

SUPPORTING INFORMATION TO ARTICLE:

Target and Suspect Screening Analysis Reveals Persistent Emerging Organic Contaminants in Soils and Sediments

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Sampling collection

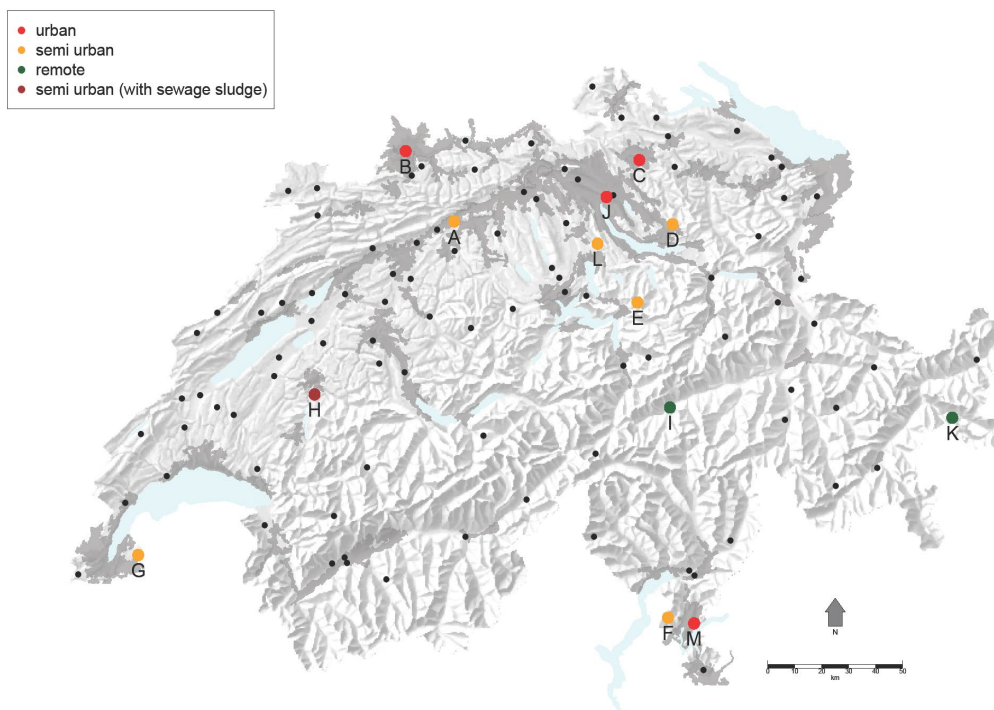


Figure S1. Map of Switzerland showing 102 NABO observation sites spread across Switzerland. Samples used in this study (13 different locations) are illustrated with red, orange, green and marron colors to indicate, urban, semi urban, remote regions. The samples were taken between 2005 and 2009. An additional sample was analyzed at sampling point H which dated back to 1994 and received domestic waste (H (2)). More details about the site can be found in Table S2.

Table S1. Selected soil properties from the Swiss National Soil Monitoring Network (NABO).

ID	Use	Silt (%)	Volume (%)	C _{org} (%)	pH (CaCl ₂)
A	Agriculture	41.0	26.8	2.9	6.2
B	Agriculture	34.0	17.0	1.1	5.5
C	City Park	30.3	20.3	2.2	5.0
M	City Park	36.0	12.0	2.5	5.6
D	Coniferous forest	27.8	30.8	5.8	3.6
E	Coniferous forest	0.0	0.0	13.1	5.8
F	Deciduous forest	0.0	0.0	11.7	3.9
G	Deciduous forest	50.5	23.5	2.7	4.4
H	Grasslands , low intensity	34.8	19.3	2.7	5.9
I	Grasslands , low intensity	36.8	12.5	3.6	5.5
J	Mixed forest	43.5	20.0	2.4	4.5
K	Protection site	28.0	19.0	3.6	5.4
L	Protection site	0.0	0.0	39.2	2.9

Sediments. Sediment cores were extracted as shown in Figure S2 and S3 using a free-fall gravity corer and stored vertically in the dark at 4°C until analysis. The sediment cores were opened and sectioned into intervals between 3 and 5 cm in thickness representing 5 to 10 years' time-frame using stainless steel slicers. Additional “blank” samples were taken from the pre-industrial sediments of the core. Sediments were transferred to airtight glass jars, frozen for 24 h and freeze-dried for 48 h. Full site description of Greifensee is provided in Table S2. Sediment cores were dated using ^{210}Pb and ^{137}Cs signals from Chernobyl (1986) and the atomic bomb tests (1963) using gamma spectroscopy (HPGe GCW 3523) with sedimentation rates from 0.3 to 1.5 cm/year as reported in Table S3. Total carbon (TC) content was measured using an elemental analyser (EURO EA 3000). Total inorganic carbon (TIC) was obtained from a titration Coulometer (CM5015). Total organic carbon (TOC) was calculated as $\text{TOC} = \text{TC} - \text{TIC}$. TOC values are reported in Table S3.

Table S2. Morphology and hydrological characteristics of Greifensee.

	Lake basin
Lake area (km ²)	8.5
Maximum depth (m)	32
Mean depth (m)	18
Lake volume (km ³)	0.15
Mean residence time (years)	1.1
	Watershed
Total area (km ²)	160

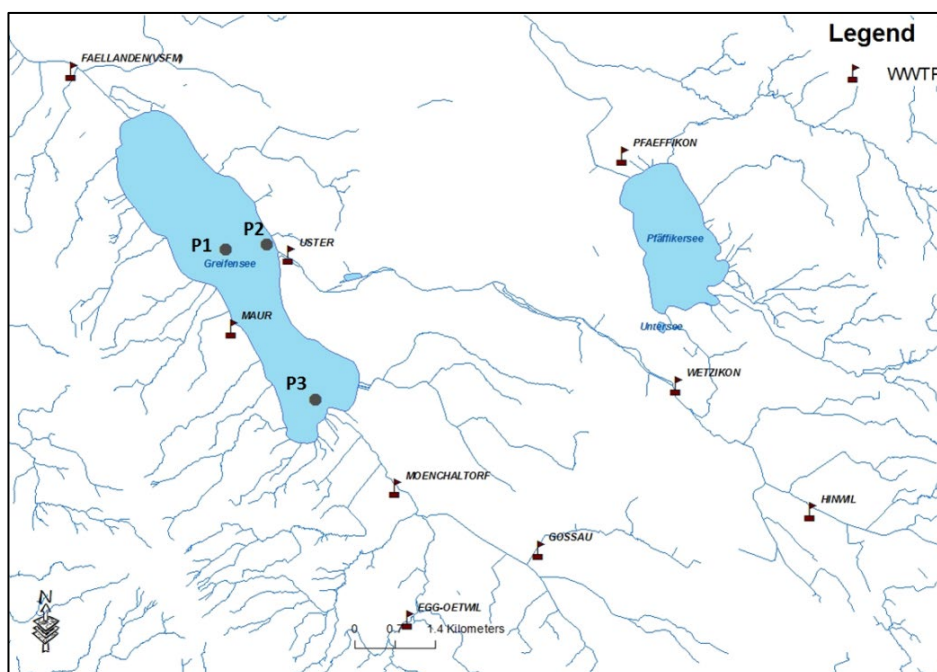


Figure S2. Map of Greifensee displaying different waste water treatment plants (WWTP) in red around the catchment as well as the tributaries. Sampling locations are displayed in black dots with P symbols. Sampling point 1 (P1, 8.678105078° N, 47.351545874° E) was collected from the deepest part of the lake. Sampling points P2 and P3 were collected near the outlet of the Uster WWTP (P2, 8.687325778° N, 47.352453944° E) and Mönchaltorfer WWTP at Aaspitz (P3, 8.697792119° N, 47.328196292° E) to cover the impact of WWTP effluents and agricultural inputs to the catchment.

*Data sources: swisstopo (art. 30 GeoIV): 5704 000 000 / DHM252003, Vector200©2015, TLM, swissboundaries, reproduced with permission of swisstopo/JA 100119.

Table S3. Sedimentation rates and total organic content (TOC) measured for P1, P2 and P3 sampling points in Greifensee. Sampling point 1 (P1) was collected from the deepest part of the lake. Sampling points P2 and P3 were collected near the outlet of the Uster WWTP and Mönchaltorfer WWTP at Aaspitz (to cover the impact of WWTP effluents and agricultural inputs to the catchment).

Deepest part		Mönchaltorfer WWTP		Aaspitz	
TOC (%)	Years (0.3 cm/year)	TOC (%)	Years (0.9 cm/year)	TOC%	Years (1.5 cm/year)
3.7	2014-2004	4.0	2014-2010	3.6	2016-2011
3.3	2004-1995	3.7	2010-2006	3.4	2011-2006
3.1	1995-1984	4.2	2006-2002	4.0	2006-2002
3.7	1984-1974	3.2	2002-1998	3.4	2002-1998
3.9	1974-1964	2.3	1998-1994	3.5	1988-1990
3.2	1964-1954	4.4	1994-1990	3.4	1990-1983
3.0	1954-1944	4.6	1990-1986	2.7	1983-1980
1.7	1944-1934	3.0	1986-1982	2.6	1980-1977
1.6	1934-1918	3.2	1982-1978	2.7	1977-1974
1.4	1918-1902	3.7	1978-1974	3.1	1974-1970
1.4	1902-1886	2.9	1970-1966	3.2	1970-1966
1.5	1889-1870	4.3	1966-1963	3.3	1966-1962
		4.4	1963-1958		
P1 (8.678105078° N, 47.351545874° E)		P2 (8.687325778° N, 47.352453944° E)		P3 (8.697792119° N, 47.328196292° E)	



Figure S3. Sediment cores from Greifensee from the deepest part of the lake (P1) and near the shore of Uster (P2) and Aaspitz (P3). Location of points is illustrated in Figure S2.

