N/D	N/D	N/D	N/D	0.591	<LOD	N/D	<LOQ	<LOD	0.030
0.49-0.95		0.020	0.204	0.022	N/D	0.003	N/D	N/D	N/D	N/D	N/D	0.061	N/D	0.026	
<0.49		0.028	0.079	0.014	N/D	N/D	N/D	<LOQ	0.103	0.012	N/D	0.038	N/D	0.023	

Table S22 (continued): Particulate concentrations of individual CUPs (in $\mu\text{g.g(PM)}^{-1}$) according to particle size (in μm) at the rural and the urban sites.

	Size fraction (μm)	Fonofos	Isoproturon	Malathion	Metamitron	Metazachlor	S-metolachlor	Metribuzin	Prochloraz	Pyrazon	Simazine	Temephos	Terbufos	Terbuthylazine	ΣCUPs	
Rural site	Winter	7.2-10	0.080	0.047	<LOQ	<LOD	N/D	N/D	<LOD	N/D	N/D	0.344	0.063	N/D	N/D	2.759
		3.0-7.2	0.020	0.047	0.034	<LOD	N/D	N/D	<LOD	0.036	N/D	0.027	N/D	0.038	N/D	0.808
		1.5-3.0	0.016	N/D	0.040	0.041	N/D	N/D	<LOQ	N/D	N/D	0.020	N/D	N/D	N/D	0.36
		0.95-1.5	0.008	N/D	<LOQ	<LOD	N/D	N/D	<LOD	N/D	N/D	0.009	N/D	N/D	N/D	0.185
		0.49-0.95	0.005	N/D	<LOQ	<LOD	N/D	N/D	0.008	N/D	N/D	0.071	N/D	N/D	N/D	0.175
		<0.49	0.009	0.006	<LOQ	<LOD	N/D	N/D	0.009	N/D	N/D	0.011	N/D	0.018	0.004	0.265
	Spring	7.2-10	0.055	6.142	<LOQ	1.286	N/D	1.834	<LOD	0.665	0.857	0.063	N/D	N/D	1.991	51.42
		3.0-7.2	0.016	3.142	<LOQ	0.945	N/D	0.931	<LOD	0.442	0.473	0.031	N/D	0.032	1.303	25.976
		1.5-3.0	0.023	3.237	<LOQ	<LOD	N/D	1.558	<LOD	0.245	0.149	0.279	N/D	0.046	1.414	31.265
		0.95-1.5	0.017	0.729	<LOQ	<LOD	N/D	0.543	<LOD	0.069	0.077	0.124	N/D	0.034	0.345	9.688
		0.49-0.95	0.015	0.101	<LOQ	N/D	N/D	0.436	<LOD	0.026	0.058	0.011	N/D	N/D	0.093	7.415
		<0.49	0.012	0.509	<LOQ	<LOD	N/D	4.590	N/D	0.032	N/D	0.127	N/D	0.026	0.263	44.134
	Summer	7.2-10	0.061	N/D	<LOQ	<LOD	0.519	1.403	<LOD	2.972	0.487	0.072	N/D	N/D	1.915	18.298
		3.0-7.2	0.015	0.012	<LOQ	0.085	0.238	0.465	<LOD	1.474	0.199	0.093	N/D	0.030	0.779	7.175
		1.5-3.0	0.033	N/D	<LOQ	0.112	0.148	1.529	<LOD	1.225	0.186	0.039	N/D	0.067	1.412	16.627
		0.95-1.5	0.032	0.036	<LOQ	<LOD	0.058	1.386	<LOD	0.400	0.089	0.037	N/D	0.063	0.892	27.612
		0.49-0.95	0.021	N/D	<LOQ	N/D	0.022	0.265	<LOD	0.074	0.054	0.016	N/D	0.027	0.140	22.54
		<0.49	0.009	0.019	<LOQ	N/D	0.281	0.146	<LOQ	0.056	N/D	0.010	N/D	0.017	0.063	23.387
Autumn	7.2-10	<LOD	0.307	N/D	N/D	0.738	N/D	N/D	0.054	N/D	0.105	N/D	N/D	<LOD	1.977	
	3.0-7.2	N/D	0.123	N/D	N/D	0.219	N/D	N/D	0.047	N/D	0.056	N/D	N/D	<LOD	0.893	
	1.5-3.0	N/D	0.695	N/D	N/D	1.002	<LOQ	N/D	0.066	N/D	N/D	N/D	0.021	0.009	3.364	
	0.95-1.5	<LOQ	0.074	N/D	N/D	0.436	N/D	N/D	N/D	N/D	0.134	N/D	N/D	N/D	1.368	
	0.49-0.95	N/D	0.010	N/D	N/D	0.273	N/D	N/D	N/D	N/D	0.099	0.009	0.016	N/D	1.138	
	<0.49	0.033	0.076	N/D	N/D	2.563	N/D	N/D	N/D	N/D	0.196	N/D	0.038	<LOQ	5.228	

