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Supporting Information

Organic Micropollutants in Rivers Downstream of the Megacity Beijing: Sources and Mass Fluxes in a Large-scale Wastewater Irrigation System

Florian Heeb[1], Heinz Singer[1,*], Benoit Pernet-Coudrier[1], Weixiao Qi[2], Huijuan Liu[2], Philipp Longrée[1], Beat Müller[1], and Michael Berg[1,*]

[1] Eawag, Swiss Federal Institute of Aquatic Science and Technology, 8600 Dübendorf, Switzerland

[2] State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco- Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

* Corresponding authors. E-mail addresses: heinz.singer@eawag.ch (H.S.), michael.berg@eawag.ch (M.B.)

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Additional information about the Haihe River System

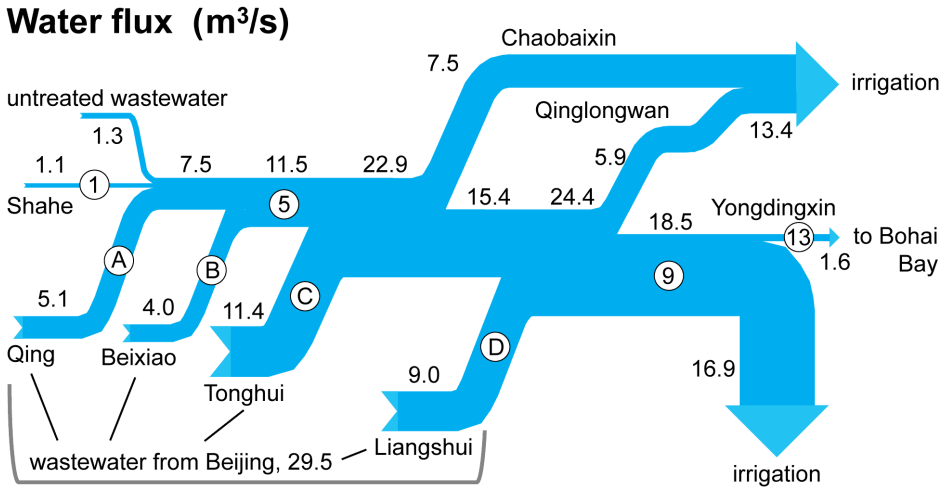


Figure S1. Flow diagram of the water discharge in the Haihe System (m3/s). The numbers 1, 5, 9 and 13 mark sampling sites where daily discharge measurements were available. The letters A to D mark the inflows of the four main channels carrying the discharge of the wastewater from the urban area of Beijing: Qing (A), Beixiao (B), Tonghui (C) and Liangshui (D). The discharge of the Chaobaixin and the Qinlongwan River represent only the amount of water that originates from the Haihe River System and do not take into account other inputs from outside of the studied system (*1*).

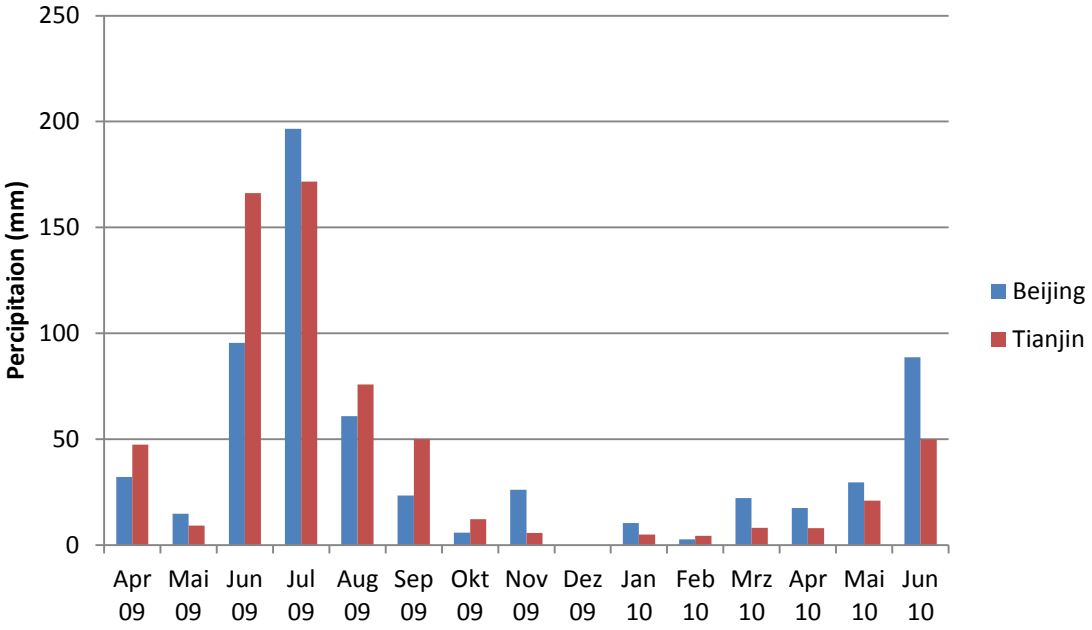


Figure S2. Monthly precipitation in Beijing and Tianjin during the study period (*2*).

Table S1. Characteristics of the five major WWTPs of Beijing. The number of inhabitants connected was calculated based on a per capita water consumption of 200 L/capita/day.

A/A/O: Anaerobic/anoxic/aerobic, MBR: Membrane bioreactor, MF: Micro filtration, RO: Reverse osmosis; UF: Ultra filtration.

WWTP Name	Inhabitants connected	Main treatment processes	Year of con-struction	Average discharge during study period (m^3/s)	Hydraulic retention time (h)	Sludge retention time (d)	Additional advanced treatment
Gaobeidian	4 925 000	A/A/O	1999	11.4	11	20	-
Xiaohongmen	3 000 000	A/A/O	2006	6.9	11	15	MF/RO
Qinghe	2 000 000	A/A/O	2004	4.6	11	13	UF / ozonation
Jiuxianqiao	1 000 000	Oxidation Ditch	2000	2.3	15	13	-
Beixiaohe	500 000	A/A/O and MBR	1990	1.2	5 (17 for MBR)	5 (20 for MBR)	RO

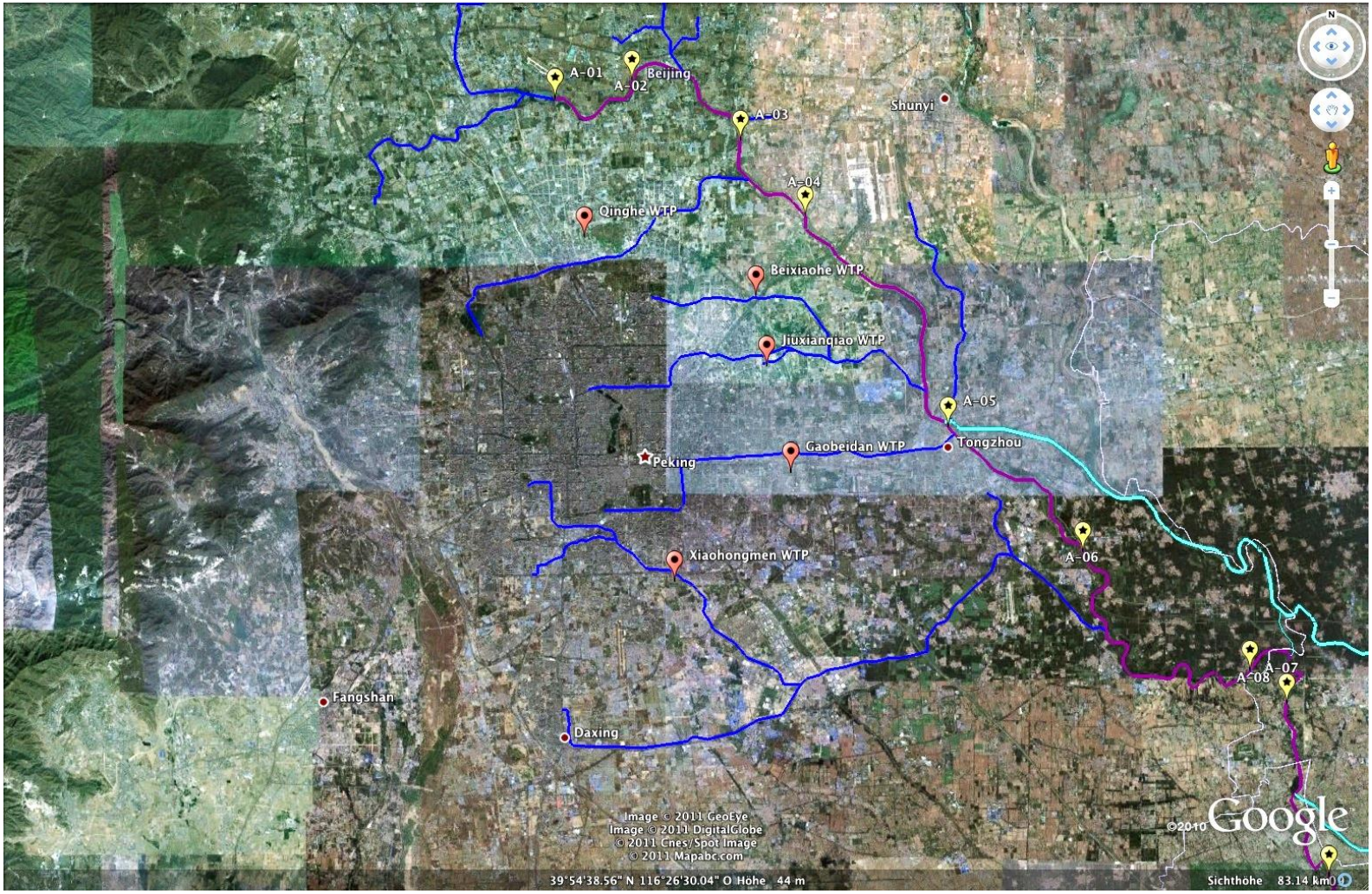


Figure S3. Location of the five major WWTPs in Beijing. The red marks with points show the location of the WWTPs within the city of Beijing. The yellow marks with stars show the location of sampling sites 1 to 7 (Satellite maps from Google Earth).

Additional information on analytical methods

Text S1: Chemicals and reagents

Unlabeled standard compounds as well as isotope labeled internal standards (ILIS, cf. Table S5) were purchased from Dr. Ehrenstorfer (Augsburg, Germany), Fluka Chemicals (Buchs, Switzerland), Riedel de Haën (Seelze, Germany), Sigma-Aldrich (Buchs, Switzerland) and Toronto Research Chemicals (North York, ON, Canada) or provided by Novartis (Basel, Switzerland). Stock solutions of all compounds and ILIS were prepared in methanol, ethanol, or acetonitrile with concentrations of 1 µg/µL. Ethanol or acetonitrile mixture solutions for the different analytes were prepared in concentrations of 0.01, 0.1 and 1 ng/mL. A ILIS mixture solution in ethanol contained 0.2 ng/µL of each substance (respectively 2.5 ng/µL of iopromide). HPLC-grade methanol and water were from Scharlau (Barcelona, Spain). Certificated reference standard mixes for selected pharmaceuticals, pesticides and x-ray contrast media, which were used for quality assurance, were purchased from Dr. Ehrenstorfer (Augsburg, Germany). All other chemicals were from Merck (Darmstadt, Germany).

Text S2: Sample preparation

Surface water and wastewater samples (250 mL) were collected in bake-out glass bottles (Schott) and sent cooled by express mail to Switzerland for analysis. There, the samples were kept at -20 °C in half-filled glass bottles stored in a horizontal position until extraction. The thawed samples were filtered through glass microfiber filters (GF/F, pore size 0.7 µm, Whatman). For the analysis of the surface water samples, 50 µL aliquots of the ILIS solution were added to a 50 mL sample volume and mixed. For the analysis of the wastewater inflows and effluents, the samples were diluted 1:4 and 1:10 (V_{sample}/V_{total}). An aliquot of 20 mL was used for analysis of surface and wastewater samples. The samples were adjusted to pH 4 prior to enrichment by adding 80 mL acetate buffer (5 M sodium acetate dissolved in water) with the autosampler.