Standards and reagents

Ethylacetate ($\geq 99.7\%$) was purchased from Sigma Aldrich (Steinheim, Germany). Acetone ($\geq 99.8\%$) and formic acid ($\geq 98\%$) from Merck (Darmstadt, Germany). Isopropanol ($\geq 99.5\%$) and methanol ($\geq 99.9\%$) were purchased from Acros Organics (New Jersey, USA) and Fisher Scientific (Wohlen, Switzerland), respectively. Nano-pure water was obtained from a Barnstead Nanopure stationary laboratory water system (Barnstead Nanopure Thermo Scientific, San Jose, U.S.). The internal standards (purity $\geq 97\%$) used for quality control, mass recalibration and intensity normalization were purchased from the following suppliers: Sigma Aldrich (Steinheim, Germany), TRC Canada (Toronto, Canada), TCI Europe (Antwerpen, Belgium), Dr. Ehrenstorfer (Augsburg, Germany), Novartis (Basel, Switzerland), ReseaLIFEchem GmbH (Burgdorf, Switzerland), CDN Isotopes Inc. (Augsburg, Germany), and Lipomed AG (Arlesheim, Switzerland).

LC-HRMS/MS analysis

Sediment and soils were extracted by pressurized liquid extraction (PLE) using an in-cell cleanup technique that employed Florisil as sorbing phase, as described in detail elsewhere (Chiaia-Hernández et al., 2017; Chiaia-Hernandez et al., 2013; Pintado-Herrera et al., 2016). Briefly, the extraction was performed with 10-mL stainless steel cells prepared by placing, from bottom to top, a 27-mm glass fiber filter, a 16.2-mm cellulose filter (Dionex, Olten, Switzerland), ~1 g of activated Florisil (60/100 mesh, Supelco, Bellefonte, USA), and an additional cellulose filter. Furthermore, ~6 g of previously freeze-dried, homogenized and weighed sediment mixed with 500 mg of hydromatrix (diatomaceous earth, Restek, Bellefonte, PA) to increase solvent channeling was added to the cell. Extraction of the cells was then carried out on an ASE 350 system (Dionex, Sunnyvale, U.S.A.) using ethyl acetate and acetone at a ratio of 70:30 (% v/v) as extraction solvents. Two static extraction cycles of 5 min at 80°C and a rinsing volume of 60% were implemented, each followed by 100 s of purging with N₂. Extracts were spiked with 60 µL of 2.0 mg/L of a mixed solution containing 98 internal standards with an absolute amount of 120 ng each. The extracts were then gently evaporated to approximately 1 mL at a temperature of 45°C with an automated evaporator system (Syncore® Polyvap from Büchi, Flawil, Switzerland). Additional evaporation with a gentle stream of N₂ at 40°C was performed to adjust the final volume to 500 µL. The final extracts were filtered into 2-mL auto sampler vials using 0.2-µm PTFE filters (BGB Analytics, Boeckten, Switzerland).

Detection of analytes was performed on an LC system connected to a Q Exactive™ Hybrid Quadrupole-Orbitrap Mass Spectrometer (Thermo Fisher Scientific, San Jose, U.S.A.) equipped with an electrospray ionization (ESI) source, as described by Chiaia-Hernandez et al., 2014. Data dependent was performed separately in the positive and negative ionization modes. Full range mass spectra were recorded over a mass range of 100 to 1100 *m/z* with a nominal resolving power of 140,000 referenced to *m/z* 400 and with a mass accuracy of ±5 ppm. High-resolution product ion spectra were acquired in MS/MS experiments with a nominal resolving power of 17,500.

The electrospray, source fragmentation and capillary voltage were set to 5 kV, 15 V and 25 V in positive ion mode, and to -4 kV, -15 V and -20 V in negative mode. The capillary temperature and tube lens were set at 300°C and 60 V in positive mode and 350°C and -70 V in negative mode. The sheath and auxiliary gas flow were set at 50 and 20 arbitrary units, respectively, for both ionization modes.

MS/MS spectra were compared with predicted MS/MS fragmentation using MetFrag (Wolf et al., 2010) in combination with ChemSpider (<http://www.chemspider.com/>) and PubChem (<https://pubchem.ncbi.nlm.nih.gov/>) databases to perform compound database searches and *in silico* fragmentation as described in detail elsewhere (Chiaia-Hernandez et al., 2014). In case standards were not available, the fragments obtained from each suspect were compared with spectral databases such as mzCloud (HighChem) and MassBank (Horai et al., 2010) and further evaluated depending on the matching fragments and predicted RT when possible.

Overall extraction recoveries and LODs were between 60 and 100% and < 10 ng/g for 95% of the target compounds for the used methods, respectively. For quantified compounds, matrix matched calibration curves were used by linear least-squares regression of the normalization of the analyte responses compared to the response of the internal standards. On average, ion suppression along the sediment core changed less than 11% from surface to bottom as shown in previous studies (Chiaia-Hernandez et al., 2013). For suspect screening, ion suppression was evaluated using >50 internal standards. Matrix and instrument blanks samples were used to account for contamination during the sample preparation and instrumental analysis. Sample duplicates, matrix and instrument blanks and check standards were randomly distributed within a sequence and account for more than 30% of the measured samples.

GC-MS/MS analysis

Sediment samples were extracted by pressurized liquid extraction (PLE) according to Pintado-Herrera et al. (2016), using an accelerated solvent extractor ASE 350 system from Dionex, (Sunnyvale, U.S.A.). A procedural blank was also performed in each extraction batch (12 cells). Purification of the extracts was performed simultaneously to the extraction (in-cell clean-up) by placing sorbents inside the cell (activated alumina 58 Å provided by Sigma-Aldrich, Madrid, Spain), whereas cellulose filters were used to separate the different layers. The target compounds were extracted from sediment aliquots (2 g dry weight, dispersed with 1.5 g of diatomaceous earth) using dichloromethane at 100 °C and 1500 psi, three static extraction cycles of 5 min each (purge time = 60 s, flush volume = 60%), 100 °C and 1500 psi. PLE extracts (30 mL) were evaporated to dryness (with a gentle stream of N₂) and re-dissolved in 500 µL of ethyl acetate, which was filtered through PTFE centrifuge filters (0.22 µm pore size, purchased from Ciromfg, Florida, U.S.A.). Finally, a derivatizing agent (10 µL of MTBSTFA from Sigma-Aldrich, Madrid, Spain) was also added to the final extract and was allowed to react with analytes at room

temperature for at least 30 min before injection to improve the signal and sensitivity of some target compounds with polar groups.

Gas chromatography (SCION 456-GC, Bruker) and mass spectrometry (SCION TQ from Bruker with CP 8400 Autosampler) were then used to identify and quantify target compounds.

Capillary gas chromatography analysis was carried out on a BR-5 ms column (30 m \times 0.25 mm i.d. \times 0.25 μ m film thickness), keeping the carrier gas flow (helium) at 1 mL min⁻¹, and the transfer line and the injection port temperatures at 280 °C. The column temperature ramp was as follows: 70 °C for 3.5 min, increased at 25 °C min⁻¹ to 180 °C, then at 10 °C min⁻¹ to 300 °C, and held for 4 min. Injection volume was 1 μ L in splitless mode and the solvent delay was set to 4 min.

The mass detector was operated in multiple reaction monitoring (MRM) mode using an electron ionization (EI) source set at 70 eV and argon as collision gas (2 mTorr). Calibration curves were constructed in ethyl acetate for each compound in the range of 5–500 ng mL⁻¹. All the data were processed using the Bruker MS Workstation 8 software.

Details of the detection methodology can be found in Pintado-Herrera et al. (Pintado-Herrera et al., 2016). Limits of detection (LOD) of the method, calculated using a signal-to-noise ratio of 3:1, and taking into account the background noise obtained for blank samples, ranged between <0.01 and 1.9 ng \cdot g⁻¹ for sediment samples.

Environmental Model

The substance behavior was assessed with a multi-compartment environmental fate model with the dimensions of Switzerland. The model was run in Microsoft Office Excel 2013. The input of the substance-specific input data as well as the collection of output data was automated using Microsoft Visual Basic for Applications (VBA).

First, a level-I model consisting of the compartments soil, water and air was applied assuming a closed system, equilibrium between compartments, and steady-state. Furthermore, a level-III model (“Small Region” Switzerland) model for sediment, soil, water and air was used assuming an open system, not at equilibrium, exchange between the different compartments but at steady state (Mackay, 2001). Parameters used in the model were the same as the level-I model with environmental parameters specific to Switzerland as described by Glüge et al., 2016. The level-III model was run with three different emission scenarios: (i) emission solely into air, (ii) emission solely into water and (iii) emission solely into soil. The emission rate was set at a

hypothetical 100 mol/h. In addition, the level-III model was used to identify chemicals that can reside >90% in sediment, with emission solely into the water compartment (the emission scenarios solely into air and soil, respectively, yield no chemicals that are >90% present in sediment). Furthermore, since specific input data (i.e. the partition coefficients octanol-water and air-water, K_{ow} , K_{aw} , and the degradation half-lives in air, water, soil and sediment, $t_{1/2,A}$, $t_{1/2,W}$, $t_{1/2,S}$, and $t_{1/2,X}$ had to be available to run a chemical in the level-III model, only complete input data sets were further evaluated. Therefore, 136 of the 9,187 chemicals (CAS numbers) in the Swiss chemical inventory (1.5%) were not considered further, since complete input data were not available (in most of these cases, no degradation half-life in air ($t_{1/2,A}$) was available). Specific parameters for the level-I and level-III model are provided in Table S4.

Table S4. Input parameters of the level-I and level-III models. Environmental parameters such as particle deposition velocity etc. were taken as listed by Mackay, 2001 Table 7.2.

Total surface area	4.13E+10 m ²
Fraction of area covered by soil	0.95
Depth of soil compartment	0.1 m
Density of soil solids	2500kg/m ³
Fraction of organic carbon in soil solids	0.04
Fraction of area covered by water	0.05
Depth of water compartment	30 m
Height of air compartment	1.00E+03 m

Table S5. Confirmed substances based on target analysis in soils using an in-house library (*Eawag-DB*). The samples were taken between 2005 and 2009 with the exception of site H (2) which dated back to 1994 and received domestic waste. More details about the site can be found in Table S1.

log K_{ow} and pK_a values were calculated by ChemAxon (Budapest, Hungary)

---- No dissociation

*Identification level was assigned according to Schymanski et al., (Schymanski et al., 2014).

