

Supplementary material

Mercury loads and fluxes from wastewater: A nationwide survey in Switzerland

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Supplementary Material, Methods

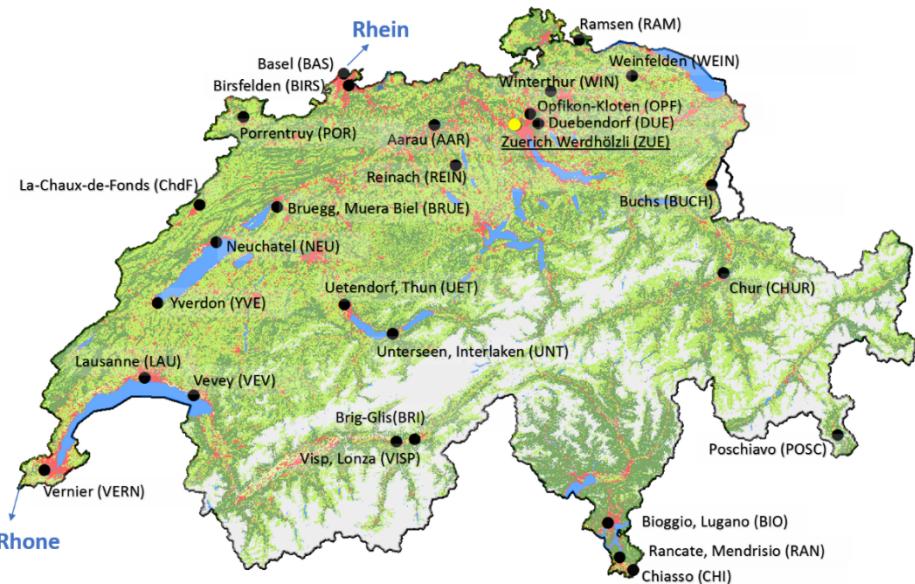


Figure S1. Map of Switzerland showing the locations of the 28 WWTPs studied in 217.

Sample handling. All bottles, vials, containers and pipet tips used in this study were acid washed successively with HCl and HNO₃ (both of Supra-quality, Roth Chemicals, Switzerland). All wastewater samples were collected in borosilicate bottles (250 mL Schott Duran) while the sewage sludge samples were collected in polypropylene (PP) jars (240 ml, Semadeni). Both were double-bagged, as recommended in earlier studies (Amde et al., 2016; Jia et al., 2011) and shipped to the Eawag laboratories within 24 h for sample processing. The liquid samples were stored at 4 °C in the dark and freeze-dried within 48 h. The sewage sludge samples were freeze-dried (LYOVAC GT 2-E, STERIS) immediately after delivery. Then the samples were homogenized by hand with an agate mortar and pestle. Liquid nitrogen was added to some of the samples for better grinding. Borosilicate vials (screw vial amber, EPA40-B, BGB) for Hg samples were pre-cleaned with HCl and subsequently burned in a hot furnace as recommended previously (Amde et al., 2016; Gill and Fitzgerald, 1985; Parker and Bloom, 2005). Acid cleaned PP vials (15 or 50 ml, Cellstar®, Greiner Bio-One GmbH) were used for the other trace elements. All chemicals for Hg acidification, digestion and analyses were Optima-grade (HCl Optima™, ultra trace Class 8, 36.46%, Fisher; Code: A466-500; HNO₃, ultra trace Class 8, 5.1, 63.01%, Fisher; Code: A467-250; H₂O₂ >30%, traceselect®, Sigma-Aldrich). For sewage sludge digestions and rinsing solutions for the ICP-QQQ, suprapure acids were used (HCl 35% and HNO₃ 69%, Supra-quality, Roth Chemicals, Switzerland). Dilutions were made with ultrapure water (MQ, Millipore, 18.2 MΩ).

Microwave digestion To avoid cross contamination in the Teflon digestion tubes (80 ml) an acid cleaning procedure under the same microwave conditions was made between each run. The same program of microwave assisted acid digestion as for the liquid samples was applied to 0.1 g (dry weight (dw)) of each sewage sludge sample. In order to avoid Hg losses due to frothing, aqua regia digests were performed without H₂O₂ (Amde et al., 2016; Lomonte et al., 2008; USEPA, 2007; Veschetti et al., 1998).