Urban site	Winter	7.2-10	N/D	0.011	0.030	N/D	N/D	N/D	N/D	0.015	N/D	0.075	0.003	N/D	N/D	0.68
		3.0-7.2	0.018	0.019	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.061	N/D	N/D	N/D	0.172
		1.5-3.0	N/D	N/D	<LOD	N/D	N/D	N/D	N/D	N/D	N/D	0.252	0.005	0.008	N/D	0.458
		0.95-1.5	N/D	<LOD	N/D	N/D	N/D	N/D	N/D	N/D	N/D	0.038	N/D	N/D	N/D	0.12
		0.49-0.95	0.034	<LOD	N/D	N/D	N/D	N/D	0.038	0.004	N/D	0.070	N/D	N/D	N/D	0.781
		<0.49	N/D	0.003	<LOQ	N/D	0.084	N/D	N/D	0.009	N/D	0.067	0.003	0.010	0.010	1.241
	Spring	7.2-10	N/D	1.018	N/D	N/D	N/D	0.119	N/D	0.043	0.138	0.058	N/D	N/D	0.366	2.77
		3.0-7.2	N/D	0.867	N/D	0.031	N/D	0.067	N/D	0.030	0.120	0.042	0.004	N/D	0.290	2.06
		1.5-3.0	N/D	0.697	0.022	N/D	N/D	0.379	N/D	0.055	N/D	0.048	N/D	N/D	0.394	3.481
		0.95-1.5	N/D	0.230	N/D	N/D	N/D	0.244	N/D	0.035	N/D	0.069	N/D	N/D	0.143	1.961
		0.49-0.95	N/D	0.039	N/D	N/D	N/D	0.029	N/D	0.013	N/D	0.069	<LOQ	N/D	0.013	0.888
		<0.49	N/D	0.162	N/D	N/D	0.003	0.209	N/D	N/D	N/D	0.076	N/D	N/D	0.035	1.908
	Summer	7.2-10	0.039	N/D	N/D	N/D	1.184	0.251	N/D	0.471	0.207	0.156	0.015	N/D	0.187	4.38
		3.0-7.2	N/D	N/D	N/D	N/D	0.277	0.187	N/D	0.469	0.047	0.108	N/D	<LOQ	0.152	2.265
		1.5-3.0	N/D	N/D	N/D	N/D	1.144	0.395	N/D	0.326	N/D	0.141	N/D	N/D	0.416	5.559
		0.95-1.5	N/D	N/D	N/D	N/D	0.395	0.246	0.341	0.093	N/D	0.204	N/D	<LOQ	0.268	3.713
		0.49-0.95	N/D	N/D	N/D	N/D	0.240	0.170	N/D	0.048	0.448	0.100	N/D	N/D	0.133	5.378
		<0.49	N/D	0.012	N/D	N/D	0.975	0.541	N/D	0.018	N/D	0.105	N/D	N/D	0.234	6.531
Autumn	7.2-10	N/D	0.489	N/D	N/D	0.522	<LOQ	N/D	0.058	N/D	0.110	N/D	0.012	0.018	1.99	
	3.0-7.2	N/D	0.498	N/D	N/D	0.391	N/D	N/D	0.079	N/D	0.121	N/D	<LOQ	<LOQ	1.554	
	1.5-3.0	N/D	0.392	N/D	N/D	0.382	0.057	N/D	0.081	N/D	0.126	N/D	<LOQ	<LOD	2.228	
	0.95-1.5	N/D	0.142	N/D	N/D	0.202	<LOQ	N/D	0.065	N/D	0.186	N/D	0.016	0.027	1.595	
	0.49-0.95	N/D	0.005	N/D	N/D	1.159	<LOD	N/D	N/D	N/D	0.087	N/D	0.005	N/D	1.592	
	<0.49	N/D	0.048	N/D	N/D	0.348	<LOD	N/D	N/D	N/D	0.138	ND	0.006	0.004	0.841	