No.	Compound Name	CAS No.	Application	log K_{ow}	pK_a	Location	Identification Level*
1	2-n-Octyl-4-isothiazolin-3-one (OIT)	26530-20-1	Pesticide	3.33	---	C	1
2	Bentazon	25057-89-0	Pesticide	0.76	2.03	B, A	1
3	Cyproconazole	94361-06-5	Pesticide	2.90	2.00/13.32	B	1
4	Cyprodinil	121552-61-2	Pesticide	3.21	3.10/13.63	B, C, H (1)	1
5	Desmedipham	13684-56-5	Pesticide	3.70	12.55	A	1
6	Dinoseb	88-85-7	Pesticide	3.24	5.08	H (1), A, B, K, M, J, I	1
7	Epoxiconazole	135319-73-2	Pesticide	3.74	2.00	A	1
8	Ethofumesate	26225-79-6	Pesticide	2.34	---	A	1
9	Fludioxonil	31341-86-1	Pesticide	3.57	14.66	B, C, M, A	1
10	Imidacloprid	138261-41-3	Pesticide	0.87	15.67	A	1
11	Ioxynil	1689-83-4	Pesticide	3.38	5.61	B	1
12	Isoproturon	34123-59-6	Pesticide	2.57	13.79	B	1
13	Metamitron	41394-05-2	Pesticide	0.44	2.78	A, B	1
14	Monolinuron	1746-81-2	Pesticide	2.08	12.12	A, B	1
15	Napropamide	15299-99-7	Pesticide	3.21	---	A, B	1
16	N-N-diethyl-3-methylbenzamid (DEET)	134-62-3	Personal care product	2.50	---	J, E	1
17	Oryzalin	19044-88-3	Pesticide	2.33	9.55	A, B, C, F	1
18	Phenmedipham	13684-63-4	Pesticide	3.86	12.55	A	1

No.	Compound Name	CAS No.	Application	log K _{ow}	pKa	Location	Identification Level*
19	Prometryn	7287-19-6	Pesticide	3.01	6.71/14.46	B	
20	Simazine	122-34-9	Pesticide	1.78	4.23/14.75	B	1
21	Tebutam	35256-85-0	Pesticide	3.71	- - -	A, C, H (2), I	1
22	Terbacil	5902-51-2	Pesticide	1.29	9.87	B	1
23	Terbutryn	886-50-0	Pesticide	2.88	6.72/14.31	B	1
24	Uniconazole	83657-22-1	Plant growth retardant	7.62	1.58/13.66	B	1

Table S6. Compounds detected in soils by gas chromatography tandem mass spectrometer (GC-MS/MS). The samples were taken between 2005 and 2009 with the exception of site H (2) which dated back to 1994 and received domestic waste. More details about the site can be found in Table S1.

*Location 97 was not part of the analysis due to the limited amount of soil available. OTNE (1-[(2R,3R)-2,3,8,8-Tetramethyl-1,2,3,4,5,6,7,8-octahydro-2-naphthalenyl]ethanone), EHS (Ethylhexyl Salicylate), TPP (Phenyl phosphate), TiBP (Triisobutyl phosphate), p,p'-DDD (Dichlorodiphenyldichloroethane), p,p'-DDE (dichlorodiphenyldichloroethylene).

Use	New location	Additives		Personal Care Products						Pesticides	
		TiBP µg/kg _{oc}	TPP µg/kg _{oc}	DEET µg/kg _{oc}	EHS µg/kg _{oc}	Galaxolide µg/kg _{oc}	Homosalate µg/kg _{oc}	OTNE µg/kg _{oc}	Tonalide µg/kg _{oc}	p,p'-DDD µg/kg _{oc}	p,p'-DDE µg/kg _{oc}
Agriculture	A	15	2	8	440	ND	ND	22	18	ND	68
Agriculture	B	45	ND	ND	1459	17	ND	9	ND	99	548
City Park	C	9	ND	ND	504	ND	30	ND	ND	ND	ND
Coniferous Forest	D	ND	ND	ND	136	ND	16	ND	ND	ND	1
Coniferous Forest	E	2	ND	12	79	ND	21	ND	ND	ND	ND
Deciduous Forest	F	ND	ND	ND	79	ND	8	ND	ND	ND	4
Deciduous Forest	G	ND	ND	ND	845	ND	39	ND	ND	ND	ND

Use	New location	Additives		Personal Care Products						Pesticides	
		TiBP μg/kg _{oc}	TPP μg/kg _{oc}	DEET μg/kg _{oc}	EHS μg/kg _{oc}	Galaxolide μg/kg _{oc}	Homosalate μg/kg _{oc}	OTNE μg/kg _{oc}	Tonalide μg/kg _{oc}	p,p'-DDD μg/kg _{oc}	p,p'-DDE μg/kg _{oc}
Grasslands , low intensity	H-1	255	15	1	511	7	47	25	37	ND	ND
Grasslands , low intensity	H-2	7	ND	2	518	4	47	ND	ND	ND	ND
Grasslands , low intensity	I	ND	ND	ND	118	ND	ND	ND	ND	ND	ND
Mixed forest	J	1	ND	ND	315	ND	55	ND	ND	ND	19
Protection site	K	7	ND	ND	361	ND	53	ND	ND	ND	ND
Protection site	L	ND	ND	ND	51	ND	10	ND	ND	ND	ND
Formula		C ₁₂ H ₂₇ O ₄ P	C ₁₈ H ₁₅ O ₄ P	C ₁₂ H ₁₇ NO	C ₁₅ H ₂₂ O ₃	C ₁₈ H ₂₆ O	C ₁₆ H ₂₂ O ₃	C ₁₆ H ₂₆ O	C ₁₈ H ₂₆ O	C ₁₄ H ₁₀ Cl ₄	C ₁₄ H ₈ Cl ₄
CAS No.		126-71-6	115-86-6	134-62-3	118-60-5	1222-05-5	118-56-9	54464-57-2	1506-02-1/21145-77-7	72-54-8	72-55-9

Table S7. Confirmed compounds based on target analysis in sediments of Greifensee using an in-house library (*Eawag-DB*). Sampling point 1 (P1) was collected from the deepest part of the lake. Sampling points P2 and P3 were collected near the outlet of the Uster WWTP and Mönchaltorfer WWTP at Aaspitz (to cover the impact of WWTP effluents and agricultural inputs to the catchment). Location and details of sediment cores can be found in Figure S2 and S3 and Table S3.

log K_{ow} and pK_a values were calculated by ChemAxon (Budapest, Hungary)

---- No dissociation

*Identification level was assigned according to Schymaski et al., (Schymanski et al., 2014).

No.	Compound Name	Formula	CAS No.	Application	Log K _{ow}	pKa	Location	Identification Level*
1	5-Methylbenzotriazole	C ₇ H ₇ N ₃	136-85-6	Industrial chemical	1.78	0.45/9.12	P2	1
2	Aldicarb	C ₇ H ₁₄ N ₂ O ₂ S	116-06-3	Pesticide	1.28	1.63/14.33	P3	1
3	Atraton	C ₉ H ₁₇ N ₅ O	1610-17-9	Pesticide	1.81	6.0/14.44	P3	1

No.	Compound Name	Formula	CAS No.	Application	Log K _{ow}	pKa	Location	Identification Level*
4	Atrazine	C ₈ H ₁₄ ClN ₅	1912-24-9	Pesticide	2.20	4.20/14.48	P2, P3	1
5	Benzotriazole	C ₆ H ₅ N ₃	95-14-7	Industrial chemical	1.3	0.22/9.04	P1	1
6	Carbamazepine	C ₁₅ H ₁₂ N ₂ O	298-46-4	pharmaceutical	2.77	15.96	P2	1
7	Climbazole	C ₁₅ H ₁₇ ClN ₂ O ₂	38083-17-9	Pesticide	4.34	6.49	P2, P3	1
8	Cyproconazole	C ₁₅ H ₁₈ ClN ₃ O	94361-06-5	Pesticide	2.90	2.0/13.32	P3	1
9	Cyprodinil	C ₁₄ H ₁₅ N ₃	121552-61-2	Pesticide	3.21	3.10/13.63	P3	1
10	DEET (N-N-diethyl-3-methylbenzamid)	C ₁₂ H ₁₇ NO	134-62-3	Personal care product	2.50	- - -	P3	1
11	Diazepam	C ₁₆ H ₁₃ ClN ₂ O	439-14-5	Pharmaceutical	3.08	2.92	P2	1
12	Diazinon	C ₁₂ H ₂₁ N ₂ O ₃ PS	333-41-5	Pesticide	4.19	4.19	P3	1
13	Difenoconazole	C ₁₉ H ₁₇ Cl ₂ N ₃ O ₃	119446-68-3	Pesticide	4.86	1.95	P3, P2	1
14	Diflufenican	C ₁₉ H ₁₁ F ₅ N ₂ O ₂	83164-33-4	Pesticide	5.11	1.60/13.28	P3, P2	1
15	Dinoseb	C ₁₀ H ₁₂ N ₂ O ₅	88-85-7	Pesticide	3.24	5.08	P3	1
16	Diuron	C ₉ H ₁₀ Cl ₂ N ₂ O	330-54-1	Pesticide	2.53	13.18	P2, P3	1
17	Epoxyconazole	C ₁₇ H ₁₃ ClFN ₃ O	133855-98-8	Pesticide	3.74	2.00	P3	1
18	Fenhexamid	C ₁₄ H ₁₇ Cl ₂ N ₂ O ₂	126833-17-8	Pesticide	4.78	7.4	P3	1
19	Fenofibrate	C ₂₀ H ₂₁ ClO ₄	49562-28-9	Pharmaceutical	5.28	- - -	P2	1
20	Fenpropimorph	C ₂₀ H ₃₃ NO	67306-03-0	Pesticide	5.17	8.49	P3	1
21	Fipronil-sulfide	C ₁₂ H ₄ Cl ₂ F ₆ N ₄ S	120067-83-6	Transformation product	5.62	0.36	P3	1
22	Fludioxonil	C ₁₂ H ₆ F ₂ N ₂ O ₂	131341-86-1	Pesticide	3.57	14.66	P2, P3	1
23	Flufenamic acid	C ₁₄ H ₁₀ F ₃ N ₂ O ₂	530-78-9	Pharmaceutical	5.25	3.88	P1	1
24	Flusilazole	C ₁₆ H ₁₅ F ₂ N ₃ Si	85509-19-9	Pesticide	4.68	2.32	P3	1