Text S3: Online SPE-LC tandem mass spectrometry

The online SPE-LC set-up was similar to the one reported by Stoob et al. (*3*) and consisted of a tri-directional autosampler (HTC PAL, CTC Analytics, Zwingen, Switzerland), a dispenser syringe, a sample loop of 20 mL, three LC pumps, two six-port valves, and an on-line extraction cartridge. The HPLC pump system consisted of a binary pump (load pump, Surveyor LC, Finnigan), a quaternary low pressure mixing gradient pump (elution pump, Rheos 2200, Flux instruments, Switzerland) for the SPE elution and the methanol gradient, and an isocratic pump (Rheos, 2000; Flux instruments, Switzerland) for the water gradient and a column oven (Portmann Instruments AG, Biel-Benken, Switzerland).

The analytical method was similar to the one described by Singer et al. (*4*). For sample enrichment, a 20 mm x 2.1 mm cartridge was used, filled with 9.1 mg Oasis HLB (Waters, Baden-Dättwil, Switzerland) and 9.8 mg of a mixture of Strata XCW, Strata XAW (Phenomenex, Munich, Germany) and Isolute ENV+ (Separtis GmbH, Gerlingen, Switzerland), mixed with a ratio of 1:1:1.5. Elution from the cartridge was achieved with methanol amended with 0.1% formic acid. In order to re-establish the initial conditions for the LC, the methanol/0.1% formic acid SPE eluate was diluted with water containing 5mM ammonium acetate. To prevent cross-contamination, the sample loop and the extraction cartridge were flushed with acetonitrile after every extraction and conditioned with water containing 5mM ammonium acetate prior to enrichment of the next sample.

Separation was achieved using a 30 x 150mm Atlantis T3 with a particle size of 3µm (Waters, Baden-Dättwil, Switzerland) at 30 °C with a total flow rate of 300 µl/ min. Solvent A was water acidified with 5mM ammonium acetate and solvent B was methanol acidified with 0.1% formic acid. The gradient was initiated with 10% B for 8 min, followed by a 18 min linear gradient to 95% B. Afterwards the column was washed with 95% B for 3 min. The column was equilibrated for 7 minutes prior to the next analysis.

The LC was coupled with an electrospray probe (ESI) to a TSQ Quantum Ultra triple quadrupole MS (Thermo Scientific, San Jose, CA, USA), operated in a scheduled selected reaction monitoring (SRM) mode with a cycle time of 1s. Analyses were performed in the positive or negative mode during the same run (Table S3 SI). For all analytes, protonated ([M+H]+) or deprotonated ([M-H]-) molecular ions were selected as precursor ions. Specific and intense product ions of each target analyte were used for quantification, and a secondary product ion was used as a qualifier ion for confirmatory purposes. Detailed specifications used for the tandem mass spectrometry are given in Table S2 SI. Details of the substance specific parameters for the ionization and detection of the analytes are given in the SI (Table S3).

Text S4: Quantification and quality control

Two transitions of the precursor ion were monitored. The concentrations were determined by using the main product ions (quantifier). To confirm the results the secondary product ion (qualifier) was used and the ion ratio between the quantifier and qualifier was checked against the standards. The ion ratios should not differ by more than 30%. Together with the retention times, they were used to ensure correct peak assignment and to confirm the quantified results. We used corresponding ILIS for quantification whenever possible. Where no labeled compounds were available, quantification was performed with another labeled standard. The most suitable ILIS was selected according to the retention time and structural analogy.

Concentrations in the samples were calculated by comparing the peak area ratios of the analytes and their assigned ILIS to the corresponding ratios in the standard solutions. Calibration curves were obtained from standards spiked in nanopure water at 1, 2, 5, 10, 20, 50, 100, 200, 500, 1000, 2000 and 5000 ng/L (respectively 25, 50, 100, 250, 500, 1000, 2500, 5000, 10000 and 25000 ng/L for iopromide). Calibration curves were produced by a weighted (1/x) linear least square regression. A standard curve was acquired at the beginning and at the end of a measurement series. Procedural blanks consisted of nanopure water. Blank samples consisted of nanopure water with ILIS but without analytes. Both were extracted in every sequence in order to uncover potential analytical interferences or possible carryover.

Limits of detection (LOD) and quantification (LOQ) were calculated on the basis of signal to noise ratios (S/N) of 3 and 10, respectively. The LOD was obtained from a spiked nanopure standard at a low concentration. For the sample based LOD, the matrix induced suppression/enhancement factor was calculated by comparing the absolute peak area of the standard in nanopure water with the absolute peak area in the spiked environmental sample. The sample based LODs were determined by multiplying the LOD with the suppression/enhancement factor. The precision of the measurements was assessed by repeatedly extracting standards spiked in nanopure water at 50, 100, 200 and 500 ng/L in every sequence. The quality of the used stock solutions was crosschecked by extracting validated

certificated reference standard mixes. For the determination of the relative recoveries, the samples were spiked prior to extraction with ILIS and with 200, 500 or 1000 ng/L of analytes. The calculated amount minus the amount already present before spiking was then divided by the spiked concentration. The average relative recoveries lay within a range of 85%-112% for all compounds expect imidacolprid (77%) and clindamycin (132%). Uncertainties due to the precision of the measurement, the analytical procedure, reference material purity, gravimetric/volumetric devices as well as standard stock solubility were calculated as described below. Detailed values concerning LOD, LOQ, relative recoveries and uncertainties for all compounds are listed in Table S2 SI.

Table S2. ESI and collision gas settings used for the tandem mass spectrometry.

Sheath gas	Nitrogen (50 arbitrary units)
Auxiliary gas	Nitrogen (10 arbitrary units)
Collision gas	Argon (1.5 mTorr)
Spray voltage	+3800 V (positive mode), -3000 V (negative mode)
Capillary temperature	350 °C

Table S3. Substance-specific settings for the tandem mass spectrometry.

	Precursor mass (m/z)	Product mass (m/z)	Collision energy (V)	Tube lens (V)	Acquisition (min) start	Acquisition (min) end	ESI
2,4-D	219.0	161.1	17	50	24.1	28.1	neg
2,4-D	219.0	125.1	18	50	24.1	28.1	neg
2,4-D 13C6	225.0	166.9	21	50	24.1	28.1	neg
2,4-D 13C6	225.0	131.0	25	50	24.1	28.1	neg
2,6-Dichlorbenzamide	190.0	173.2	18	77	16.7	20.7	pos
2,6-Dichlorbenzamide	190.0	145.2	28	77	16.7	20.7	pos
2-Amino-Benzimidazole	134.1	65.2	32	93	12.6	16.6	pos
2-Amino-Benzimidazole	134.1	92.2	23	93	12.6	16.6	pos
2-Naphtalene sulfonic acid	207.0	142.7	25	57	20.5	24.5	neg
2-Naphtalene sulfonic acid	207.0	79.5	39	57	20.5	24.5	neg
4-Acetamidoantipyrine	246.1	228.1	13	63	15.9	19.9	pos
4-Acetamidoantipyrine	246.1	204.1	13	63	15.9	19.9	pos
5-Methyl-Benzotriazole *	134.1	77.0	22	80	20.2	24.2	pos
5-Methyl-Benzotriazole *	134.1	79.1	17	80	20.2	24.2	pos
5-Methyl-Benzotriazole D6	140.0	81.0	28	80	20.2	24.2	pos
5-Methyl-Benzotriazole D6	140.0	85.0	20	80	20.2	24.2	pos
Atenolol acid D5	273.2	150.1	24	81	13.5	17.5	pos
Atenolol acid D5	273.2	196.1	17	81	13.5	17.5	pos
Atenolol acid	268.2	145.0	24	81	13.5	17.5	pos
Atenolol acid	268.2	191.1	17	81	13.5	17.5	pos
Atenolol D7	274.2	190.0	18	93	11.6	15.6	pos
Atenolol D7	274.2	226.1	16	93	11.6	15.6	pos
Atenolol	267.2	190.0	18	93	11.6	15.6	pos
Atenolol	267.2	225.1	16	93	11.6	15.6	pos
Atrazine-2-Hydroxy D5	203.0	161.1	14	96	15.7	19.7	pos
Atrazine-2-Hydroxy D5	203.0	86.0	20	96	15.7	19.7	pos
Atrazine-2-Hydroxy	198.1	156.1	14	96	15.7	19.7	pos
Atrazine-2-Hydroxy	198.1	86.0	20	96	15.7	19.7	pos
Atrazine D5	221.1	179.0	19	91	23.7	27.7	pos
Atrazine D5	221.1	101.0	30	91	23.7	27.7	pos
Atrazine-desethyl-2-hydroxy	170.1	128.0	16	81	11.2	15.2	pos
Atrazine-desethyl-2-hydroxy	170.1	86.1	21	81	11.2	15.2	pos
Atrazine	216.1	174.0	19	91	23.7	27.7	pos
Atrazine	216.1	104.0	30	91	23.7	27.7	pos
Atrazine-Desethyl 15N3	193.0	151.0	17	94	20	24	pos
Atrazine-Desethyl 15N3	193.0	108.0	23	94	20	24	pos
Atrazine-Desethyl	188.1	146.0	17	94	20	24	pos
Atrazine-Desethyl	188.1	104.1	23	94	20	24	pos

	Precursor mass (m/z)	Product mass (m/z)	Collision energy (V)	Tube lens (V)	Acquisition (min) start	Acquisition (min) end	ESI
Atrazine-Desisopropyl D5	179.0	105.0	22	80	17.7	21.7	pos
Atrazine-Desisopropyl D5	179.0	137.0	14	80	17.7	21.7	pos
Atrazine-Desisopropyl	174.1	104.0	22	80	17.7	21.7	pos
Atrazine-Desisopropyl	174.1	132.0	14	80	17.7	21.7	pos
Azoxystrobin acid	390.1	372.4	11	62	23.2	27.2	pos
Azoxystrobin acid	390.1	344.1	23	62	23.2	27.2	pos
Azoxystrobin	404.1	372.4	13	63	24.2	28.2	pos
Azoxystrobin	404.1	344.4	23	63	24.2	28.2	pos
Bentazone D6	245.1	132.1	27	60	22.2	26.2	neg
Bentazone D6	245.1	198.0	25	60	22.2	26.2	neg
Bentazone	239.1	132.4	27	60	22.2	26.2	neg
Bentazone	239.1	197.0	25	60	22.2	26.2	neg
Benzotriazole D4	124.0	69.1	15	70	17.8	21.8	pos
Benzotriazole D4	124.0	96.0	12	70	17.8	21.8	pos
Benzotriazole	120.0	65.1	15	70	17.8	21.8	pos
Benzotriazole	120.0	92.0	12	70	17.8	21.8	pos
Bezafibrate D4	366.1	320.0	10	109	24.7	28.7	pos
Bezafibrate D4	366.1	143.0	17	109	24.7	28.7	pos
Bezafibrate	362.1	316.0	10	109	24.7	28.7	pos
Bezafibrate	362.1	139.0	17	109	24.7	28.7	pos
Carbamazepine-10,11-dihydro-10,11 Dihydroxy	271.1	180.3	29	72	20.1	24.1	pos
Carbamazepine-10,11-dihydro-10,11 Dihydroxy	271.1	253.4	5	72	20.1	24.1	pos
Carbamazepine-10,11-epoxid	253.1	180.3	26	57	20.6	24.6	pos
Carbamazepine-10,11-epoxid	253.1	210.3	14	57	20.6	24.6	pos
Carbamazepine-10,11-epoxid 13C,D2	257.1	183.2	26	57	20.6	24.6	pos
Carbamazepine-10,11-epoxid 13C,D2	257.1	213.2	14	57	20.6	24.6	pos
Carbamazepine 13C,D2	240.3	195.0	17	91	22.7	26.7	pos
Carbamazepine 13C,D2	240.3	194.0	22	91	22.7	26.7	pos
Carbamazepine	237.1	194.0	17	91	22.7	26.7	pos
Carbamazepine	237.1	192.0	22	91	22.7	26.7	pos
Carbendazim D4	196.1	164.0	14	84	16.5	20.5	pos
Carbendazim D4	196.1	136.1	31	84	16.5	20.5	pos
Carbendazim	192.1	160.0	14	84	16.5	20.5	pos
Carbendazim	192.1	132.1	31	84	16.5	20.5	pos
Clarithromycin	748.5	590.4	15	118	22.4	26.4	pos
Clarithromycin	748.5	158.0	22	118	22.4	26.4	pos
Clarithromycin D3	751.5	593.4	15	118	22.4	26.4	pos
Clarithromycin D3	751.5	161.0	22	118	22.4	26.4	pos
Climbazole	293.1	197.1	15	54	22.9	26.9	pos
Climbazole	293.1	225.1	14	54	22.9	26.9	pos
Clindamycin	425.2	126.0	33	97	19.6	23.6	pos
Clindamycin	425.2	377.1	15	97	19.6	23.6	pos