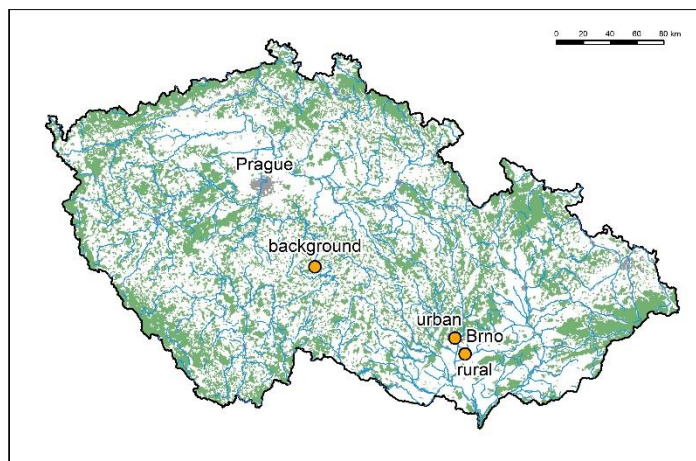


Figure S1: Map of the sampling sites

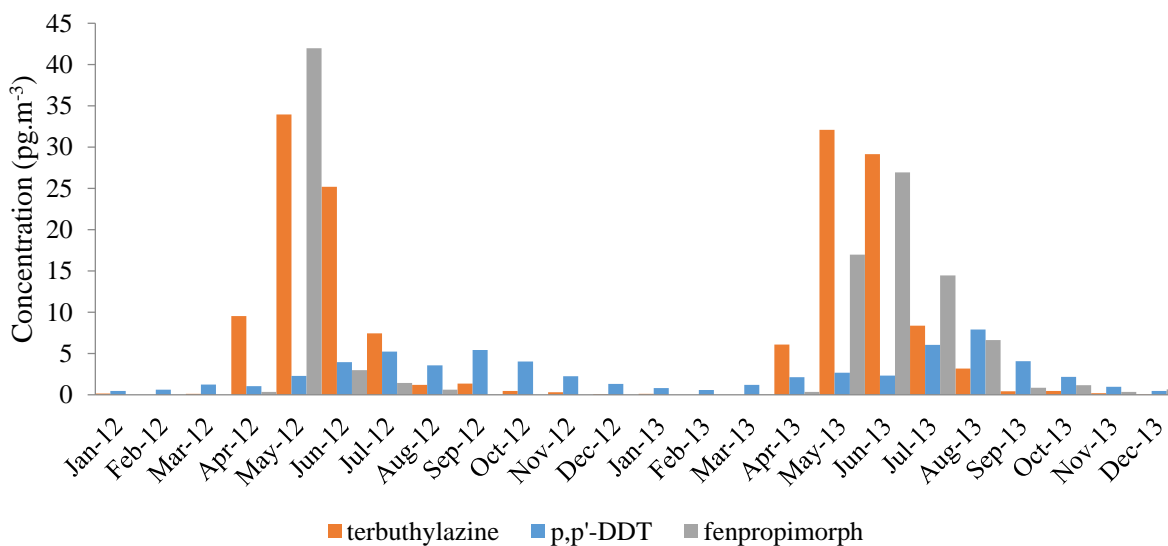


Figure S2: Monthly concentrations (in pg.m^{-3}) of terbuthylazine, *p,p'*-DDT and fenpropimorph.

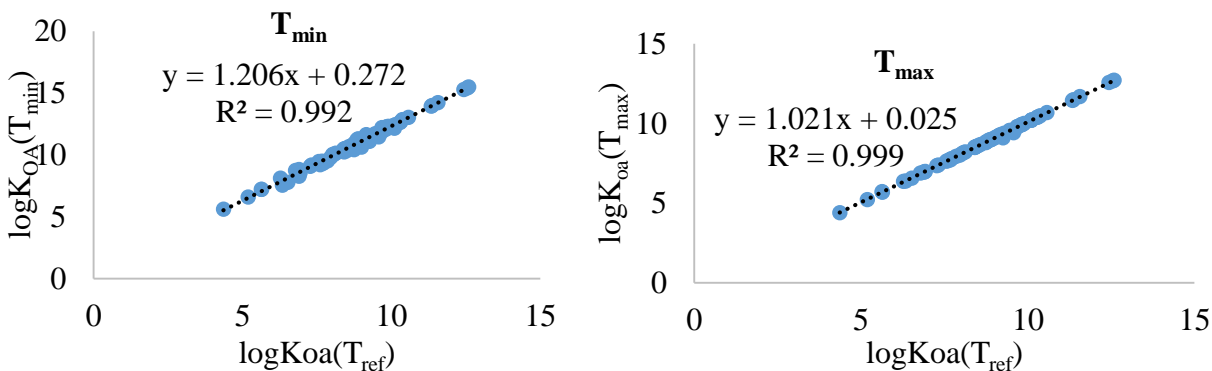


Figure S3: Linear regression of $\log K_{OA}(T)$ versus $\log K_{OA}(T_{ref})$ ($T_{ref} = 25^{\circ}\text{C}$) of OCPs, CBs, PCBs, PAHs, CAR and PCNs for T_{min} ($T_{min} = -13.3^{\circ}\text{C}$) and T_{max} ($T_{max} = 22.8^{\circ}\text{C}$) used to determine the $\log