No.	Compound Name	Formula	CAS No.	Application	Log K _{ow}	pKa	Location	Identification Level*
25	Galaxolidone	C18H24O2	507442-49-1	Personal care product	4.65	- - -	P1	1
26	Irgarol	C11H19N5S1	28159-98-0	Pesticide	2.99	6.68/14.13	P1, P2, P3	1
27	Irgarol-descyclopropyl	C8H15N5S	30125-65-6	Transformation product	2.22	6.87/14.36	P3	1
28	Isoproturon	C12H18N2O	34123-59-6	Pesticide	2.57	13.79	P3	1
29	Lidocaine	C14H22N2O	137-58-6	pharmaceutical	2.84	7.75/13.78	P2	1
30	Lufenuron	C17H8Cl2F8N2O3	103055-07-8	Pesticide	6.12	9.02	P1	1
31	Methylbenzotriazole	C7H7N3	29385-43-1	Corrosion Inhibitor	1.78	0.47/9.29	P3, P1	1
32	Metribuzin	C8H14N4OS	21087-64-9	Pesticide	1.96	2.46	P3	1
33	Diuron-desmethyl	C8H8Cl2N2O	3567-62-2	Pesticide	2.31	13.31	P3	1
34	Octocrylene	C24H27NO2	6197-30-4	Personal care product	6.78	- - -	P1	1
35	OIT (2-Octyl-2H-isothiazol-3-one)	C11H19NOS	26530-20-1	Pesticide	3.33	- - -	P3	1
36	Oryzalin	C12H18N4O6S	19044-88-3	pesticide	2.33	9.55	P3	1
37	Pirimicarb	C11H18N4O2	23103-98-2	Pesticide	1.8	5.00	P3	1
38	Prometon	C10H19N5O	1610-18-0	Pesticide	2.23	5.97/14.27	P3	1
39	Prometryn	C10H19N5S	7287-19-6	Pesticide	3.01	6.71/14.46	P1, P3	1
40	Propiconazole	C15H17Cl2N3O2	60207-90-1	Pesticide	4.33	1.95	P1, P2, P3	1
41	Prosulfocarb	C14H21NOS	52888-80-9	Pesticide	4.17	- - -	P3	1
42	Pyrimethanil	C12H13N3	53112-28-0	Pesticide	2.43	3.44/13.67	P3	1
43	Simazin	C7H12ClN5	122-34-9	Pesticide	1.78	4.23/14.75	P3	1
44	Tebuconazole*	C16H22ClN3O	107534-96-3	Pesticide	3.69	2.01/13.85	P3	1
45	Tebutam	C15H23NO	35256-85-0	Pesticide	3.71	- - -	P3	1
46	Terbumeton	C10H19N5O	33693-04-8	Pesticide	2.09	5.97/14.13	P3	1

No.	Compound Name	Formula	CAS No.	Application	Log K _{ow}	pKa	Location	Identification Level*
47	Terbutylazine	C ₉ H ₁₆ ClN ₅	5915-41-3	Pesticide	2.48	4.18/14.77	P2, P3	1
48	Terbutryn	C ₁₀ H ₁₉ N ₅ S	886-50-0	Pesticide	2.88	6.72/14.31	P1, P3	1
49	Terbutylazin-desethyl*	C ₇ H ₁₂ ClN ₅	30125-63-4	Pesticide	1.82	4.35/14.22	P3	1
50	Tonalide	C ₁₈ H ₂₆ O	21145-77-7	Personal care product	4.96	- - -	P1, P2, P3	1
51	Triclocarban	C ₁₃ H ₉ Cl ₃ N ₂ O	101-20-2	Personal care product	4.93	11.42	P1, P2	1
52	Triclosan	C ₁₂ H ₇ Cl ₃ O ₂	3380-34-5	Antimicrobial	4.98	7.68	P1, P2, P3	1

Table S8. Compounds detected in sediment samples from Greifensee by GC-MS/MS. Sampling point 1 (P1) was collected from the deepest part of the lake. Sampling points P2 and P3 were collected near the outlet of the Uster WWTP and Mönchaltorfer WWTP at Aaspitz (to cover the impact of WWTP effluents and agricultural inputs to the catchment). Location and details of sediment cores can be found in Figure S2 and S3 and Table S3.

*Identification level was assigned according to Schymanski et al., (Schymanski et al., 2014).

No.	Compound Name	Acronym	CAS No.	Formula	Use	Location	Identification Level*
1	1-[(2R,3R)-2,3,8,8-Tetramethyl-1,2,3,4,5,6,7,8-octahydro-2-naphthalenyl]ethanone	OTNE	54464-57-2	C ₁₆ H ₂₆ O	Personal care product	P1, P2, P3	1
2	2-Ethyl hexyl diphenyl phosphate	EHDPP	1241-94-7	C ₂₀ H ₂₇ O ₄ P	Additives	P1, P2, P3	1
3	2-ethyl methoxycinnamate	EHMC	5466-77-3	C ₁₈ H ₂₆ O ₃	Personal care product	P2, P3	1
4	2-Ethylhexyl phosphate	TEHP	78-42-2	C ₂₄ H ₅₁ O ₄ P	Additives	P1, P2, P3	1
5	Celestolide	ADBI	13171-00-1	C ₁₇ H ₂₄ O	Personal care product	P1, P2, P3	1
6	Chlorpyrifos		2921-88-2	C ₉ H ₁₁ Cl ₃ NO ₃ PS	Pesticide	P2, P3	1
7	Dichlorodiphenyldichloroethane	p,p'-DDD	72-54-8	C ₁₄ H ₁₀ Cl ₄	Pesticide	P1, P2, P3	1
8	Dichlorodiphenyldichloroethylene	p,p'-DDE	72-55-9	C ₁₄ H ₈ Cl ₄	Pesticide	P1, P2, P3	1

No.	Compound Name	Acronym	CAS No.	Formula	Use	Location	Identification Level*
9	N-N-diethyl-3-methylbenzamid	DEET	134-62-3	C ₁₂ H ₁₇ NO	Personal care product	P2, P3	1
10	Ethyl hexyl salycilate	EHS	118-60-5	C ₁₅ H ₂₂ O ₃	Personal care product	P3	1
11	Galaxolide	HHCB	1222-05-5	C ₁₈ H ₂₆ O	Personal care product	P1, P2, P3	1
12	Homosalate	HMS	118-56-9	C ₁₆ H ₂₂ O ₃	Personal care product	P1, P2, P3	1
13	Octocrylene	OC	6197-30-4	C ₂₄ H ₂₇ NO ₂	Personal care product	P1, P2, P3	1
14	Permetrin	cis-isomer	52645-53-1	C ₂₁ H ₂₀ Cl ₂ O ₃	Pesticide	P2, P3	1
	Permetrin	trans-isomer	52645-53-1	C ₂₁ H ₂₀ Cl ₂ O ₃	Pesticide	P2, P3	1
15	Terbutryn		886-50-0	C ₁₀ H ₁₉ N ₅ S	Pesticide	P1, P2, P3	1
16	Tonalide	AHTN	21145-77-7	C ₁₈ H ₂₆ O	Personal care product	P1, P2, P3	1
17	Traseolide	ATII	68140-48-7	C ₁₈ H ₂₆ O	Personal care product	P1, P2, P3	1
18	Triisobutylphosphate	TiBP	126-71-6	C ₁₂ H ₂₇ O ₄ P	Additives	P2, P3	1
19	Triphenylphosphate	TPP	115-86-6	C ₁₈ H ₁₅ O ₄ P	Additives	P1, P2, P3	1
20	Tris-n-butyl phosphate	TNBP	126-73-8	C ₁₂ H ₂₇ O ₄ P	Additives	P1, P2, P3	1

Environmental fate model- Substances in Soil

According to the level-I “Unit World” model of Switzerland, 2,550 of the 9,187 (28% of entire Swiss DB) modelled CAS numbers are predicted to be >90% distributed in soil as shown in Figure S4.

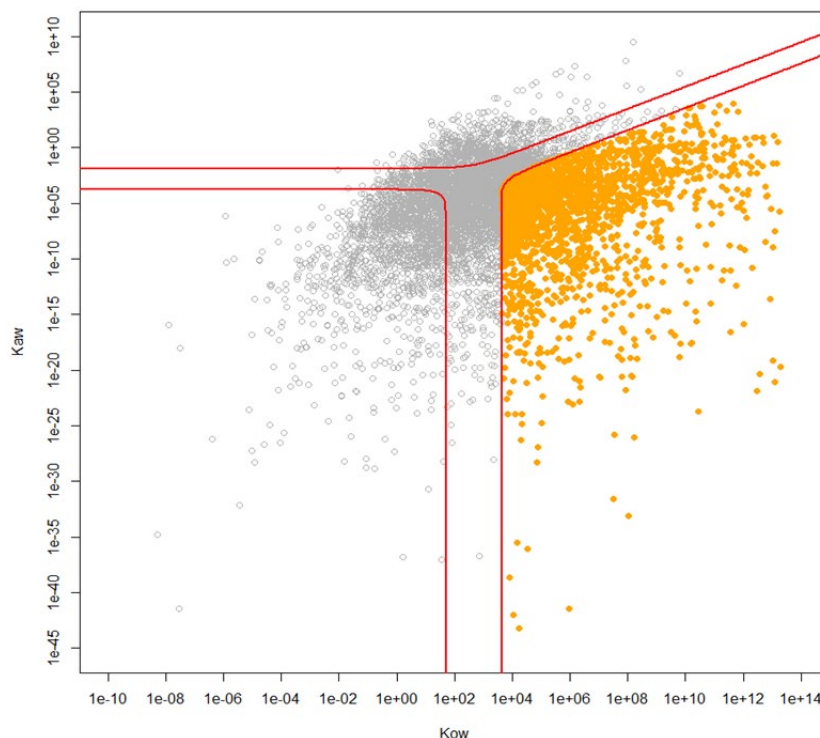


Figure S4. Chemical space of 9,187 CAS numbers from the Swiss DB (grey dots) with 2,550 CAS numbers (orange dots) indicated to be >90% distributed in soil by the level-I model. Red lines show ratio 0.90:0.10 between the different compartments (top: 90% in air, bottom left: 90% in water, bottom right: 90% in soil).

According to the level-III model of Switzerland, 2,190 (24%) modelled chemicals of the entire Swiss DB are indicated to be >90% distributed in soil when an emission solely into the air compartment is used as shown in Figure S5. Emission to air implies that the chemicals have to survive deposition from air to soil before they can build up a reservoir in the soil. In contrast, when emission solely into the soil compartment is used, 5,758 (64%) modelled CAS numbers of entire Swiss DB are found to be >90% distributed in soil, which is too many substances for the following steps. Therefore, the more restrictive filtering with emission to air was used to identify chemicals with a high potential for accumulation in the soil.