	Precursor mass (m/z)	Product mass (m/z)	Collision energy (V)	Tube lens (V)	Acquisition (min) start	Acquisition (min) end	ESI
Clofibric acid	213.0	85.0	11	80	24.5	28.5	neg
Clofibric acid	215.0	129.0	18	80	24.5	28.5	neg
Clofibric acid D4	219.0	133.0	18	80	24.5	28.5	neg
Clofibric acid D4	217.0	85.0	11	80	24.5	28.5	neg
Caffeine	195.1	138.1	18	81	16.7	20.7	pos
Caffeine	195.1	110.1	19	81	16.7	20.7	pos
Caffeine D9	204.0	144.1	18	81	16.7	20.7	pos
Caffeine D9	204.0	116.1	19	81	16.7	20.7	pos
DEET	192.1	91.2	30	84	23.7	27.7	pos
DEET	192.1	119.2	17	84	23.7	27.7	pos
DEET D7	199.2	98.0	30	84	23.7	27.7	pos
DEET D7	199.2	126.1	17	84	23.7	27.7	pos
Diazepam	285.1	193.1	31	107	24.9	28.9	pos
Diazepam	285.1	154.0	26	107	24.9	28.9	pos
Diazepam d5	290.1	198.1	31	107	24.9	28.9	pos
Diazepam d5	290.1	154.0	26	107	24.9	28.9	pos
Diclofenac	296.0	214.0	34	80	26.1	30.1	pos
Diclofenac	296.0	215.0	17	80	26.1	30.1	pos
Diclofenac D4	302.0	218.0	34	80	26.1	30.1	pos
Diclofenac D4	302.0	219.0	17	80	26.1	30.1	pos
Diuron D6	239.0	78.1	15	89	25.1	29.1	pos
Diuron D6	239.0	160.0	22	89	25.1	29.1	pos
Diuron	233.0	72.1	15	89	25.1	29.1	pos
Diuron	233.0	160.0	22	89	25.1	29.1	pos
Erythromycin	734.5	576.3	14	125	21.3	25.3	pos
Erythromycin	734.5	158.0	23	125	21.3	25.3	pos
Erythromycin-H2O	716.5	158.0	33	113	21.6	25.6	pos
Erythromycin-H2O	737.5	160.0	23	125	21.6	25.6	pos
Erythromycin-H2O 13C2	737.5	579.0	14	125	21.6	25.6	pos
Erythromycin-H2O 13C2	716.5	558.3	10	113	21.6	25.6	pos
Fluconazole	307.1	220.0	17	96	18.8	22.8	pos
Fluconazole	307.1	238.0	16	96	18.8	22.8	pos
Fluconazole D4	311.1	242.1	16	96	18.8	22.8	pos
Fluconazole D4	311.1	223.0	17	96	18.8	22.8	pos
Hydrochlorothiazide	296.0	268.9	18	88	13.5	17.5	neg
Hydrochlorothiazide	296.0	205.0	22	88	13.5	17.5	neg
Imidacloprid	256.1	209.0	16	64	17.8	21.8	pos
Imidacloprid	256.1	175.0	17	64	17.8	21.8	pos
Iopromide	791.9	299.8	60	140	13.8	17.8	pos
Iopromide	791.9	572.7	25	140	13.8	17.8	pos
Iopromide D3	794.9	575.7	25	140	13.8	17.8	pos
Iopromide D3	794.9	299.8	60	140	13.8	17.8	pos
Lidocaine D10	245.2	96.1	17	74	15.1	19.1	pos

	Precursor mass (m/z)	Product mass (m/z)	Collision energy (V)	Tube lens (V)	Acquisition (min) start	Acquisition (min) end	ESI
Lidocaine D10	245.2	64.1	34	74	15.1	19.1	pos
Lidocaine	235.2	86.1	17	74	15.1	19.1	pos
Lidocaine	235.2	58.1	34	74	15.1	19.1	pos
MCPA	199.0	141.2	19	51	24.2	28.2	neg
MCPA	201.0	143.1	19	51	24.2	28.2	neg
MCPA D6	207.0	149.1	19	51	24.2	28.2	neg
MCPA D6	205.0	147.2	19	51	24.2	28.2	neg
Mefenamic acid D3	245.1	212.0	25	80	27.2	31.2	pos
Mefenamic acid D3	245.1	227.0	15	80	27.2	31.2	pos
Mefenamic acid	242.1	224.0	15	80	27.2	31.2	pos
Mefenamic acid	242.1	209.0	25	80	27.2	31.2	pos
Metalaxyl	280.2	220.1	12	55	23.7	27.7	pos
Metalaxyl	280.2	192.1	16	55	23.7	27.7	pos
Metolachlor D6	290.1	258.1	15	52	26	30	pos
Metolachlor D6	290.1	176.1	23	52	26	30	pos
Metolachlor-ESA	328.1	80.0	43	75	25.6	29.6	neg
Metolachlor-ESA	328.1	121.0	25	75	25.6	29.6	neg
Metolachlor	284.0	252.1	15	52	26	30	pos
Metolachlor	284.0	176.1	23	52	26	30	pos
Metolachlor-OXA	278.1	174.2	18	56	24	29	neg
Metolachlor-OXA	278.1	206.2	14	56	24	29	neg
Metoprolol D7	275.2	159.0	17	91	16.5	20.5	pos
Metoprolol D7	275.2	133.0	21	91	16.5	20.5	pos
Metoprolol	268.2	159.0	17	91	16.5	20.5	pos
Metoprolol	268.2	133.0	21	91	16.5	20.5	pos
Metronidazole	172.1	82.1	24	71	14	18	pos
Metronidazole	172.1	128.0	14	71	14	18	pos
N,N Didesvenlafaxine	250.2	215.4	10	79	17	22.9	pos
N,N Didesvenlafaxine	250.2	232.4	5	79	17	22.9	pos
N Desvenlafaxine	264.2	246.2	7	95	16	22.9	pos
N Desvenlafaxine	264.2	121.1	30	95	16	22.9	pos
N Desvenlafaxine D3	267.2	249.3	7	95	16	22.9	pos
N Desvenlafaxine D3	267.2	121.1	30	95	16	22.9	pos
N4-Acetylsulfamethazine	321.1	186.0	26	101	17.5	21.5	pos
N4-Acetylsulfamethazine	321.1	134.1	34	101	17.5	21.5	pos
N4-Acetyl-Sulfamethoxazole	296.1	198.0	17	103	19	23	pos
N4-Acetyl-Sulfamethoxazole	296.1	134.0	23	103	19	23	pos
N4-Acetyl-Sulfamethoxazole D5	301.1	203.0	17	103	19	23	pos
N4-Acetyl-Sulfamethoxazole D5	301.1	139.0	23	103	19	23	pos
Phenazone D3	192.1	77.0	37	95	17.8	21.8	pos
Phenazone D3	192.1	131.0	17	95	17.8	21.8	pos
Phenazone	189.1	77.0	37	95	17.8	21.8	pos
Phenazone	189.1	131.0	17	95	17.8	21.8	pos

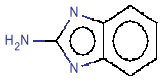
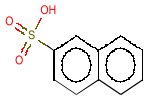
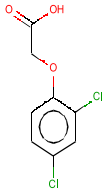
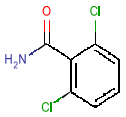
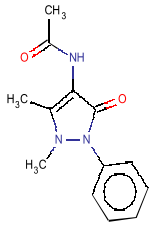
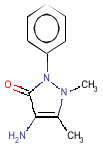
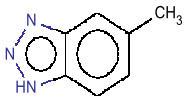
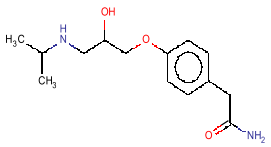
	Precursor mass (m/z)	Product mass (m/z)	Collision energy (V)	Tube lens (V)	Acquisition (min) start	Acquisition (min) end	ESI
Prometryn	242.1	158.0	23	63	24.7	28.7	pos
Prometryn	242.1	200.1	17	63	24.7	28.7	pos
Propazine-2-hydroxy	212.2	128.0	22	77	17.5	21.5	pos
Propazine-2-hydroxy	212.2	85.9	29	77	17.5	21.5	pos
Sotalol	273.1	255.0	11	87	11.1	15.1	pos
Sotalol	273.1	213.0	15	87	11.1	15.1	pos
Sotalol-D6	279.1	261.0	11	87	11.1	15.1	pos
Sotalol-D6	279.1	214.0	15	87	11.1	15.1	pos
Sucralose-FA D6	447.1	401.0	13	91	17.7	21.7	neg
Sucralose-FA D6-FA	447.1	365.0	17	91	17.7	21.7	neg
Sucralose-FA	441.1	395.0	13	91	17.7	21.7	neg
Sucralose-FA	441.1	359.0	17	91	17.7	21.7	neg
Sulfadiazine D4	255.1	96.1	32	73	13.1	17.1	pos
Sulfadiazine D4	255.1	160.0	20	73	13.1	17.1	pos
Sulfadiazine	251.1	156.0	14	80	13.1	17.1	pos
Sulfadiazine	251.1	108.0	21	80	13.1	17.1	pos
Sulfamethazine 13C6	285.1	186.0	16	80	16.4	20.4	pos
Sulfamethazine 13C6	285.1	124.0	24	80	16.4	20.4	pos
Sulfamethazine	279.0	186.0	16	80	16.4	20.4	pos
Sulfamethazine	279.0	124.0	24	80	16.4	20.4	pos
Sulfamethoxazole D4	258.1	160.0	15	94	17	21	pos
Sulfamethoxazole D4	258.1	96.0	24	94	17	21	pos
Sulfamethoxazole	254.1	156.0	15	94	17	21	pos
Sulfamethoxazole	254.1	92.0	24	94	17	21	pos
Sulfapyridine	250.1	156.0	14	87	14.7	18.7	pos
Sulfapyridine	250.1	184.0	13	87	14.7	18.7	pos
Sulfapyridine D4	254.0	160.0	16	87	14.7	18.7	pos
Sulfapyridine D4	254.0	96.0	29	87	14.7	18.7	pos
Tramadol D6	270.2	64.1	16	62	16.4	20.4	pos
Tramadol D6	270.2	45.1	78	62	16.4	20.4	pos
Tramadol	264.2	58.1	16	62	16.4	20.4	pos
Tramadol	264.2	42.1	78	62	16.4	20.4	pos
Trimetoprim D9	300.1	234.0	10	98	14	18	pos
Trimetoprim D9	300.1	264.0	14	98	14	18	pos
Trimetoprim	291.1	230.0	10	98	14	18	pos
Trimetoprim	291.1	261.0	14	98	14	18	pos
Tritosulfuron	446.0	195.3	18	108	24.6	28.6	pos
Tritosulfuron	446.0	145.2	34	108	24.6	28.6	pos
Valsartan 15N,13C5	442.3	207.2	20	85	24.8	28.8	pos
Valsartan 15N,13C5	442.3	291.2	14	85	24.8	28.8	pos
Valsartan	436.2	207.0	27	85	24.8	28.8	pos
Valsartan	436.2	291.1	18	85	24.8	28.8	pos
Venlafaxine D6	284.2	58.1	18	63	18.7	22.7	pos