Table S1. Matrices and program for microwave assisted digestion of sewage sludge and raw wastewaters.

	Sample	MQ	H ₂ O ₂	HCl	HNO ₃
Raw wastewater	40 mL	-	1 mL	3 mL	1 mL
Sewage sludge	0.1 g	2 mL	-	12 mL	4 mL
Microwave ETHOS 1, MSL GmbH (program: 0-55 min (800 W) 80-110 °C, 55-70 min (100 W) 110-50 °C)					

Analytical methods. THg, dissolved THg was measured with the ICP-QQQ (Agilent 8900) (Table new S2). For comparison THg concentrations of sewage sludge samples were also measured with AMA (LCBIE, Pau). The speciation of the sewage sludge samples was measured with ID-GC-ICP-MS (LCBIE, Pau). Trace and major elements (Li, Na, Mg, Al, Si, Cl, K, Ca, P, S, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Br, Sr, Mo, Cd, Sb, Ba, La, Ce, W, Ti, Pb, Th, U) were measured with ICP-QQQ (Agilent 8900). The amount of total suspended solids (TSS) was determined by gravimetric analysis after filtration (0.2 µm cellulose acetate filter, Satorius) of 20 mL of wastewater samples.

Table S2. Instrumental configuration of the ICP-QQQ (Agilent 8900).

Instrument parameters	Settings / Values
Sample introduction	ISIS
RF power	1550 W
Nebulizer / spray chamber	Micromist / Scott double pass (2 °C)
Cones / lenses	Pt / x-lenses
Carrier / Makeup gas	1.07 / 0.15 L·min ⁻¹
Sampling depth	9 mm
Nebulizer pump speed	0.1 rps
He cell gas	5 mL min ⁻¹
H ₂ cell gas	5.5 mL min ⁻¹
O ₂ cell gas	30 % O ₂ + 1 mL min ⁻¹ H ₂
Monitored masses (m/z)	¹⁷⁵ Lu (+16 in O ₂ mode), ¹⁸⁶ W (+16 in O ₂ mode), ^{201, 202} Hg (on mass)
Integration time	0.5 s for Hg and 0.3 s for Lu and W

Table S3. Background concentration, method detection limits (LOD, 3 σ -criterion) and quantification limit (QL, (3.33*LOD) for total and speciation analyses by ICP-QQQ, AMA and IDL-GC-ICP-MS for speciation.

ICP-QQQ (^{201}Hg He/O ₂)	BEC (ng/L)	LOD (ng/L)	QL _{THg} (ng/L)	MDL _{calc} sludge (ng/g) / liquid digest (ng/L) (digest of 0.1 g sludge, dilution to 50 mL) or liquid digest (40 mL)(ng/L)
Liquids	1.4	0.3	1.0	
SD	0.8	0.0	0.1	
Liquid digests	1.2	1.0	3.5	1.3
SD	0.3	0.3	0.9	
Sludge (freezedried)	1.3	1.3	4.3	0.6
SD	0.7	0.8	2.8	0.4
AMA/IDL-GC-ICP-MS (on freezedried sludge)	LOD THg (ng/g)	QL _{THg} (ng/g)	LOD MMHg (ng Hg/g)	QL _{MMHg} (ng/g)
AVG	7.9	26.4	0.11	0.40
SD	1.1	3.8	0.05	0.16
			IHg (ng Hg/g)	QL _{IHg} (ng/g)
			0.05	0.15
			0.01	0.03

Table S4. Method detection limits (LOD, 3 σ -criterion) for total and speciation analyses by ICP-QQQ, AMA and IDL-GC-ICP-MS for speciation.

ICP-QQQ (^{201}Hg He/O ₂)	LOD THg (ng/L)
Liquids	0.3
SD	0.0
AMA/IDL-GC-ICP-MS (on freezedried sludge)	LOD MMHg (ng Hg/g)
AVG	0.11
SD	0.05

Table S5. Replicate analyses for sewage sludge certified reference materials.

CRM	Target value (ng/g)	ICP-QQQ * (201 Hg He)	SD	n	THg (AMA)** ng/g dw	SD ng/g dw	n	
NIST-2782 Domestic sludge	1100	190	1096	44	4	1087	39	6
BCR-144 Sewage Sludge	1499	220	1327	208	8	1347	34	3
NIST-2781 Industrial sludge	3680	140			3412	476	4	

*The data for ICP-QQQ result from triplicate analyses of the respective references. **The AMA data are single measurements.