The outcome of the level-I model was directly compared to the level-III model results with emission solely into air. The comparison of the level-I and level-III model results showed 1,379

CAS numbers from the Swiss DB (15%) that were indicated by both models to be >90% present in soil. In addition, 1,171 CAS numbers (13%) and 811 CAS numbers (9%) from the Swiss DB were shown to reside >90% in soil solely in the level-I and level-III model, respectively.

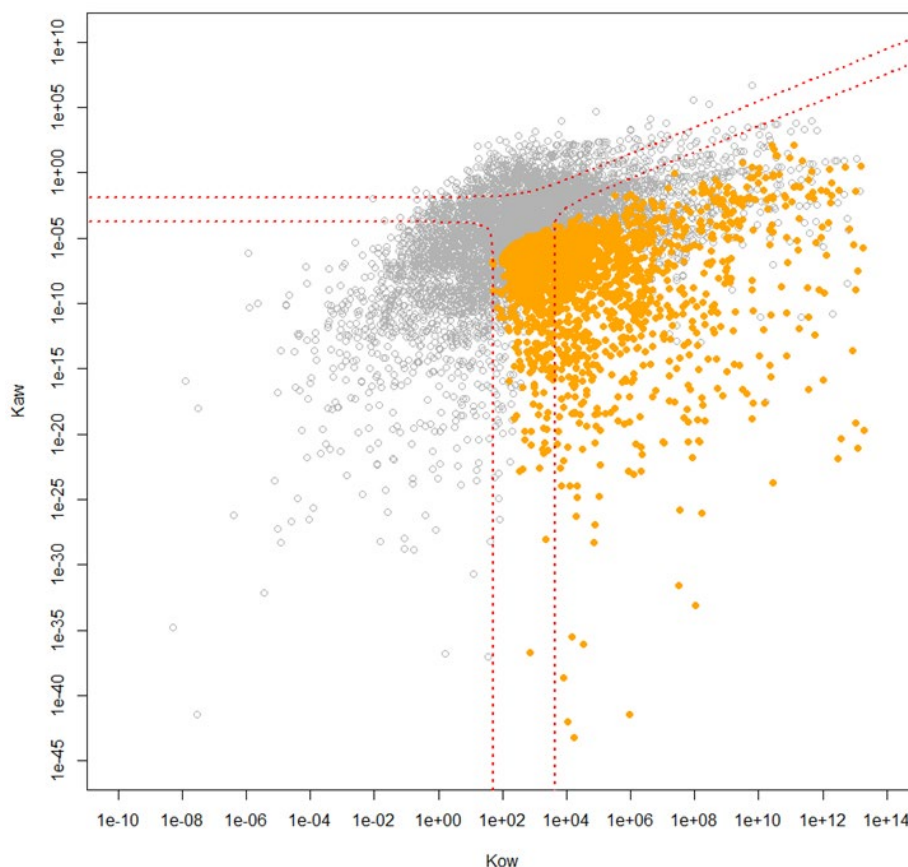


Figure S5. Chemical space of 9,187 CAS numbers from the Swiss DB (grey dots) with 2,190 CAS numbers (orange dots) predicted to be >90% distributed in soil by the Level III model (811 predicted CAS numbers are unique to Level 3). Red lines show equilibrium lines indicate the ratio of 0.90:0.10 between the different compartments (top: 90% in air, bottom left: 90% in water, bottom right: 90% in soil).

Environmental fate model- Substances in Sediment

The level-III model was also used to identify the CAS numbers that will reside >90% in sediment after emission solely into the water compartment. Because of the high number of substances found to be present in soil when the full Swiss DB was analyzed, as described above, only substances containing halogens were selected for this analysis. Accordingly, from the 9,187 CAS numbers in the Swiss DB, only 2,371 CAS numbers were run through the level-III model, with

emissions solely into water. The results show that 188 of the 2,371 Swiss CAS numbers (8%) were found to be >90% distributed in sediment as shown in Figure S6.

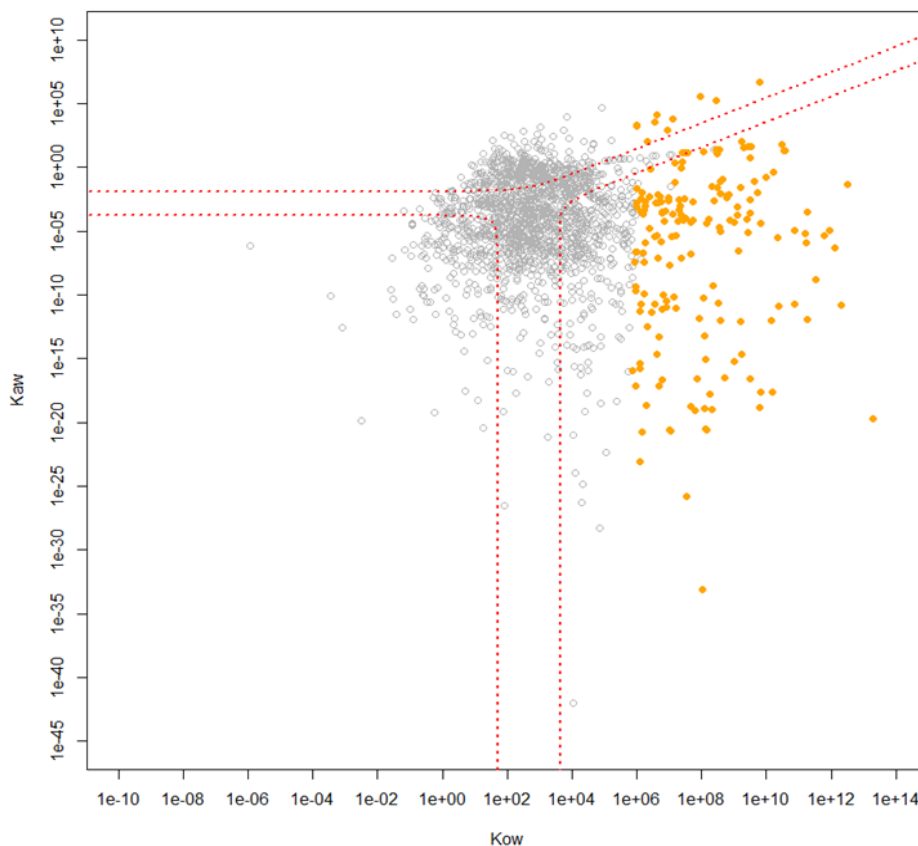


Figure S6. Chemical Space of 2,371 Swiss CAS halogenated CAS numbers from the Swiss DB. Orange dots illustrate 188 substances (CAS numbers) found to be >90% in sediment with the level-III model, emissions to water. Red dotted lines show equilibrium lines indicate the ratio of 0.90:0.10 between the different compartments according to the level-I model between the different compartments (top: 90% in air, bottom left: 90% in water, bottom right: 90% in sediment).

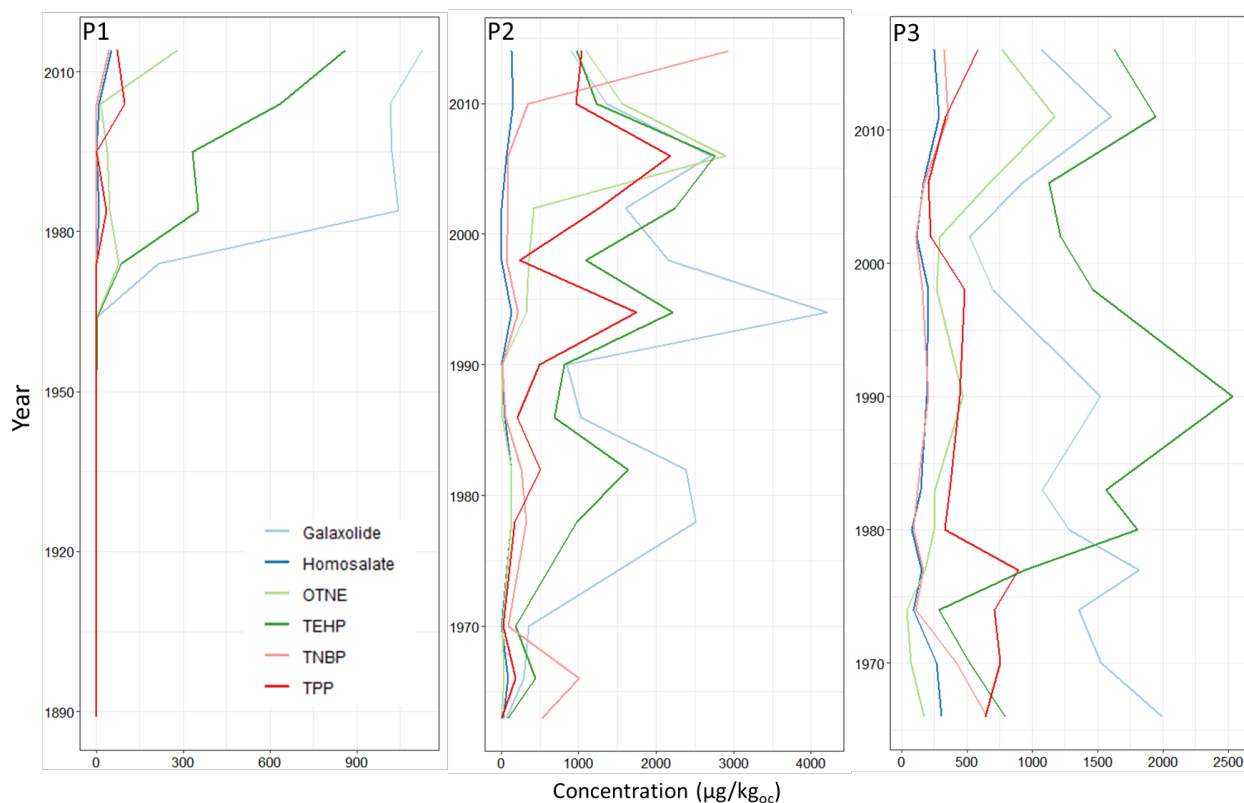


Figure S7. Temporal concentration trends of selected personal care products (galaxolide, homosalate and OTNE) and organosphosphate esters (2-ethylhexyl phosphate (TEHP), tris-n-butyl phosphate (TNBP), and phenyl phosphate (TPP)) analyzed by GC-MS/MS. Increasing concentrations over time are observed for TNBP (P1, P2 and P3), TEHP (P1), TPP (P3), OTNE (P1), and homosalate (P1, P2 and P3).