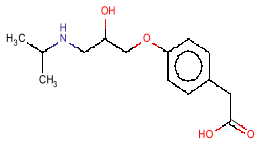
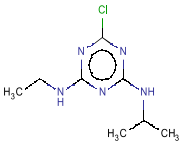
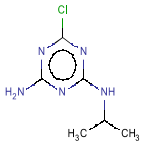
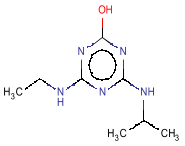
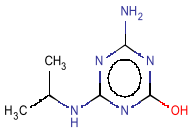
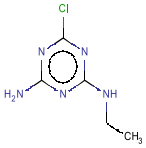
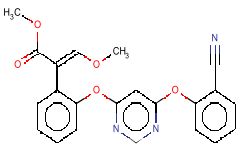
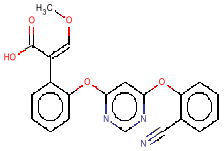
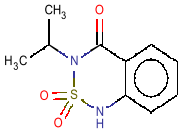
	Precursor mass (m/z)	Product mass (m/z)	Collision energy (V)	Tube lens (V)	Acquisition (min) start	Acquisition (min) end	ESI
Venlafaxine D6	284.2	266.2	10	63	18.7	22.7	pos
Venlafaxine	278.2	58.1	18	63	18.7	22.7	pos
Venlafaxine	278.2	260.2	10	63	18.7	22.7	pos

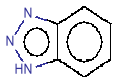
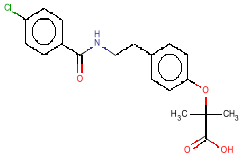
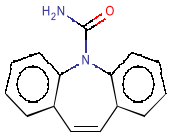
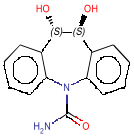
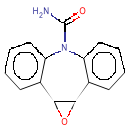
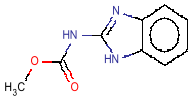
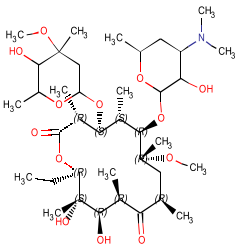
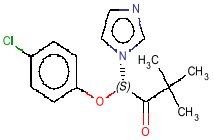
Quantifier is printed in bold

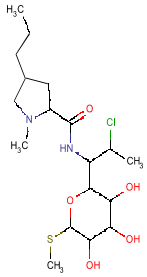
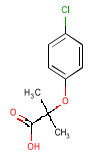
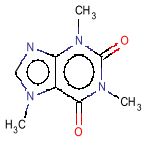
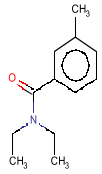
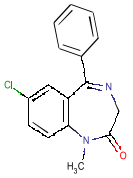
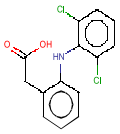
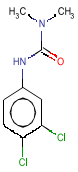
* As the chromatographic separation of 4- and 5-metyhl benzotriazole is not sufficient, the determined methyl benzotriazole concentration in environmental samples reflects the sum of 4- and 5-methylbenzotriazol. The reference standard used was 5-metyhl benzotriazole. More details can be found in (5).

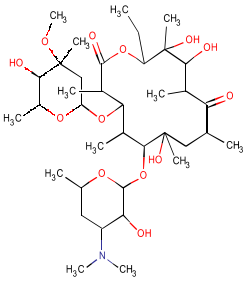
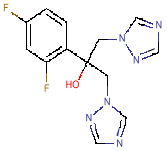
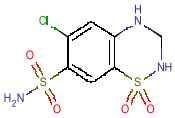
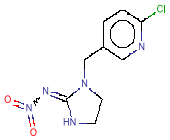
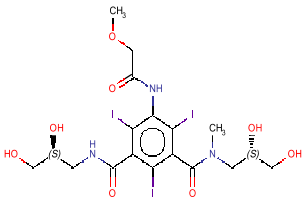
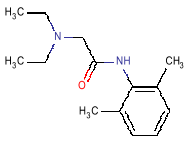
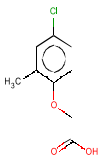
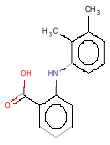
Table S4. Substance type, CAS-Number and structure of the measured micropollutants.

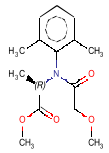
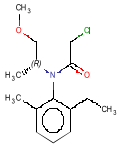
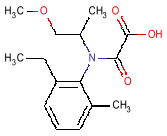
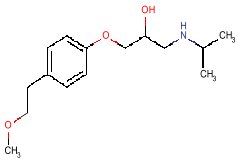
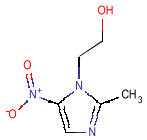
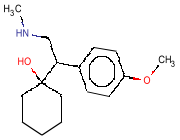
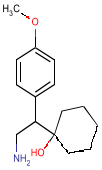
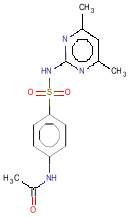
Substance	Substance class	CAS number	Structure
2-Amino-benzimidazole	Fungicides metabolite	934-32-7	
2-Naphtalene sulphonic acid	Industrial chemical	120-18-3	
2,4-D	Herbicides	94-75-7	
2,6-Dichlor-benzamide	Herbicides metabolite	2008-58-4	
4-Acetamido-antipyrine	Analgesic metabolite	83-15-8	
4-Aminoantipyrine	Analgesic metabolite	83-07-8	
5-Methyl-Benzotriazole	Corrosion inhibitor	136-85-6	
Atenolol	Beta-blocker	29122-68-7	

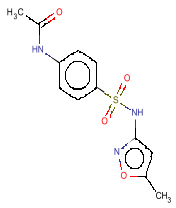
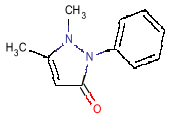
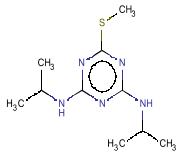
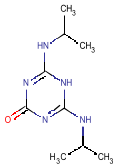
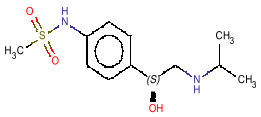
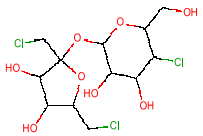
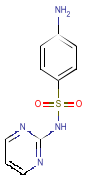
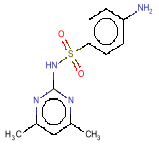
Substance	Substance class	CAS number	Structure
Atenolol acid	Beta-blocker metabolite	56392-14-4	
Atrazine	Herbicide	1912-24-9	
Atrazine-desethyl	Herbicide metabolite	6190-65-4	
Atrazine-2-hydroxy	Herbicide metabolite	2163-68-0	
Atrazine-desethyl-2-hydroxy	Herbicide metabolite	19988-24-0	
Atrazine-6-desisopropyl	Herbicide metabolite	1007-28-9	
Azoxystrobin	Fungicide	131860-33-8	
Azoxystrobin acid	Insecticide metabolite		
Bentazone	Herbicide	25057-89-0	

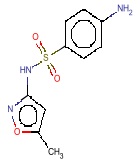
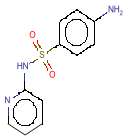
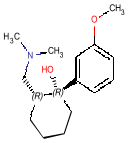
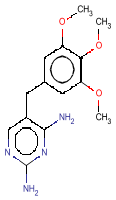
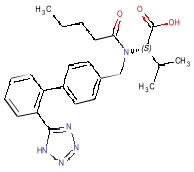
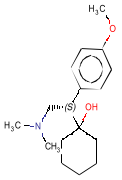
Substance	Substance class	CAS number	Structure
Benzotriazole	Corrosion inhibitor	95-14-7	
Bezafibrate	Lipid regulator	41859-67-0	
Carbamazepine	Anticonvulsant	298-46-4	
Carbamazepine-dihydro-dihydroxy	Anticonvulsant metabolite	58955-93-4	
Carbamazepine epoxide	Anticonvulsant metabolite	36507-30-9	
Carbendazim	Fungicide	10605-21-7	
Clarithromycin	Antibiotic	81103-11-9	
Climbazole	Antifungal	38083-17-9	

Substance	Substance class	CAS number	Structure
Clindamycin	Analgesic	18323-44-9	
Clofibric acid	Lipid regulator metabolite	882-09-7	
Caffeine	Food ingredient	58-08-2	
DEET	Insect repellent	134-62-3	
Diazepam	Anxiolytic	439-14-5	
Diclofenac	Analgesic	15307-86-5	
Diuron	Herbicide	330-54-1	

Substance	Substance class	CAS number	Structure
Erythromycin	Antibiotic	114-07-8	
Fluconazole	Antifungal	86386-73-4	
Hydrochlorothiazide	Diuretic-Cardiovascular	58-93-5	
Imidacloprid	Insecticide	138261-41-3	
Iopromide	X-ray contrast media	73334-07-3	
Lidocaine	Anesthetic	137-58-6	
MCPA	Herbicide	94-74-6	
Mefenamic acid	Analgesic	61-68-7	

Substance	Substance class	CAS number	Structure
Metalaxyl	Fungicide	57837-19-1	
Metolachlor	Herbicide	51218-45-2	
Metolachlor OXA	Herbicide metabolite	152019-73-3	
Metoprolol	Beta-blocker	37350-58-6	
Metronidazole	Antibiotic	443-48-1	
N-Desmethyl-venlafaxine	Antidepressant metabolite	149289-30-5	
N,N-Didesmethyl-venlafaxine	Antidepressant metabolite	93413-77-5	
N4-Acetyl-sulfamethazine	Antibiotic metabolite	100-90-3	

Substance	Substance class	CAS number	Structure
N4-Acetyl-sulfamethoxazole	Antibiotic metabolite	21312-10-7	
Phenazone	Analgesic	60-80-0	
Prometryn	Herbicide	7287-19-6	
Propazine-2-hydroxy	Herbicide metabolite	7374-53-0	
Sotalol	Beta-blocker	3930-20-9	
Sucralose	Sweetener	56038-13-2	
Sulfadiazine	Antibiotic	68-35-9	
Sulfamethazine	Antibiotic	57-68-1	

Substance	Substance class	CAS number	Structure
Sulfamethoxazole	Antibiotic	723-46-6	
Sulfapyridine	Antibiotic	144-83-2	
Tramadol	Analgesic	27203-92-5	
Trimethoprim	Antibiotic	738-70-5	
Valsartan	Angiotensin II antagonist	137862-53-4	
Venlafaxine	Antidepressant	93413-69-5	

Text S5: Uncertainty calculation for the quantified concentrations

The combined relative uncertainty ($relU_{tot}$) of the measured concentrations of micropollutants was calculated as:

$$relU_{tot} = \sqrt{(relU_{bias})^2 + \left(relU_{precision}\right)^2 + (relU_{matetrial})^2 + \left(relU_{stability}\right)^2}$$

$relU_{bias}$: Relative uncertainty due to the bias of the analytical procedure

$relU_{precision}$: Relative uncertainty due to the precision of the measurements

$relU_{material}$: Relative uncertainty due to material purity as well as imprecision of gravimetric and volumetric devices

$relU_{stability}$: Relative uncertainty due to the stability of standard stock solutions

$relU_{bias}$ was calculated based on the relative recoveries of every compound:

$$relU_{bias} = \frac{\sqrt{\left(\frac{1 - mean\ recovery}{k}\right)^2 + \left(\frac{SD}{\sqrt{n}}\right)^2}}{mean\ recovery}$$

n: number of measured recoveries
SD: Standard deviation of the measured recoveries
k: coverage factor (k=1 for one standard deviation)

$relU_{precision}$ was calculated based on repeated measurements of standards spiked in nanopure water at 50, 100, 200 and 500 ng/L in every sequence:

$$relU_{precision} = \sqrt{\frac{\left(RSD_{50}{}^2 \times (n_{50} - 1)\right) + \left(RSD_{100}{}^2 \times (n_{100} - 1)\right) + \left(RSD_{200}{}^2 \times (n_{200} - 1)\right) + \left(RSD_{500}{}^2 \times (n_{500} - 1)\right)}{(n_{50} - 1) + (n_{100} - 1) + (n_{200} - 1) + (n_{500} - 1)}}$$

With:

$$RSD_{level} = \frac{\sqrt{\frac{\sum (x - level)^2}{(n_{level} - 1)}}}{level}$$

level: concentration level of the corresponding standard
n_{level}: number of repeated measurements of a standard at a given concentration level
RSD: Relative standard deviations of all standards at a given concentration level

$relU_{material}$ was estimated to be 2%.