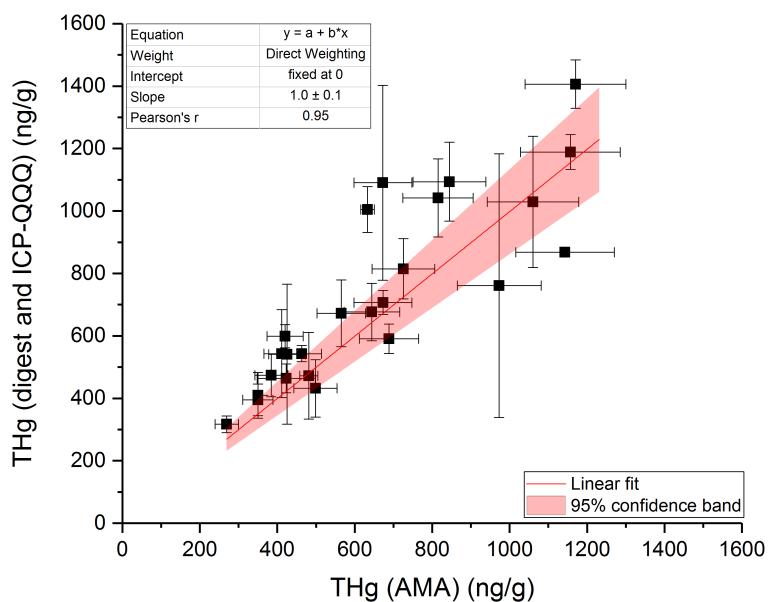


Figure S2. Comparison of total mercury (THg) analyses of 28 samples collected in 20017 by AMA and ICP-QQQ. THg concentrations determined by direct analyses with AMA did not show significant differences to the total concentrations after aqua regia digestion. ICP-QQQ analyses as well as the certified references were analyzed by both techniques (Kolmogorov-Smirnov, Median test sowie paired t-test). The mean recoveries for the certified references were $94 \pm 8\%$ for ICP-QQQ and $94 \pm 5\%$ for AMA.