Screening Organic Contaminants Based on Filtered Data

The output of the environmental fate model was further filtered by retrieving the molecular formula and structure from the SMILES codes and CAS numbers using different software and databases such as Chemaxon, ChemSpider and PubChem (JChem, 2016; RSC). The output list was reduced by removing compounds with salt adducts and already present in the output list, substances with the same structure but different CAS numbers, substances not likely to be ionized by ESI using rules (presence or absence of functional groups) as described by Moschet et al., 2014, substances with known low ionization efficiency (e.g. pyrethroids), and structural isomers and enantiomers. Retention times (RT) for suspected compounds were predicted using a linear correlation between measured retention time and $\log K_{ow}$ for 164 references standards as described elsewhere ($Rt \text{ (min)} = 1.65 \cdot \log K_{ow} + 4.36$) (Chiaia-Hernandez et al., 2013) with an error window of $\pm 4 \text{ min.}$ Workflows are illustrated in Figure S8 and S9.

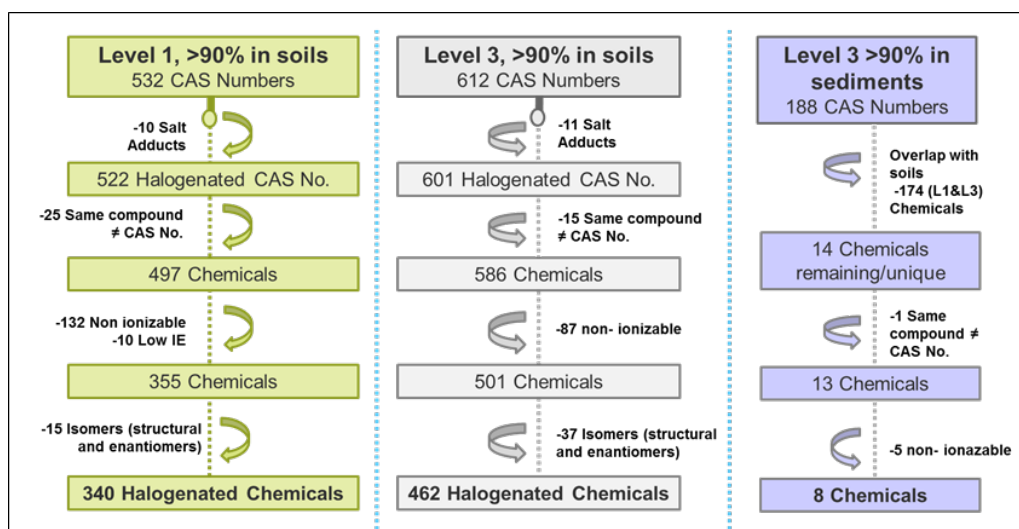


Figure S8. Organization and filtering of compounds obtained from the multimedia fate model. The filter steps led to a set of 340 CAS numbers for chemicals in soil from the level-I model and another set of 462 CAS numbers for chemicals in soil from the level-III model, with an overlap of 289 CAS numbers. In addition, there were 8 CAS numbers for chemicals in sediment that were not yet in any of the two soil sets.

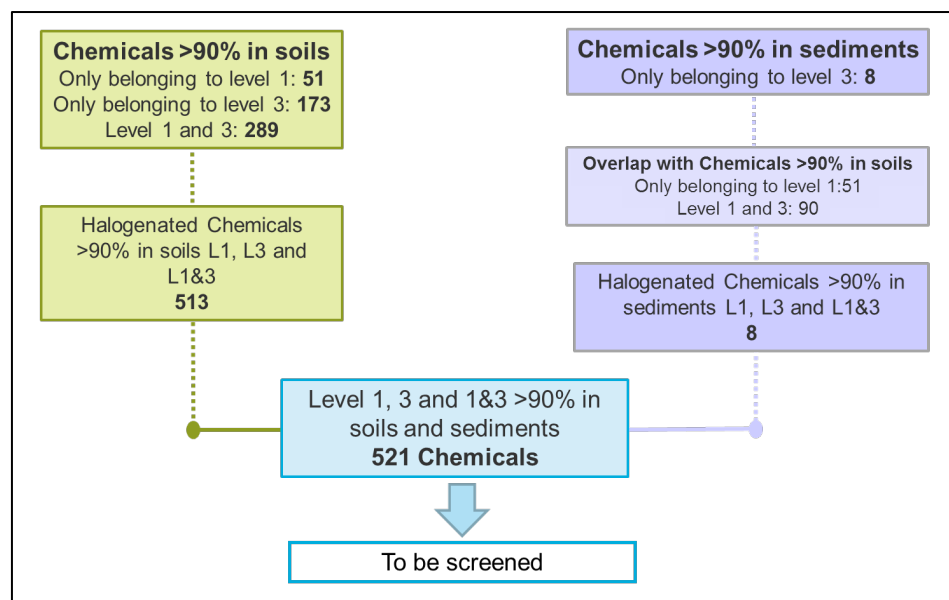


Figure S9. Filtering and compilation of 521 chemicals used as suspect candidates (289 in soil, level-I and level-III model; 51 in soil, level-I model only; 173 in soil, level-III model only; 8 in sediment, level-III model) to detect further organic contaminants in soils and sediments. Details of the 521 screened chemicals can be found at the Appendix B file.

Table S9. Confirmed substances of halogenated substances (Swiss HDB) from the Swiss chemical inventory with fractions of 0.9 or above in soils. The samples were taken between 2005 and 2009 with the exception of site H (2) which dated back to 1994 and received domestic waste. More details about the site can be found in Table S1. Highlighted substances overlap with detectable compounds using an in-house library (Eawag DB).

log K_{ow} and pK_a values were calculated by ChemAxon (Budapest, Hungary)

---- No dissociation

*Identification level was assigned according to Schymanski et al., (Schymanski et al., 2014).

No.	Compound Name	CAS No.	Application	Location	log K _{ow}	pKa	Identification Level
1	2-Chlorobenzophenone	5162-03-8, 134-85-0	Various sources	H (1), A	4.04	---	1
2	Atrazine	1912-24-9	Pesticide	H (1), B, C, A	2.20	4.20/14.48	1
3	Bromochlorophene	15435-29-7	Antimicrobial	M	6.20	5.86	1
4	Difeconazole	119446-68-3	Pesticide	B	4.86	1.95	1
5	Diflufenican	83164-33-4	Pesticide	B	5.11	1.60/13.28	1
6	Dimefuron	34205-21-5	Pesticide	B	3.44	12.9	1
7	Diuron	330-54-1	Pesticide	B	2.53	13.18	1
8	Diuron-desmethyl	3567-62-2	Pesticide	B	2.31	13.31	1
9	Fluazinam	79622-59-6	Pesticide	C	6.93	6.69	1
10	Flucifuron	370-50-3	Pesticide	H (1), H (2), B, C, M, A	6.08	11.38	1
11	Fludioxonil	31341-86-1	Pesticide	B, C, M, A	3.57	14.66	1
12	Hexachlorophene	70-30-4	Antimicrobial	H (1), H (2), B, C, M, H, D	7.08	5.15	1
13	PFOS	1763-23-1	Industrial chemical	B, A, C, M, E, D, F, H (2), I, J, K, L, H (1)	5.43	---	1
14	Propiconazole	60207-90-1	Pesticide	B, C, M	4.33	1.95	1

No.	Compound Name	CAS No.	Application	Location	log K _{ow}	pKa	Identification Level
15	Triclocarban	101-20-2	Antimicrobial	H (1), H (2), B, C, M, A, I	4.98	11.42	1
16	Triclosan	3380-34-5	Antimicrobial	H (1), H (2), B, C, M, A, I	5	7.68	1

Table S10. Suspect Screening Analysis of halogenated substances (Swiss-HDB) from the Swiss chemical inventory in soils with fractions of 0.9 or above in soils and with level of identification > 3. The samples were taken between 2005 and 2009 with the exception of site H (2) which dated back to 1994 and received domestic waste. More details about the site can be found in Table S1.

¹ Smiles obtained from Strempe et al., (Strempe et al., 2012).

*Identification level was assigned according to Schymanski et al., (Schymanski et al., 2014).

No.	Compound Name	CAS No.	Location	Formula	Smiles ¹	Δ m/z (ppm)	Δ RT (min)	Isotopic Pattern Match (%)	Ionization	Identification Level *	Fragments predicted found
1	2-(chloromethyl)-4-nitrophenol	2973-19-5	A, B, C, M, I	C ₇ H ₆ ClNO ₃	<chem>ClCc1cc(ccc1O)N(=O)=O</chem>	1.4	0.9	94	negative	2	2
3	Dichlorophenol	576-24-9, 95-77-2, 591-35-5, 583-78-8	B	C ₆ H ₄ Cl ₂ O	<chem>Clc1c(O)cccc1Cl</chem>	0.4	0.3	100	negative	2	3
4	Niclofolan	10331-57-4	K	C ₁₂ H ₆ Cl ₂ N ₂ O ₆	<chem>Clc1cc(c2cc(Cl)cc(N(=O)=O)c2O)c(O)c(c1)N(=O)=O</chem>	0.4	-0.5	98	negative	2	2