$relU_{stability}$ was estimated to be 0.5% for stable compounds and 20% for less stable compounds.

The resulting uncertainties for the concentrations of all measured compounds are listed in Table S5. For the caution of the uncertainties of the mass flows, several factors were considered in addition to the uncertainties in the underlying concentrations. Uncertainties in the discharge data and uncertainties due to the representativeness of the samples were estimated to be 10% each. Uncertainties in the loads due to concentrations below the LOD and LOQ were considered by calculating maximum and minimum load scenarios.

Table S5. Quality assurance data.

Compound	LOQ (ng/L)	LOD (ng/L)	Internal standard	Number of recoveries	Average recovery (%)	Standard deviation (%)	$relU_{bias}$ (%)	$relU_{precision}$ (%)	$relU_{material}$ (%)	$relU_{stability}$ (%)	Utot (%)
2-Aminobenzimidazole	6	2		5	100	10	4	27	2	0.5	27
2,4-D	43	14	IStd	19	94	8	20	10	2	0.5	22
2,6-Dichlorbenzamide	60	20		18	85	15	6	9	2	0.5	11
4-Acetamidoantipyrine	31	10		8	88	27	15	33	2	0.5	37
4-Aminoantipyrine	3	1		5	105	33	15	28	2	20.0	38
5-Methyl-Benzotriazole	8	3	IStd	16	94	10	16	47	2	0.5	50
Atenolol	10	3	IStd	18	101	7	7	9	2	0.5	11
Atenolol_acid	5	2	IStd	5	100	7	2	6	2	0.5	6
Atrazine	29	10	IStd	19	95	11	3	5	2	0.5	6
Atrazine-desethyl	9	3	IStd	19	93	7	6	6	2	0.5	8
Atrazine-2-hydroxy	16	5	IStd	18	104	28	7	4	2	0.5	8
Atrazine-desethyl-2-hydroxy	8	3		18	93	40	8	6	2	0.5	10
Atrazine-desisopropyl	5	2	IStd	19	94	6	12	58	2	0.5	59
Azoxystrobin	30	10		17	96	17	6	10	2	0.5	12
Azoxystrobin_acid	4	1		18	92	14	6	26	2	0.5	27
Bentazone	10	3	IStd	9	98	6	9	31	2	0.5	32
Benzotriazole	16	5	IStd	10	110	64	3	17	2	0.5	18
Bezafibrate	164	55	IStd	19	98	10	23	7	2	0.5	24
Carbamazepine	16	5	IStd	19	95	10	3	6	2	0.5	7
Carbamazepine-10,11-dihydro..	2	1		17	98	11	6	7	2	0.5	9
Carbamazepine-10,11-epoxide	49	16	IStd	19	100	7	3	18	2	0.5	18
Carbendazim	3	1	IStd	11	92	17	2	9	2	0.5	9
Clarithromycin	14	5	IStd	19	97	9	10	23	2	0.5	25
Climbazole	6	2		17	90	12	4	8	2	0.5	9
Clindamycin	7	2		19	132	53	11	17	2	0.5	20
Clofibric acid	15	5	IStd	19	101	13	34	15	2	0.5	37
Caffeine	12	4	IStd	12	99	8	3	7	2	0.5	8
DEET	58	19	IStd	19	102	7	2	6	2	0.5	6
Diazepam	7	2	IStd	19	93	5	3	6	2	0.5	7
Diclofenac	17	6	IStd	19	98	19	7	9	2	0.5	11
Diuron	7	2	IStd	18	105	7	5	7	2	0.5	9
Erythromycin	22	7	IStd	17	90	12	5	8	2	0.5	10
Fluconazole	6	2	IStd	19	97	9	10	16	2	20.0	28
Hydrochlorothiazide	3	1	IStd	11	100	9	4	6	2	0.5	8
Imidacloprid	15	5		19	77	13	3	10	2	0.5	11
Iopromide	18	6	IStd	13	112	47	23	30	2	0.5	38

Compound	LOQ (ng/L)	LOD (ng/L)	Internal standard	Number of recoveries	Average recovery (%)	Standard deviation (%)	$relU_{bias}$ (%)	$relU_{precision}$ (%)	$relU_{material}$ (%)	$relU_{stability}$ (%)	Utot (%)
Lidocaine	89	30	IStd	19	96	6	18	10	2	0.5	20
MCPA	7	2	IStd	17	99	12	4	5	2	0.5	7
Mefenamic acid	69	23	IStd	19	90	7	3	26	2	0.5	27
Metalaxyl	6	2		19	98	20	10	2	2	0.5	11
Metolachlor	9	3	IStd	19	93	5	5	19	2	0.5	19
Metolachlor OXA	2	1		19	100	9	7	7	2	0.5	10
Metoprolol	12	4	IStd	5	102	6	2	6	2	0.5	7
Metronidazole	11	4		19	95	24	3	10	2	0.5	11
N-Desvenlafaxine	24	8	IStd	5	98	7	7	17	2	0.5	19
N,N-Desvenlafaxine	8	3		5	100	13	14	17	2	0.5	22
N4_Acetyl-Sulfamethazine	2	1		19	90	12	6	49	2	0.5	49
N4_Acetyl-Sulfamethoxazole	14	5	IStd	19	92	15	10	10	2	0.5	14
O,N-Desvenlafaxine	43	14	IStd	5	103	8	9	10	2	0.5	13
Phenazone	10	3	IStd	18	97	11	4	6	2	20.0	21
Prometryn	3	1		16	98	27	7	12	2	0.5	14
Propazine-2-hydroxy	2	1		14	112	21	13	7	2	0.5	15
Sotalol	6	2	IStd	5	100	6	3	7	2	0.5	8
Sucralose	113	38	IStd	5	87	18	15	32	2	0.5	35
Sulfadiazine	30	10	IStd	17	95	13	6	5	2	0.5	8
Sulfamethazine	3	1	IStd	19	96	7	4	6	2	0.5	8
Sulfamethoxazole	33	11	IStd	18	97	9	4	7	2	0.5	8
Sulfapyridine	15	5	IStd	19	94	11	6	6	2	0.5	9
Tramadol	14	5	IStd	5	103	5	4	10	2	0.5	11
Trimetoprim	15	5	IStd	18	105	14	6	24	2	0.5	25
Valsartan	6	2	IStd	5	109	5	10	15	2	0.5	18
Venlafaxine	7	2	IStd	5	109	7	9	8	2	0.5	13

Text S6: Additional details on the mass flow calculations

The concentration of micropollutant *i* in the wastewater entering the HRS between sites *j* and *j-1* was calculated via:

$$c_{WW\ i,(j,j-1)} = \frac{\sum_n (c_{n,i} * Q_{WW,n}) + c_{s,i} * (Q_{WW\ (j,j-1)} - \sum_n Q_{WW,n})}{Q_{WW\ (j,j-1)}} \quad \text{(S 1)}$$

with:

$$c_{n,i} = \overline{c_{eff,n,i}} * (1 - r_{adv,n,i} * f_{adv,n}) \quad \text{(S 2)}$$

n: Sampled WWTP discharging its treated wastewater into the HRS between *j* and *j-1*.

$Q_{WW,n}$: Total discharge of the WWTP *n*.

$c_{n,i}$: Average concentration of micropollutant *i* in the wastewater originating form the WWTP *n*.

$Q_{WW,(j,j-1)}$: Total discharge of wastewater into HRS between sites *j* and *j-1*.

$c_{s,i}$: Average concentration of micropollutant *i* in the wastewater originating form various smaller WWTPs discharging between *j* and *j-1*. It was approximated as the discharge-proportional average of the effluent concentrations of all sampled WWTPs.

$\overline{c_{eff,n,i}}$: Average of the concentrations of micropollutant *i* measured in the samples of the conventionally treated effluent of WWTP *n*.

$f_{adv,n}$: Fraction of the total discharge of the WWTP *n* that is treated by advanced treatment technologies.

$r_{adv,n,i}$: Estimated removal rate of micropollutant *i* in by the advanced treatment technology applied in WWTP *n* based on values reported in literature (6-8).

Additional results

Table S6. Results of the screening with high resolution LC-MS (method details see (9)).

Compound	Metabolite of	LOQ (ng/L)	June 2009, Site 12 (ng/L)	June 2009 Site 7 (ng/L)
Pesticides				
2,4-D		10	35	30
Acetochlor		30	-	-
Alachlor		30	-	-
Aldicarb		20	-	-
Asulam		50	-	-
Atraton		0.1	-	-
Atrazine		0.5	55	66
Azoxystrobin		0.5	-	4
Bentazon		0.1	20	2
Bromazil		0.5	-	-
Bromoxynil		0.1	-	-
Carbetamide		1	-	-
Chloridazon		1	-	-
Chlortoluron		0.2	-	-
Clomazone		0.1	-	-
Cymoxanil		100	-	-
Cyproconazole		1.5	-	-
Cyprodinil		0.5	-	-
Desmedipham		25	-	-
Diazinon		n.b.	-	-
Dicamba		2	-	-
Dichlorprop		0.5	-	-
Diflufenican		5	-	-
Dimethachlor		0.5	-	-
Dimethenamid		0.1	-	-
Dinoseb		40	-	-
Epoxiconazole		2	-	-
Ethofumesate		4	-	-
Fenpropimorph		0.1	-	-
Fipronil		0.4	-	1
Fluazifop		0.5	-	-
Fluazinam		-	-	-
Fludioxonil		0.1	-	-
Flufenacet		0.5	-	-
Fluroxypyr		2	-	-
Flusilazole		5	-	-
Hexazinone		0.1	-	-
Imidacloprid		5	16	79
Ioxynil		0.1	-	-
Isoproturon		0.2	-	-
Kresoxim-methyl		1	-	-
Linuron		1	-	-
MCPA		1	-	-
MCPB		-	-	-
Mecoprop		0.7	-	-
Mesotrione		15	-	-
Metalaxyl		1	-	50
Metamitron		50	-	-
Metazachlor		0.5	-	-
Metolachlor		0.2	-	-