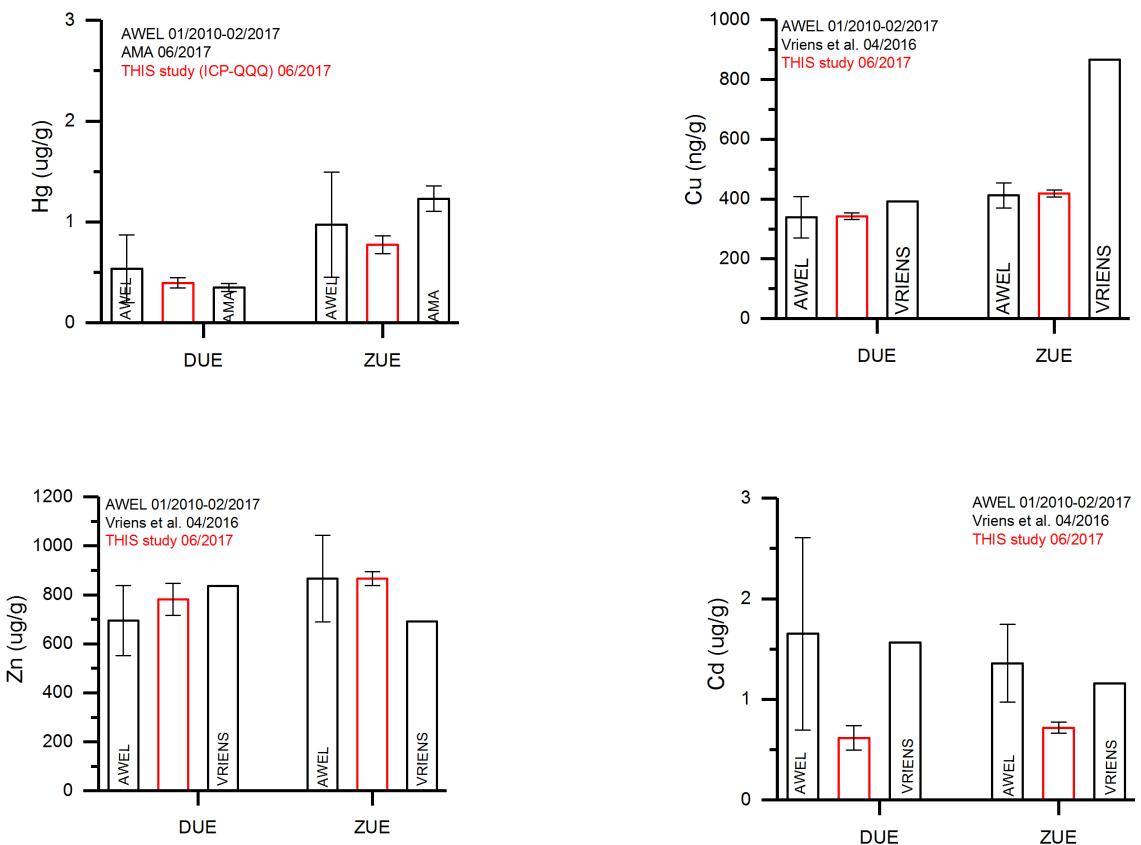


Figure S3. Comparison of THg, Cu, Zn, and Cd concentrations of this study with long-term monitoring data of the Canton of Zurich 2010-2017 (AWEL, 2017) and Vriens et. al. (Vriens et al., 2017).

Supplementary Material, Results and Discussion

Table S6. Overview of THg concentrations (ng/L) reported for raw wastewater and treated wastewater in Switzerland (this study) and WWTPs of other countries.

Country	No. of studied WWTPs	Raw wastewater [THg, ng/L]		Treated wastewater [THg, ng/L]		Removal efficiency (%)	Reference
		range	average	range	average		
Switzerland	28 + 64	9–754	112±159	<LOD–92	8±20	96±4	this study
China, Jiaozuo	1		2000±2700	1.8–43		>90	(Mao et al., 2016)
China	62		3400±2600		160±130		(Liu et al., 2018)
UK	1		92±60	10–60		66.3	(Hargreaves et al., 2016)
Germany, Frankfurt am Main	2		303	<LOD–3.63		99.7	(Fricke et al., 2015)
UK	16		66±55	n.a.		>75	(Gardner et al., 2013)
USA, New York	1	80–860	310±239	24–48		92	(Gbondo-Tugbawa et al., 2010)
USA, St Paul, Minnesota	1	169–376	242	2.05–3.92		98	(Balogh and Nollet, 2008)
Brazil, Sao Paulo	1	0–500	130±90	0–240	50±70	61.5	(Oliveira et al., 2007)
Canada, Winnipeg	3	2–150	61	3–14		88	(Bodaly et al., 1998)