No.	Compound Name	CAS No.	Location	Formula	Smiles ¹	Δ m/z (ppm)	Δ RT (min)	Isotopic Pattern Match (%)	Ionization	Identification Level *	Fragments predicted found
2	3,4,6-trichloro-2-nitrophenol	82-62-2	H (1, 2), A, B, C, K, M, I	C6H2Cl3NO3	<chem>O=N(=O)c1c(O)c(Cl)cc(Cl)c1Cl</chem>	0.5	1.1	71	negative	3	4
5	Clomestron	4091-75-2	I, F, H (1)	C19H23ClO2	<chem>COc1ccc2c(CCC3C2CC2(C)C3CC(Cl)C2=O)c1</chem>	2.1	-0.4	57	negative	3	1
6	2,3,4,6-Tetrachlorophenol	58-90-2, 25167-83-3, 4901-51-3, 935-95-5	H (1), A, B, I	C6H2Cl4O	<chem>Oc1c(Cl)cc(Cl)c(Cl)c1Cl</chem>	-0.2	1.9	100	negative	3	1
7	Pentachlorophenol	87-86-5	H (1, 2), A, B, C, M, I, G	C6HCl5O	<chem>Clc1c(O)c(Cl)c(Cl)c(Cl)c1Cl</chem>	1.0	3.0	100	negative	3	1
8	2,4,6-tribromophenol	118-79-6	H (1)	C6H3Br3O	<chem>Brcc1cc(Br)c(O)c(Br)c1</chem>	0.6	1.7	100	negative	3	1
9	2,6-dibromobenzoic acid	601-84-3	H (1, 2), A, B, C, M, J, I, G	C7H4Br2O2	<chem>OC(=O)c1c(Br)cccc1Br</chem>	0.2	-1.9	100	negative	3	1
10	3,5-Dibromosalicylaldehyde	90-59-5	A, B, M, H (1, 2), I, G, I, G, C, J, I	C7H4Br2O2	<chem>O=Cc1cc(Br)cc(Br)c1O</chem>	0.2	-2.2	100	negative	3	1
11	4-chloro-1-(4-methoxyphenyl)butan-1-one	40877-19-8	G	C11H13ClO2	<chem>ClCCCC(=O)c1ccc(OC)cc1</chem>	0.2	2.6	94	negative	3	1

No.	Compound Name	CAS No.	Location	Formula	Smiles ¹	Δ m/z (ppm)	Δ RT (min)	Isotopic Pattern Match (%)	Ionization	Identification Level *	Fragments predicted found
12	2,6-dibromo-4-nitrophenol	99-28-5	B, C	C ₆ H ₃ Br ₂ NO ₃	<chem>BrC1cc(cc(Br)c1O)N(=O)=O</chem>	1.1	0.7	100	negative	3	2

Table S11. Confirmed halogenated substances (Swiss HDB) from the Swiss chemical inventory with fractions of 0.9 or above in sediments of Greifensee. Sampling point 1 (P1) was collected from the deepest part of the lake. Sampling points P2 and P3 were collected near the outlet of the Uster WWTP and Mönchaltorfer WWTP at Aaspitz (to cover the impact of WWTP effluents and agricultural inputs to the catchment). Location and details of sediment cores can be found in Figure S2 and S3 and Table S3. Highlighted substances overlap with detectable compounds using an in-house library (Eawag DB).

log K_{ow} and pK_a values were calculated by ChemAxon (Budapest, Hungary)

---- No dissociation

*Identification level was assigned according to Schymanski et al., (Schymanski et al., 2014).

No.	Compound Name	CAS No.	Application	Formula	Location	log K _{ow}	pK _a	Identification Level*
1	Atrazine	1912-24-9	Pesticide	C ₈ H ₁₄ ClN ₅	P2, P3	2.20	4.20/14.48	1
2	Bromochlorophene	15435-29-7	Antimicrobial	C ₁₃ H ₈ Br ₂ Cl ₂ O ₂	P1, P2	6.20	5.86	1
3	Chlorobenzhydrol	16071-26-4	Various	C ₁₃ H ₁₁ ClO	P2	3.59	13.71	1
4	Chlorophene	120-32-1	Pesticide/Pharma	C ₁₃ H ₁₁ ClO	P1, P2	9.06	4.37	1
5	Chlorpyrifos	2921-88-2	Pesticide	C ₉ H ₁₁ Cl ₃ NO ₃ PS	P2	4.78	---	1
6	Chlortoluron	15545-48-9	Pesticide	C ₁₀ H ₁₃ ClN ₂ O	P3	2.44	13.53	1
7	Climbazole	38083-17-9	pesticide	C ₁₅ H ₁₇ ClN ₂ O ₂	P2, P3	4.34	6.49	1
8	Dichlorophene	97-23-4	Pesticide/Antimicrobial	C ₁₃ H ₁₀ Cl ₂ O ₂	P1	4.67	7.22	1

No.	Compound Name	CAS No.	Application	Formula	Location	log K _{ow}	pKa	Identification Level*
9	Difenoconazole	119446-68-3	Pesticide	C ₁₉ H ₁₇ Cl ₂ N ₃ O ₃	P3, P2	4.86	1.95	1
10	Diuron	330-54-1	Pesticide	C ₉ H ₁₀ Cl ₂ N ₂ O	P2, P3	2.53	13.18	1
11	Diuron-desmethyl	3567-62-2	pesticide	C ₈ H ₈ Cl ₂ N ₂ O	P4	2.31	13.31	1
12	Fluazinam	79622-59-6	Pesticide	C ₁₃ H ₄ Cl ₂ F ₆ N ₄ O ₄	P2	6.93	6.69	1
13	Flucofuron	370-50-3	Pesticide	C ₁₅ H ₈ Cl ₂ F ₆ N ₂ O	P1, P2	6.08	11.38	1
14	Fludioxonil	131341-86-1	Pesticide	C ₁₂ H ₆ F ₂ N ₂ O ₂	P2, P3	3.57	14.66	1
15	Hexachlorophene	70-30-4	Antimicrobial	C ₁₃ H ₆ Cl ₆ O ₂	P1, P2	7.08	5.15	1
16	Mandipropamid	374726-62-2	Pesticide	C ₂₃ H ₂₂ ClNO ₄	P3	3.71	14.47	1
17	Penconazole	66246-88-6	Pesticide	C ₁₃ H ₁₅ Cl ₂ N ₃	P2, P3	4.19	2.06	1
18	Pencycuron	66063-05-6	Pesticide	C ₁₉ H ₂₁ ClN ₂ O	P2	5.01	13.37	1
19	Phosalone	2310-17-0	Pesticide	C ₁₂ H ₁₅ ClNO ₄ PS ₂	P2	3.75	---	1
20	Prochloraz	67747-09-5	Pesticide	C ₁₅ H ₁₆ Cl ₃ N ₃ O ₂	P1, P2, P3	3.62	2.55	1
21	Propazine	139-40-3	Pesticide	C ₉ H ₁₆ ClN ₅	P2, P3	2.61	4.17/14.31	1
22	Propiconazole	60207-90-1	Pesticide	C ₁₅ H ₁₇ Cl ₂ N ₃ O ₂	P1, P2, P3	4.33	1.95	1
23	Terbutylazine	5915-41-3	Pesticide	C ₉ H ₁₆ ClN ₅	P2, P3	2.48	4.18/14.77	1
24	Triclocarban	101-20-2	personal care product	C ₁₃ H ₉ Cl ₃ N ₂ O	P1, P2	4.93	11.42	1
25	Triclosan	3380-34-5	Antimicrobial	C ₁₂ H ₇ Cl ₃ O ₂	P1, P2, P3	4.98	7.68	1

Table S12. Suspect Screening Analysis of halogenated substances (Swiss HDB) from the Swiss chemical inventory with fractions of 0.9 or above and with level of identification > 3 in sediments of Greifensee. Location and details of sediment cores can be found in Figure S2 and S3 and Table S3.

log K_{ow} and pK_a values were calculated by ChemAxon (Budapest, Hungary)

---- No dissociation

¹ Smiles obtained from Strempe et al., (Strempe et al., 2012).

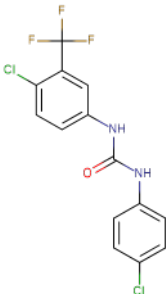
*Identification level was assigned according to Schymanski et al., (Schymanski et al., 2014).

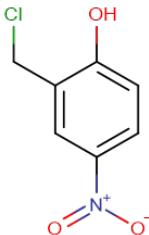
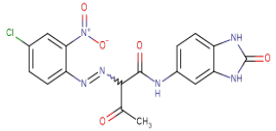
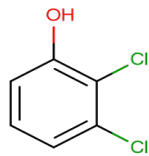
No	Compound Name	CAS No.	Formula	Smiles ¹	Δ m/z (ppm)	Δ Rt (min)	Ionization	Isotopic Pattern Match (%)	Identification Level *	Fragments predicted found
1	2-[2-(4-chloro-2-nitrophenyl)diazen-1-yl]-3-oxo-N-(2-oxo-1,3-dihydro-1,3-benzodiazol-5-yl)butanamide	12236-62-3 /50694-80-9	C17H13ClN6O5	<chem>O=c1[nH]c2ccc(NC(=O)C(N=Nc3ccc(Cl)cc3N(=O)=O)C(=O)C)c2[nH]1</chem>	1.33	3.6	negative	90	2	2
2	1-[4-chloro-3-(trifluoromethyl)phenyl]-3-(4-chlorophenyl)urea*	369-77-7	C14H9Cl2F3N2O	<chem>Clc1ccc(cc1)NC(=O)Nc1ccc(Cl)c(c1)C(F)(F)F</chem>	0.32	1.9	negative	91	2	2
3	Arildone	56219-57-9	C20H29ClO4	<chem>COc1ccc(OCCCCC(C(=O)CC)C(=O)CC)c(Cl)c1</chem>	-0.22	-0.9	negative	98	3	2
4	Clomestron	4091-75-2	C19H23ClO2	<chem>COc1ccc2c(CCC3C2CCC2(C)C3CC(Cl)C2=O)c1</chem>	4.96	-0.5	negative	50	3	1
5	2,3,4,6-Tetrachlorophenol	58-90-2, 25167-83-3, 4901-51-3, 935-95-5	C6H2Cl4O	<chem>Oc1c(Cl)cc(Cl)c(Cl)c1Cl</chem>	-0.35	2.0	negative	64	3	1

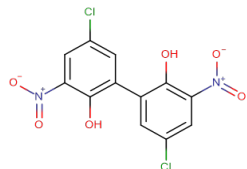
No	Compound Name	CAS No.	Formula	Smiles ¹	Δ m/z (ppm)	Δ Rt (min)	Ionization	Isotopic Pattern Match (%)	Identification Level *	Fragments predicted found
6	p-Chlorocinnamic acid	1615-02-7, 1866-38-2, 3752-25-8	C ₉ H ₇ ClO ₂	<chem>OC(=O)C=Cc1ccc(Cl)cc1</chem>	0.28	-2.5	negative	97	3	1
7	Dibromsalan	1320-09-8, 87-12-7	C ₁₃ H ₉ Br ₂ NO ₂	<chem>BrC1CCC(CC1)NC(=O)C1CC(Br)CCC1O</chem>	-0.44	2.4	negative	100	3	1
8	2-(4-bromo-3-hydroxyquinolin-2-yl)-2H-indene-1,3-dione	10319-14-9, 233-701-7	C ₁₈ H ₁₀ BrNO ₃	<chem>O=C1C(C(=O)c2ccccc2c1)nc2ccccc2c(Br)c1O</chem>	-0.25	-0.9	negative	100	3	1

¹1-[4-chloro-3-(trifluoromethyl)phenyl]-3-(4-chlorophenyl)urea is commonly known as cloflucarban used as disinfectant and is used in antimicrobial soaps and deodorants.