Compound	Metabolite of	LOQ (ng/L)	June 2009, Site 12 (ng/L)	June 2009 Site 7 (ng/L)
Metribuzin		0.5	-	-
Metsulfuron-methyl		1	-	-
Monuron		0.1	-	-
Napropamide		1	-	-
Nicosulfuron		4	-	-
Orbencarb		0.5	-	-
Pethoxamid		0.1	-	-
Phenmedipham		25	-	-
Pinoxaden		-	-	-
Pirimicarb		0.5	-	-
Prochloraz		20	-	-
Prometon		0.1	-	2
Prometryn		0.2	300	14,000
Propachlor		0.1	-	-
Propaquizafop		10	-	-
Prosulfocarb		1	-	-
Pyrimethanil		1	-	-
Rimsulfuron		2	-	-
Simazine		0.5	-	-
Simeton		0.1	-	-
Sulcotrione		10	-	-
Tebuconazole		2	-	-
Tebutam		0.1	-	-
Terbumeton		0.1	-	-
Terbutylazine		0.5	-	-
Thifensulfuron-methyl		2	-	-
Trinexapac-ethyl		2	-	-
Tritosulfuron		-		
Biocide and pesticide metabolites				
2,6-Dichlorbenzamide	Dichlobenil	1	-	-
2-Methyl-4-amino-6-methoxy-s-triazine	Thifensulfuron-methyl	2	-	-
3,5,6-Trichloro-2-pyridinol	Chlorpyrifos	2	-	-
3-Phenoxybenzoic acid	Permethrin	5	-	-
4-Isopropylaniline	Isoproturon	-	-	-
6-Pyrimidinol, 2-isopropyl-4-methyl	Diazinon	50	-	-
Acetochlor ESA	Acetochlor	0.5	11	-
Acetochlor OXA	Acetochlor	0.5	17	-
Alachlor ESA	Alachlor	0.5	11	-
Alachlor OXA	Alachlor	0.5	17	-
Atrazine-2-hydroxy	Atrazine	0.1	13	9
Atrazine-6-desisopropyl	Atrazine	1	-	-
Atrazine-desethyl	Atrazine	0.5	23	39
Atrazine-desethyl-2-hydroxy	Prometon/Atrazin	3	10	23
Benzoic acid, 2-(aminosulfonyl)-, methyl ester	Metsulfuron-methyl Tribenuron	-	-	-
Benzoic acid, 3,5-dibromo-4-hydroxy-	Bromoxynil	2	-	-
Bifenox acid	Bifenox	1	-	-
Chloridazone-desphenyl	Chloridazon	50	-	-
Chloridazone-methyl-desphenyl	Chloridazon	4	-	-
Diazoxon	Diazinon	0.1	-	-
Dimethachlor ESA	Dimethachlor	2.5	-	-
Dimethachlor OXA	Dimethachlor	2.5	-	-
Dimethenamid ESA	Dimethenamid	0.5	-	-
Dimethenamid OXA	Dimethenamid	0.5	-	-

Compound	Metabolite of	LOQ (ng/L)	June 2009, Site 12 (ng/L)	June 2009 Site 7 (ng/L)
Diuron-desdimethyl	Diuron	3	-	-
Diuron-monomethyl (DCPMU)	Diuron	50	-	6
Ethofumesate-2-keto	Ethofumesate	-	-	-
Fipronil-desulfinyl	Fipronil	-	-	-
Fipronil-sulfide	Fipronil	0.5	-	-
Fipronil-sulfone	Fipronil	0.5	-	-
Flufenacet ESA	Flufenacet	1	-	-
Flufenacet OXA	Flufenacet	0.5	-	-
Formamide, N-(2,4-dimethylphenyl)-	Amitraz	15	-	-
Isoproturon-desmethyl	Isoproturon	0.5	-	-
Isoproturon-N-monodemethyl	Isoproturon	0.5	-	-
Mesotrione MNBA	Mesotrion	100	-	-
Metamitron-desamino	Metamitron	0.5	-	-
Metazachlor ESA	Metazachlor	1.5	-	-
Metazachlor OXA	Metazachlor	0.5	-	-
Metolachlor ESA	Metolachlor	1	-	-
Metolachlor OXA	Metolachlor	0.5	-	-
Metolachlor-Morpholinon	Metolachlor	0.5	-	-
Metribuzin-deamino (DA)	Metribuzin	0.2	-	-
Metribuzin-diketo (DK)	Metribuzin	100	-	-
N'-(2,4-Dimethylphenyl)-N-methylformamidine	Amitraz	2	-	-
N,N-Dimethyl-N'-phenylsulphamide (DMSA)	Dichlofluanid	1	-	-
N,N-Dimethyl-N'-p-tolylsulphamide	Tolyfluanid	1	-	-
Propachlor ESA	Propachlor	2	-	-
Propachlor OXA	Propachlor	5	-	-
Propazine-2-hydroxy	Prometon	0.5	140	480
Simazine-2-hydroxy	Simazin	1	-	-
Sulcotrione CMBA	Sulcotrione	40	-	-
Terbutylazine-2-hydroxy	Terbutylazin	0.5	-	-
Terbutylazine-desethyl	Terbutylazine	0.5	-	-
Terbutylazine-desethyl-2-hydroxy	Terbutylazine	0.5	-	-
Trinexapac	Trinexapac-ethyl	-	-	-
2-n-Octyl-4-isothiazolin-3-one (OI)		0.1	-	-
4,5-Dichloro-2-n-octyl-3(2H)-isothiazolone (DCOIT)		n.b.	-	-
Benzisothiazolone (BIT)		-	-	-
Bronopol		-	-	-
Carbendazim		2	510	610
Diuron		1	26	51
Iodopropynyl butylcarbamate (IPBC)		-	-	-
Irgarol 1051		0.1	-	-
Propiconazole		2	-	10
Terbutryn		0.2	-	-
Triclosan		2	23	84
2-Aminobenzimidazole	Carbendazim	0.1	51	88
Irgarol-descyclopropyl	Irgarol	0.1	-	-
Pharmaceuticals				
5-Fluorouracil		-	-	-
Aminopyrine		2	-	-
Antipyrene (Phenazone)		0.5	190	30
Atenolol		0.5	-	-
Azithromycin		2	-	-
Bezafibrate		1	-	-
Carbamazepine		0.5	48	28

Compound	Metabolite of	LOQ (ng/L)	June 2009, Site 12 (ng/L)	June 2009 Site 7 (ng/L)
Cilastatin		-	-	-
Ciprofloxacin		-	-	-
Clarithromycin		1.5	-	-
Climbazol		-	590	1'300
Clindamycin		1	11	21
Clotrimazole		-	-	-
Cyclophosphamide		5	-	-
Cytarabine		3	-	-
Dexamethasone		1	-	-
Diatrizoate		-	-	-
Diazepam		0.5	70	6
Diclofenac		1	-	33
Erythromycin		1	67	200
Exemestane		1	-	-
Fenofibrate		5	-	-
Fluconazole		1	120	69
Fluoxetine		0.5	-	-
Furosemide		10	-	-
Gabapentin		30	-	-
Gemcitabine		150	-	-
Gentamicins		50	-	-
Hydrochlorothiazide		10	13	44
Ibuprofen		2	-	-
Ifosfamide		2	-	-
Indomethacin		2	-	-
Iohexol		-	7'500	710
Iomeprol		-	-	-
Iopamidol		-	-	-
Iopromide		150	390	340
Ioxitalamic acid		-	-	-
Ketoprofen		1	-	-
Lamotrigine		1	14	-
Levetiracetam		5	-	-
Lidocaine		0.1	28	42
Mefenamic acid		1	-	-
Metformin		2	3'100	3'100
Methylprednisolone		5	-	-
Metoprolol		1	18	40
Metronidazole		5	-	-
Moclobemide		0.5	-	-
Morphine		-	-	-
Naproxen		2	-	-
Norfloxacin		-	-	-
Oseltamivir		0.1	-	-
Pantoprazol		-	-	-
Paracetamol		10	-	-
Primidone		5	-	-
Propanolol		0.2	-	-
Ranitidine		2	-	-
Reserpine		-	-	-
Ritonavir		2	-	-
Roxithromycin		0.5	-	-
Sotalol		2	-	-
Sulfadiazine		10	140	170
Sulfadimethoxine		5	-	-
Sulfamethazine		2	-	8

Compound	Metabolite of	LOQ (ng/L)	June 2009, Site 12 (ng/L)	June 2009 Site 7 (ng/L)
Sulfamethoxazole		0.2	520	320
Sulfapyridine		2	-	27
Sulfathiazole		10	-	-
Tramadol		0.5	-	17
Trimethoprim		0.5	27	34
Tylosin		50	-	-
Valsartan		20	-	37
Venlafaxine		0.2	-	-
Verapamil		0.1	-	-
Pharmaceutical metabolites				
2',2'-Difluoro-2'-deoxyuridine	Gemcitabine	-	-	-
4-Acetamidoantipyrine	Metamizole	0.1	1'000	670
4-Aminoantipyrine	Aminopyrine	-	1'300	-
4-Formylaminoantipyrine	Aminopyrine	0.1	950	530
4-(Methylamino)antipyrine	Metamizole	-	-	-
4-Trifluoromethylphenol	Fluoxetine	10	-	-
Atenolol acid (Metoprolol acid)	Atenolol/Metoprolol	0.5	430	270
Atenolol-desisopropyl	Atenolol	150	-	-
Carbamazepine epoxide	Carbamazepine	2	-	-
Carbamazepine-10,11-dihydro-10,11-dihydroxy	Carbamazepine	-	160	130
Clofibric acid	Clofibrate	1	88	23
D617	Verapamil	0.2	-	-
Desvenlafaxine	Venlafaxin	0.5	-	-
Dioxoaminopyrine	Aminopyrine	0.5	570	85
Fenofibric acid	Fenofibrate	0.5	-	-
Iminostilbene	Carbamazepine	5	-	-
N,N-Didesmethylvenlafaxine	Venlafaxin	10	-	-
N,O-Didesmethylvenlafaxine	Venlafaxin	20	-	-
N4-Acetylsulfadiazine	Sulfadiazin	10	22	48
N4-Acetylsulfadimethoxine	Sulfadimethoxin	1	-	-
N4-Acetylsulfamethazine	Sulfamethazin	2.5	-	-
N4-Acetylsulfamethoxazole	Sulfamethoxazole	3	130	100
N4-Acetylsulfathiazole	Sulfathiazol	5	-	-
N-Desmethylvenlafaxine	Venlafaxin	0.5	-	-
Oseltamivir carboxylate	Oseltamivir	2	-	-
Ranitidine-N-oxide	Ranitidine	-	-	-
Ranitidine-S-oxide	Ranitidine	-	-	-
Ritalinic acid	Methylphenidate	1	-	-
Industrial chemicals				
2,7-Naphthalenedisulfonic acid		-	-	-
2-Amino-1,5-naphthalenedisulfonic acid		-	-	-
2-Naphthalenesulfonic acid		10	260	1'100
4,4'-Dinitro-2,2'-stilbenedisulfonic acid (DNS)		-	-	-
4-Amino-N-acetyl-N-methylaniline		100	-	-
Amsonic acid (DAS)		50	-	-
Diglyme		-	-	-
Dinitrostilbenedisulfonic acid		50	-	-
N-Methylacetanilide		2.5	-	-
Sulfanilic acid		-	-	-
Surfynol 104A (TMDD)		30	-	-
Household chemicals				
4-/5-Methyl-1H-benzotriazole		20	1'100	970

Compound	Metabolite of	LOQ (ng/L)	June 2009, Site 12 (ng/L)	June 2009 Site 7 (ng/L)
Benzotriazole		20	1'200	1'700
Acesulfam		-	8'300	6'900
Sucralose		10	2'000	1'100
Benzophenone-3		2	-	-
Galaxolidon	Galaxolid	10	-	-
Caffeine		50	330	2'500
Diethyltoluamide (DEET)		0.1	270	190
Bisphenol A		-	-	-
Benzothiazole		-	-	-

Table S7. Import and export statistics of pharmaceuticals for China in 2009. The difference between the production and the amounts exported can serve as an approximation for the consumption in China (10).