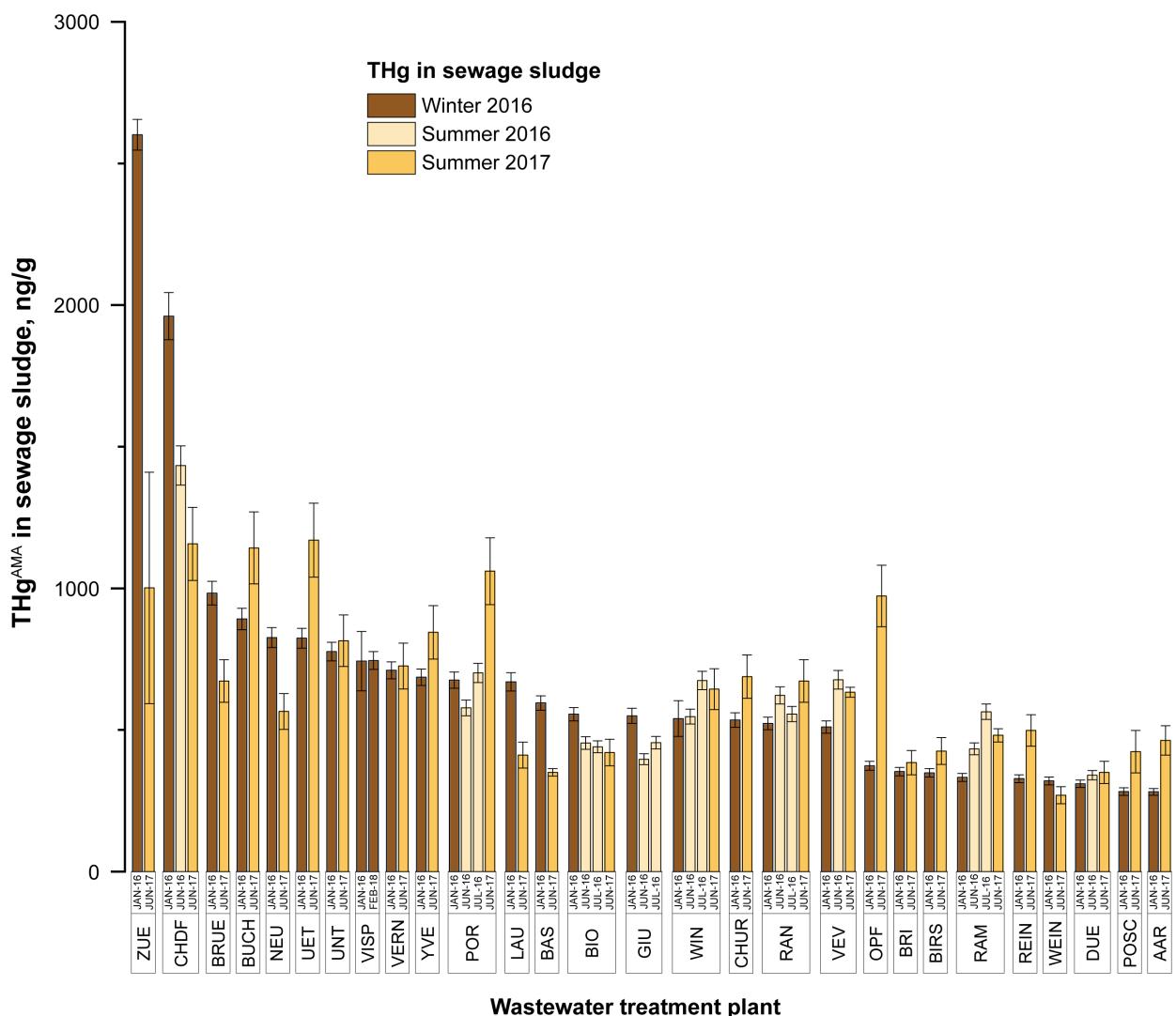


Figure S4. Variability of THg concentrations in sewage sludges collected in the WWTPs studied in 2016 and 2017 (AMA measurements).

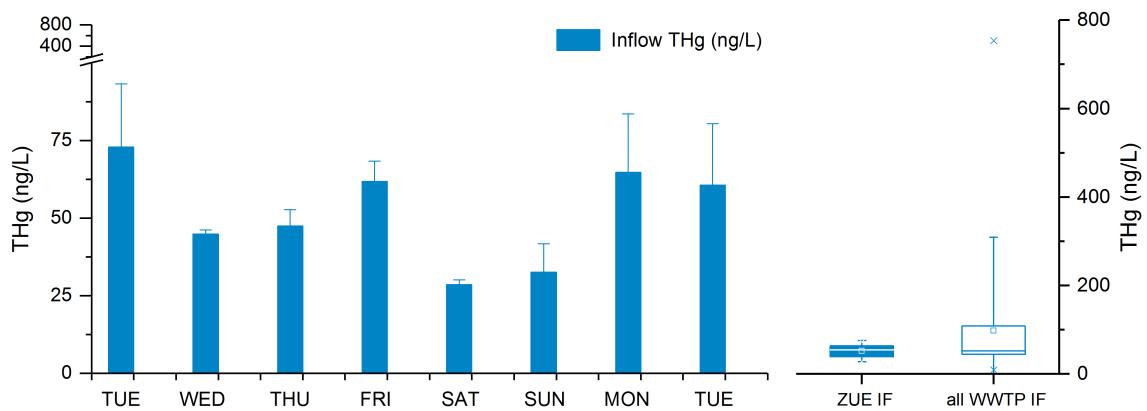


Figure S5. Daily variations in THg concentrations in raw wastewater (inflow) of the WWTP Werdhölzli. The corresponding THg loads ranged from 4 to 12.3 ng/d with 35% variability (percentage of the standard deviation to the mean) showing the lowest concentrations and loads during the weekend. The samples were collected as 24 h composites over one week from Tuesday to Tuesday. Treated wastewater showed concentrations below the detection limit of 0.3 ng/L (quantification limit = $3.33 \times \text{LOD} = 1.1 \text{ ng/L}$). Boxplots on the right show the weekly THg inflow concentrations compared to the combined data from the 28 Swiss WWTPs of this study. The mean is indicated as square, the median as a line within the box and the whiskers indicate a value of $1.5 \times \text{SD}$. The outliers are indicated as star.