Table S13. Suspect screening found in soils and sediments at level of identification 2. MS and MS/MS information can be found at Tables S10 and S12.

No.	Compound Name	CAS No.	Application	Structure	Δ m/z (ppm)	Δ RT (min)	Isotopic Pattern Match (%)	Ionization	Matrix
1	1-[4-chloro-3-(trifluoromethyl)phenyl]-3-(4-chlorophenyl)urea*	369-77-7	Pharma		0.32	1.9	91	Negative	Sediment

No.	Compound Name	CAS No.	Application	Structure	$\Delta m/z$ (ppm)	ΔRT (min)	Isotopic Pattern Match (%)	Ionization	Matrix
2	2-(chloromethyl)-4-nitrophenol	2973-19-5	Many sources		1.4	0.9	94	Negative	Soil
3	2-[2-(4-chloro-2-nitrophenyl)diazen-1-yl]-3-oxo-N-(2-oxo-1,3-dihydro-1,3-benzodiazol-5-yl)butanamide	12236-62-3/50694-80-9	No Data		1.33	3.60	90	Negative	Sediment
4	Dichlorophenol	576-24-9, 95-77-2, 591-35-5, 583-78-8	Many sources		0.4	0.3	100	Negative	Soil

No.	Compound Name	CAS No.	Application	Structure	$\Delta m/z$ (ppm)	ΔRT (min)	Isotopic Pattern Match (%)	Ionization	Matrix
5	Niclofolan	10331-57-4	Pharmaceutical (used widely in sheep)		0.4	-0.5	98	Negative	Soil

*1-[4-chloro-3-(trifluoromethyl)phenyl]-3-(4-chlorophenyl)urea is commonly known as cloflucarban used as disinfectant and is used in antimicrobial soaps and deodorants.

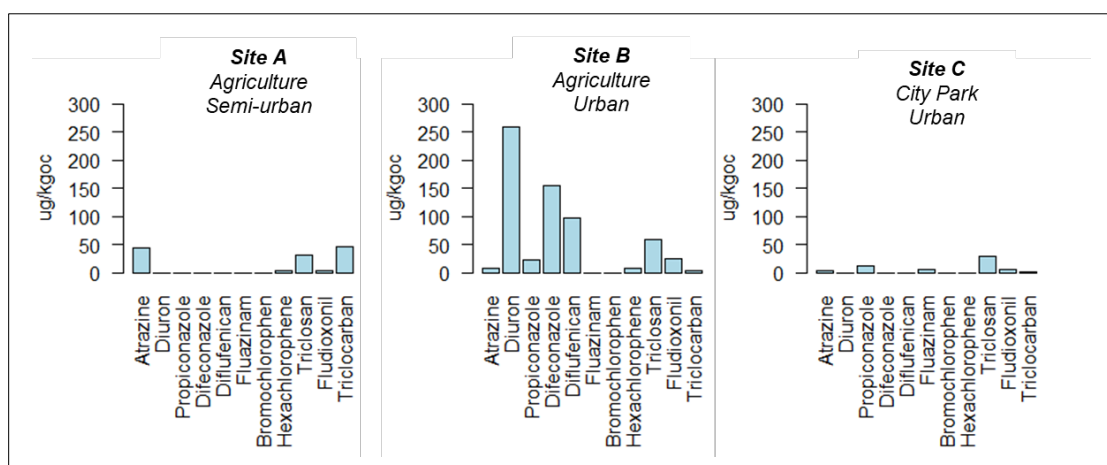


Figure S10. Concentrations in $\mu\text{g}/\text{kg}_{\text{oc}}$ of selected compounds of the most contaminated sites (A, B and C) detected in soils. The samples were taken between 2005 and 2009. More details about the site can be found in Table S1.

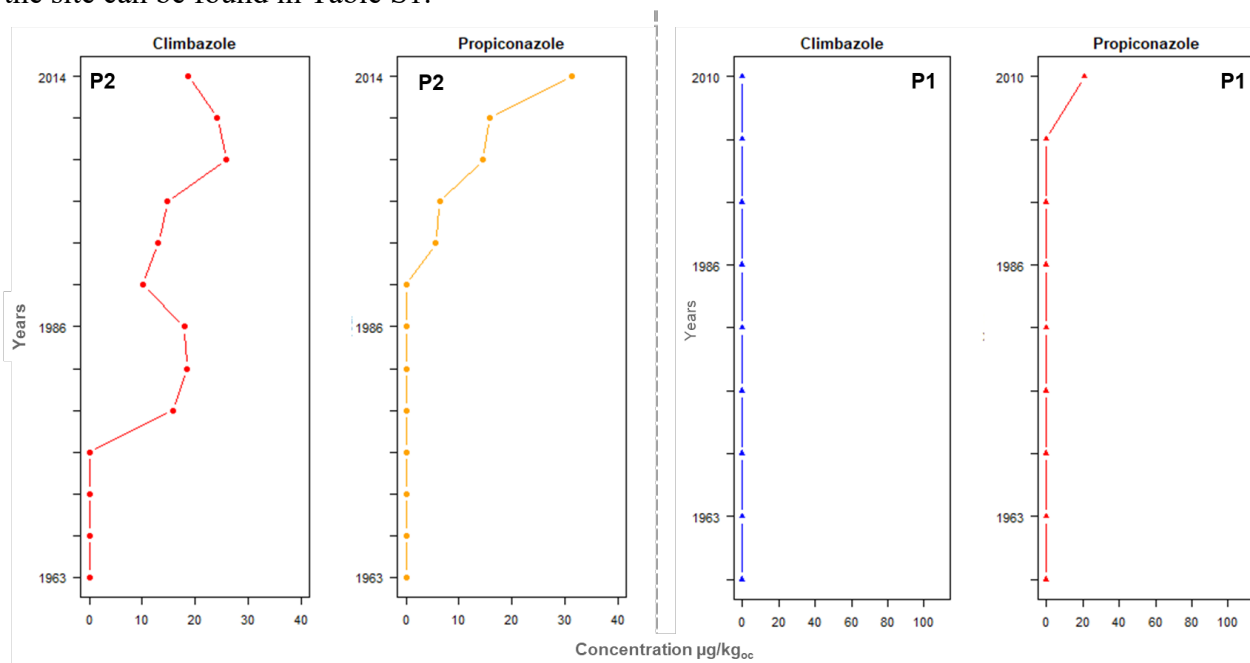


Figure S11. Temporal concentration trends of the fungicides climbazole and propiconazole from Uster (P2) with impact of WWTP effluents showing different trends with regards to location P1 (deepest part of the lake, left). Map location and details of sediment cores can be found in Figure S2 and S3 and Table S3.

References

- Chiaia-Hernandez A, Schymanski E, Kumar P, Singer H, Hollender J. Suspect and nontarget screening approaches to identify organic contaminant records in lake sediments. *Analytical and Bioanalytical Chemistry* 2014; 406: 7323-7335.
- Chiaia-Hernández AC, Günthardt BF, Frey MP, Hollender J. Unravelling Contaminants in the Anthropocene Using Statistical Analysis of Liquid Chromatography–High-Resolution Mass

- Spectrometry Nontarget Screening Data Recorded in Lake Sediments. *Environmental Science & Technology* 2017; 51: 12547-12556.
- Chiaia-Hernandez AC, Krauss M, Hollender J. Screening of Lake Sediments for Emerging Contaminants by Liquid Chromatography Atmospheric Pressure Photoionization and Electrospray Ionization Coupled to High Resolution Mass Spectrometry. *Environmental Science & Technology* 2013; 47: 976-986.
- Glüge J, Bogdal C, Scheringer M, Hungerbühler K. What determines PCB concentrations in soils in rural and urban areas? Insights from a multi-media fate model for Switzerland as a case study. *Science of The Total Environment* 2016; 550: 1152-1162.
- HighChem. (2016), mzCloud <https://mzcloud.org>, HighChem Ltd., Bratislava, Slovakia. Accessed 17.08.2016.
- Horai H, Arita M, Kanaya S, Nihei Y, Ikeda T, Suwa K, et al. MassBank: A public repository for sharing mass spectral data for life sciences. *Journal of Mass Spectrometry* 2010; 45: 703-714.
- JChem. ChemAxon software. Version 16.10.300 <https://www.chemaxon.com>. 2016.
- Mackay D. Multimedia Environmental Models: CRC Press 2001.
- Moschet C, Wittmer I, Simovic J, Junghans M, Piazzoli A, Singer H, et al. How a Complete Pesticide Screening Changes the Assessment of Surface Water Quality. *Environmental Science & Technology* 2014; 48: 5423-5432.
- Pintado-Herrera MG, González-Mazo E, Lara-Martín PA. In-cell clean-up pressurized liquid extraction and gas chromatography–tandem mass spectrometry determination of hydrophobic persistent and emerging organic pollutants in coastal sediments. *Journal of Chromatography A* 2016; 1429: 107-118.
- RSC. (2014) ChemSpider <http://www.chemspider.com/>, Royal Society of Chemistry, UK (accessed 21/3/2014).
- Schymanski EL, Jeon J, Gulde R, Fenner K, Ruff M, Singer HP, et al. Identifying Small Molecules via High Resolution Mass Spectrometry: Communicating Confidence. *Environmental Science & Technology* 2014; 48: 2097-2098.
- Stempel S, Scheringer M, Ng CA, Hungerbühler K. Screening for PBT Chemicals among the “Existing” and “New” Chemicals of the EU. *Environmental Science & Technology* 2012; 46: 5680-5687.
- Wolf S, Schmidt S, Muller-Hannemann M, Neumann S. In silico fragmentation for computer assisted identification of metabolite mass spectra. *BMC Bioinformatics* 2010; 11: 148.