	Production (t)	Export (t)	Difference (t)
Analgesics			
Paracetamol	53336	31108	22228
Metamizole	12586	7240	5346
Adefovir dipivoxil	6428	4003	2425
Aminophenazone	2014	33	1982
Phenacetin	1767	136	1632
Ibuprofen	2828	1813	1015
Benorilate	650		650
Methyl salicylate	400		400
Indometacin	379	67	312
Diclofenac	397	119	278
Naproxen	590	324	266
Mefenamic acid	999	779	220
Propyphenazone	343	274	68
Antibiotics			
Oxytetracycline	20905	5219	15686
Amoxicillin	11095	2030	9065
Lincomycin	3348	1211	2137
Penicilin G Sodium	2070	61	2009
Erythromycin Thiocyanate	2960	1035	1925
Sulfaguanidine	1730		1730
Cephalexin	1590	3	1588
Neomycin sulfate	2406	1198	1207
Metronidazole	1346	205	1141
Moroxydine	999		999
Trimethoprim	2269	1311	959
Amoxicillin chloride	907	82	825
Sulfamethoxazole	1071	290	781
Chloramphenicol	1505	761	745
Tetracycline hydrochloride	3293	2550	743
Cefamezin	865	129	736
Leucomycin	743	11	733
Amantadine	799	79	720
Doxycycline Hyclate	2225	1520	705
Oxytetracyclin-Hydrochlorid	2920	2231	690
Tetracycline	674	10	664
Acetylspiramycin	655	5	650
Norfloxacin	1124	530	594
Sulfamethazine	1806	1244	562
Cefotaxime	592	55	537
Streptomycine	527	7	520
Fosfomycin Sodium	1496	995	501
Ciprofloxacin	957	467	490
Amikacin	618	136	482
Phenoxymethylpenicillin	554	91	463
Gentamicin	595	171	424

	Production (t)	Export (t)	Difference (t)
Procain Benzylpenicillin	1064	675	389
Kanamycin	388		388
Cefoperazone Sodium	387	17	370
Kanamycin Monosulfate	401	55	346
Roxithromycin	339	24	315
Griseofulvin	296		296
Sulfadiazine	1803	1516	287
Berberine	286		286
Ribavirin	316	44	272
Erythromycin Ethylsuccinate	262		262
Azithromycin	336	85	251
Rifamycin	248		248
Rifampicin	419	178	242
Sulbactam + Cefopcrazone	231	0	231
Penicillin G potassium	249	23	226
Levofloxacin	364	153	211
Furazolidone	1174	993	181
Ofloxacin	459	279	180
Polymyxin E	452	272	180
Sulbactam	178	9	168
Ceftazidime	183	21	162
Cefuroxime	164	10	154
Aciclovir	263	116	147
Clindamycin	307	163	144
Clotrimazole	132		132
Piperacillin Sodium	122	3	120
Isoniazid	231	113	118
Pyrazinamide	111		111
Benzathine Penicillin G	178	67	111
Erytromycin	110		110
Meleumycin	107		107
Phenazone	211	104	107
Clarithromycin	104	30	74
Carbamazepine	385	307	78
Diazepam	27	14	13
Primidone	19	18	1
Antiparasitics			
Piperazine	9696	26	9670
Niclosamide	755	170	586
Levamisole	717	152	565
Beta-blocker			
Atenolol	17	0	17
Sotalol	0	0	0
Cardiovascular drugs			
Troxerutin	605	159	446
Lovastatin	205	17	188
Captopril	411	267	143
Methyldopa	283	256	27

	Production (t)	Export (t)	Difference (t)
Other			
Piracetam	2809	1025	1783
Coffein	11924	10246	1678
Aminophylline	510	187	323
Guaifenesin	613	421	192
Theophylline	882	696	186
Chlorzoxazone	163		163
Chorionic gonadotropin	329	179	151
Phenobarbital	296	151	145
Tramadol	17	3	14
Bezafibrat	10	7	3

Table S8. Consumption statistics of pesticides in China in 2009 (11).

	CAS-Number	Consumption (t)
Dichlorvos	62-73-7	40600
Glyphosate	1071-83-6	>10000
Molosultap	52207-48-4	>10000
Acetochlor	34256-82-1	>10000
Thiophanate-methyl	23564-05-8	5000-10000
Carbendazim	10605-21-7	5000-10000
Chlorothalonil	1897-45-6	1000-5000
Isoprothiolane	50512-35-1	1000-5000
Fenaminosulf	140-56-7	1000-5000
Metalaxyl	57837-19	1000-5000
Validamycin	37248-47-8	1000-5000
Pyrimethanil	53112-28-0	1000-5000
Tricyclazole	41814-78-2	1000-5000
Triadimefon	43121-43-3	1000-5000
Paraquat methosulfate	2074-50-2	1000-5000
Trifluralin	1582-09-8	1000-5000
Bentazone	25057-89-0	1000-5000
Atrazine	1912-24-9	>5000
2,4-D	94-75-7	>5000
Diphacinone	82-66-6	674

Table S9. Summary of statistical analysis of the concentrations of all measured substances in the Haihe River System (ng/L).

	Number of valid detects	Number of positive detects	Min	1st Quartile	Median	Mean	3rd Quartile	Max
Household and industrial chemicals								
2-Naphtalene sulphonic acid	49	30	22	89	240	310	370	1700
5-Methyl-benzotriazole	84	82	5	140	230	360	500	2400
Benzotriazole	84	83	360	660	1100	1500	2100	6300
Caffeine	84	57	29	130	320	860	1300	5000
Climbazol	84	83	11	81	310	310	450	1000
DEET	84	84	4	110	140	200	250	590
Sucralose	84	82	300	740	1100	1200	1600	3400
Pesticides								
2,4-D	81	31	30	30	30	50	30	200
Atrazine	84	84	5	17	31	66	67	590
Azoxystrobin	84	15	2	2	6	8	9	22
Bentazon	83	37	8	8	8	20	29	69
Carbendazim	84	84	27	270	390	490	620	1900
Diuron	84	77	11	11	11	25	35	140
Imidacloprid	84	67	9	14	33	59	73	430
MCPA	84	1	96	96	96	96	96	96
Metalaxyl	84	84	5	5	17	25	33	120
Metolachlor	84	31	1	3	3	16	7	190
Prometryn	83	81	19	74	160	460	380	4100
Pesticide metabolites								
2,6-Dichlorbenzamide	84	38	16	60	84	86	110	200
2-Aminobenzimidazole	49	49	8	37	58	75	76	400
Atrazine-2-hydroxy	84	81	4	4	12	16	20	51
Atrazine-6-desiso-propyl	84	13	15	35	61	87	160	200
Atrazine-desethyl	84	78	8	16	24	62	43	520
Atrazine-desethyl-2-hydroxy	76	19	6	10	15	18	24	43
Azoxystrobin acid	84	3	5	10	14	14	18	22
Metolachlor OXA	80	43	6	24	33	41	52	120
Propazine-2-hydroxy	66	64	5	11	23	51	54	660
Pharmaceuticals								
Atenolol	49	14	6	13	19	83	22	940
Bezafibrate	84	24	8	14	21	40	34	170
Carbamazepine	84	84	2	27	50	53	69	160
Clarithromycin	81	62	3	8	20	37	55	180
Clindamycin	83	68	8	8	36	48	72	160
Diazepam	84	44	9	9	22	42	67	140

	Number of valid detects	Number of positive detects	Min	1st Quartile	Median	Mean	3rd Quartile	Max
Diclofenac	75	60	4	12	35	47	80	190
Erythromycin	59	58	3	54	110	130	170	400
Fluconazole	84	84	4	59	79	84	110	200
Hydrochlorothiazide	81	57	8	20	39	82	120	540
Iopromide	82	78	45	220	460	730	870	4000
Lidocaine	75	72	4	24	39	42	56	120
Mefenamic acid	84	28	3	3	5	6	7	19
Metoprolol	49	34	6	26	36	46	58	150
Metronidazole	84	16	12	38	100	100	130	250
Phenazone	84	83	26	56	72	85	100	430
Sotalol	49	19	3	3	7	8	10	19
Sulfadiazine	35	32	15	50	85	91	120	220
Sulfamethazine	75	71	2	7	11	19	22	190
Sulfamethoxazole	84	81	17	97	140	180	240	600
Sulfapyridine	84	59	8	8	23	31	35	210
Tramadol	49	40	7	14	18	18	22	42
Trimethoprim	83	81	8	28	60	82	120	340
Valsartan	49	17	8	11	14	69	61	440
Venlafaxine	49	2	4	5	6	6	8	9
Pharmaceutical metabolites								
4-Acetamidoantipyrine	84	83	270	740	1000	1000	1300	2200
4-Aminoantipyrine	49	5	4	9	9	16	28	31
Atenolol acid	35	35	48	170	320	370	560	730
Carbamazepine epoxide	53	48	2	6	11	11	16	25
Carbamazepine-dihydro-dihydroxy	82	81	53	160	250	260	340	680
Clofibric acid	79	44	6	21	25	32	39	120
N,N-Didesmethyl-Venlafaxine	49	7	22	25	39	49	62	110
N4-Acetyl-Sulfamethazine	84	34	7	7	7	18	21	85
N4-acetyl-Sulfamethoxazole	82	69	22	83	120	130	180	300
N-Desmethyl-Venlafaxine	49	2	15	18	20	20	22	25

Text S7: Significance of observed levels to the aquatic ecosystems

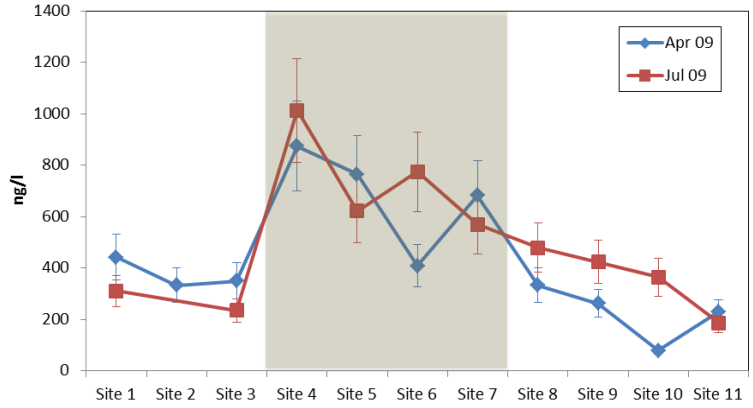
To assess the potential impacts of the observed levels of micropollutants to the aquatic ecosystems of the Haihe River System, the detected levels are compared to Environmental Quality Standards (EQS) as defined by the European Parliament and Council (12) or as suggested by several institutes according to the Technical Guidance Document of the European Union (13).

Table S10. Comparison of the observed levels of micropollutants in the Haihe River to Environmental Quality Standards.

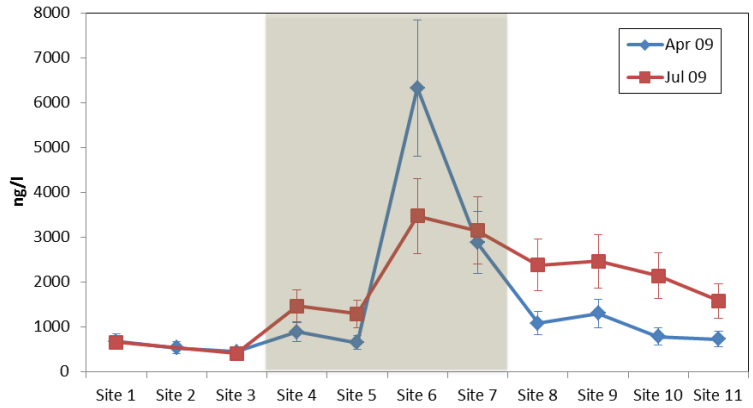
	AA-EQS (ng/L)	Annual average Site 5 (ng/L)	Annual average Site 9 (ng/L)	MAC-EQS (ng/L)	Range Haihe River (ng/L)	Percentage of Samples above MAC-EQS
Household chemicals						
Methyl-benzotriazole	75'000 [b]	320	790	200'000 [b]	<LOD - 2420	0% (n = 84)
Benzotriazole	30'000 [b]	1320	2050	120'000 [b]	<LOD - 6330	0% (n = 84)
Pesticides						
2,4-D	300 [e]	4	9	1'300 [e]	<LOD – 200	0% (n = 81)
Atrazine	600 [a]	36	55	2'000 [a]	3 – 590	0% (n = 84)
Carbendazim	340 [c]	**490**	**540**	560 [c]	27 – 1900	**29%** (n = 84)
Diuron	20 [c]	**48**	**33**	60 [c]	<LOD – 140	**5%** (n = 84)
Imidacolprid	67 [d]	55	59	200 [d]	<LOD – 430	**4%** (n = 84)
Pharmaceuticals						
Atenolol	150'000 [b]	15	22	330'000 [b]	<LOD – 940	0% (n = 49)
Bezafibrate	460 [b]	19	13	76'000 [b]	<LOD – 180	0% (n = 84)
Carbamazepine	500 [b]	40	52	2'550'000 [b]	2 – 160	0% (n = 84)
Clarithromycin	60 [b]	**60**	49	110 [b]	<LOD – 180	**5%** (n = 81)
Diclofenac	50 [b]	**64**	**57**	-	-	-
Mefenamic acid	4'000 [b]	7	3	40'000 [b]	<LOD – 19	0% (n = 84)
Metroprolol	64'000 [b]	55	34	76'000 [b]	<LOD – 150	0% (n = 49)
Sulfamethoxazole	600 [a]	170	170	2'700 [a]	<LOD – 600	0% (n = 84)
Trimethoprim	60'000 [a]	120	120	1'100'000 [a]	<LOD - 340	0% (n = 83)