K_{OA}$ of all CUPs except chlorpyrifos. Experimental values from references 7-13.(Harner and Bidleman, 1998a; Harner and Mackay, 1995; Odabasi and Cetin, 2012b; Odabasi et al., 2006a, 2006b; Shoeib and Harner, 2002b; Zhang et al., 2009)

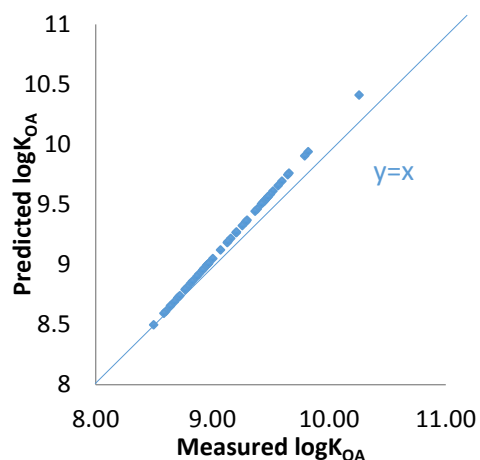


Figure S4: Comparison of $\log K_{OA}$ values of chlorpyrifos at different temperatures determined based on experimental values (Odabasi and Cetin, 2012b) and extrapolated from the regression analysis.

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