[a] European Parliament and Council (12), [b] Swiss Centre for Applied Ecotoxicology (14), [c] Junghans and Homazava (15), [d] Posthuma-Doodeman (16), [e] Crane, Maycock et al. (17)

A: Climbazole



B: Benzotriazole



C: Atrazine

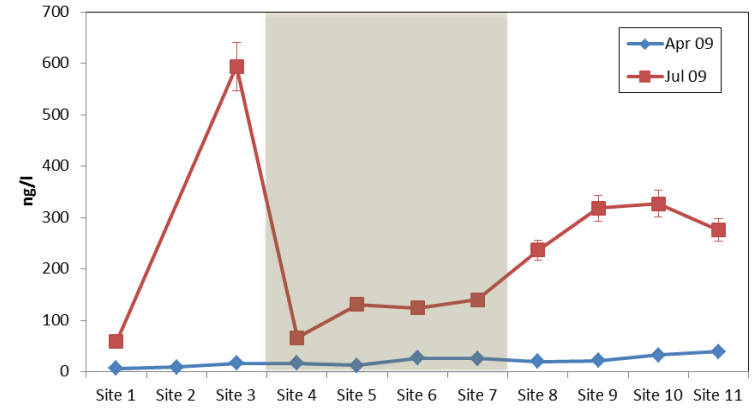


Figure S4. Spatial variation of climbazole, benzotriazole and atrazine concentrations of selected micropollutants along the Haihe River in April and July 2009. The brown shade marks the urban area of Beijing. Within this area, the discharge of Beijing's WWTPs is entering the Haihe River System through the four main tributaries. The error bars mark the uncertainty of the analysis ($relU_{tot}$).

Table S11. Average per capita loads of selected micropollutants in the wastewater of Beijing compared to values from Switzerland. For the loads in Beijing, the values in brackets denote the 0.25 and 0.75 percentile. The per capita loads for Beijing and Switzerland were calculated based on the concentrations in the untreated wastewater, assuming an average water consumption of 200 L/day per person (18, 19).

	Per capita loads (g/day)			
	Beijing		Switzerland	
	5 WWTPs 3 samplings[a]		1 WWTP 11 samplings[b]	17 WWTPs 1-5 samplings[c]
Pesticides				
Atrazine	2	(1.2-2)	7	
Atrazine-desethyl	4	(2-5)	9	
Carbendazim	114	(86-126)	16	6
Diuron	5	(3-5)	16	
Household chemicals				
Benzotriazole	306	(173-310)	2098	2700
5-Methyl-benzotriazole	90	(66-123)	410	
Pharmaceuticals				
Bezafibrate	15	(10-22)	99	76
Carbamazepine	12	(8-16)	120	141
Clarithromycin	28	(16-35)	97	
Clindamycin	<LOQ		14	116
Clofibric acid	<LOQ		3	
Diclofenac	40	(31-49)	347	305
Erythromycin	71	(66-74)	16	
Iopromide	2006	(560-3020)	1057	
Mefenamic acid	3	(2-5)	587	382
N4-Acetylsulfamethoxazole	53	(6-95)	151	
Phenazone	9	(5-12)	8	
Sulfadiazine	60	(52-69)	27	
Sulfamethazine	2	(2-3)	2	
Sulfamethoxazole	62	(34-79)	68	120
Sulfapyridine	42	(36-48)	37	
Trimethoprim	32	(7-57)	38	

[a] this study, based on measurements in the inflow. [b] Hollender, Zimmermann et al. (6), based on concentrations measured in the primary clarifier. [c] Götz, Kase et al. (20), based on concentrations measured in the primary clarifier.

Table S12. Average inflow- and effluent concentrations as well as average removal rates of selected micropollutants for the sampled WWTPs in Beijing. The values in the bracket denote the 0.25 and the 0.75 percentile. Only values for micropollutants that occurred abundantly in the raw wastewater are shown. The removal rates were first calculated for all WWTPs for all three sampling campaigns and then averaged afterwards.

	Average Inflow (ng/L, N=11)		Average Effluent (ng/L, N=12)		Average removal rates	
4-Acetamidoantipyrine	1707	(1443-1997)	1660	(1490-1894)	-4%	(-21-11%)
5-Methyl-benzotriazole	452	(331-615)	347	(291-427)	26%	(13-42%)
Atenolol acid	1006	(894-1151)	504	(278-631)	44%	(25-71%)
Benzotriazole	1531	(864-1550)	448	(328-553)	64%	(59-72%)
Carbamazepine	62	(42-79)	69	(62-71)	-27%	(-56-0%)
Carbamazepine-10,11-dihydro-10,11-dihydroxy	594	(489-663)	456	(431-482)	17%	(4-34%)
Carbendazim	568	(431-629)	477	(390-538)	10%	(6-14%)
Clarithromycin	139	(79-177)	92	(36-119)	26%	(-3-45%)
Climbazol	684	(572-835)	460	(318-569)	27%	(18-48%)
Caffeine	15498	(5528-24377)	290	(35-183)	88%	(90-100%)
DEET	199	(148-223)	76	(16-88)	58%	(45-85%)
Diclofenac	201	(156-244)	164	(134-177)	11%	(-13-32%)
Erythromycin	355	(330-372)	204	(156-265)	35%	(25-40%)
Fluconazole	125	(99-148)	135	(124-148)	-16%	(-39-6%)
Imidacloprid	198	(52-189)	74	(47-81)	17%	(-9-46%)
Iopromide	10028	(2800-15101)	3398	(743-4392)	67%	(49-91%)
Lidocaine	115	(63-144)	99	(61-126)	-8%	(-27-14%)
Sucralose	3294	(2625-3678)	2621	(2437-2805)	11%	(-6-30%)
Sulfadiazine	300	(260-344)	95	(71-122)	67%	(63-75%)
Sulfamethoxazole	308	(169-396)	155	(122-189)	25%	(4-71%)
Sulfapyridine	211	(179-239)	77	(64-87)	61%	(53-68%)

.

Table S13. Average annual loads of micropollutants from July 2009 to June 2010 at sites 1, 5 and 9.

Compound	Site 1 (kg/year)		Site 5 (kg/year)		Site 9 (kg/year)	
Household Chemicals						
2-Naphtalenesulphonic acid	10.8	± 16%	153	± 16%	24	± 14%
5-Methyl-benzotriazole	2.7	± 6%	124	± 6%	596	± 7%
Benzotriazole	19.0	± 10%	516	± 12%	1356	± 10%
Caffeine	15.3	± 10%	659	± 5%	261	± 9%
Climbazol	8.6	± 9%	208	± 8%	268	± 8%
DEET	8.5	± 5%	92	± 5%	112	± 5%
Sucralose	28.8	± 15%	522	± 13%	849	± 13%
Pesticides						
2,4-D	0.1	± 15%	4	± 5%	9	± 5%
Atrazine	0.7	± 7%	16	± 7%	30	± 8%
Azoxystrobin	0.0	± 0%	2	± 17%	2	± 19%
Bentazon	0.1	± 12%	2	± 8%	5	± 12%
Carbendazim	10.5	± 11%	195	± 10%	346	± 9%
Diuron	0.2	± 5%	21	± 7%	21	± 5%
Imidacloprid	0.2	± 28%	22	± 16%	37	± 14%
MCPA	0.0	± 0%	4	± 10%	8	± 10%
Metalaxyl	0.8	± 10%	14	± 11%	20	± 10%
Metolachlor	0.0	± 14%	2	± 11%	1	± 7%
Prometryn	22.1	± 8%	295	± 10%	326	± 11%
Pesticide metabolites						
2,6-Dichlorbenzamide	1.6	± 20%	17	± 23%	21	± 18%
2-Aminobenzimidazole	1.8	± 16%	26	± 18%	39	± 15%
Atrazine-2-hydroxy	0.2	± 7%	6	± 8%	7	± 5%
Atrazine-desisopropyl	0.2	± 16%	6	± 10%	13	± 12%
Atrazine-desethyl	1.3	± 8%	18	± 8%	40	± 8%
Atrazine-desethyl-2-hydroxy	0.1	± 45%	2	± 36%	1	± 35%
Azoxystrobin acid	0.0	± 0%	1	± 23%	2	± 18%
Metolachlor OXA	0.8	± 6%	16	± 7%	19	± 6%
Propazin-2-hydroxy	2.3	± 10%	23	± 13%	22	± 10%
Pharmaceuticals						
Atenolol	0.1	± 12%	3	± 11%	3	± 10%
Bezafibrate	0.1	± 12%	5	± 5%	7	± 6%
Carbamazepine	2.9	± 5%	15	± 5%	34	± 5%
Clarithromycin	0.5	± 10%	22	± 6%	33	± 6%
Clindamycin	0.3	± 32%	24	± 14%	50	± 17%
Diazepam	0.1	± 11%	2	± 6%	3	± 6%
Diclofenac	1.7	± 10%	24	± 6%	44	± 6%
Erythromycin	4.3	± 19%	70	± 12%	129	± 13%
Fluconazole	2.4	± 5%	34	± 4%	76	± 4%
Hydrochlorothiazide	0.4	± 14%	46	± 6%	76	± 6%
Iopromide	9.3	± 11%	504	± 10%	956	± 10%
Lidocaine	0.7	± 5%	19	± 4%	42	± 4%
Mefenamic acid	0.0	± 15%	3	± 6%	2	± 6%
Metoprolol	0.4	± 15%	22	± 9%	18	± 8%
Metronidazole	0.0	± 0%	23	± 12%	59	± 11%
Phenazone	2.6	± 10%	21	± 8%	54	± 8%
Sotalol	0.0	± 0%	3	± 8%	2	± 8%
Sulfadiazine	2.8	± 9%	37	± 7%	78	± 6%
Sulfamethazine	0.3	± 6%	3	± 6%	8	± 6%

Compound	Site 1 (kg/year)		Site 5 (kg/year)		Site 9 (kg/year)	
Sulfamethoxazole	5.4	± 6%	62	± 5%	111	± 5%
Sulfapyridine	0.2	± 10%	17	± 6%	16	± 5%
Tramadol	0.5	± 9%	6	± 10%	11	± 8%
Trimethoprim	2.3	± 16%	44	± 10%	83	± 10%
Valsartan	0.1	± 21%	16	± 17%	3	± 13%
Venlafaxine	0.0	± 0%	0	± 10%	1	± 8%
Pharmaceutical metabolites						
4-Acetamidoantipyrine	37.3	± 15%	445	± 14%	851	± 14%
4-Aminoantipyrine	0.0	± 0%	1	± 31%	1	± 25%
Atenolol acid (Metoprolol acid)	9.1	± 7%	202	± 5%	514	± 5%
Carbamazepine epoxide	0.2	± 7%	6	± 7%	8	± 7%
Carbamazepine-10,11-dihydro-10,11-dihydroxy	6.7	± 8%	114	± 7%	219	± 8%
Clofibric acid	0.1	± 13%	4	± 6%	12	± 6%
N-Desmethylvenlafaxine	0.0	± 0%	1	± 15%	1	± 12%
N,N-Didesmethylvenlafaxine	0.8	± 50%	5	± 49%	0	± 25%
N4-Acetylsulfamethazine	0.3	± 8%	1	± 7%	3	± 7%
N4-Acetylsulfamethoxazole	3.9	± 7%	40	± 7%	94	± 7%

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