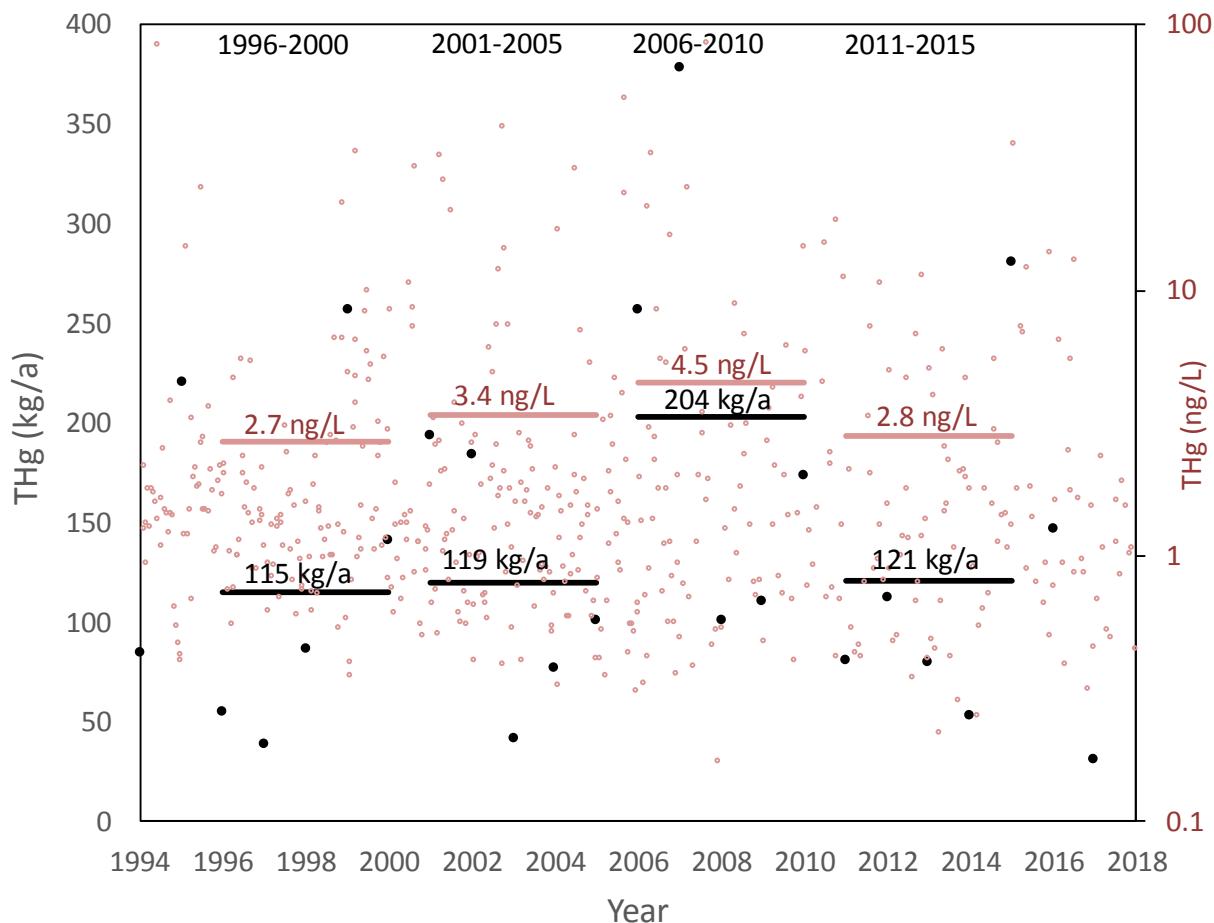


Figure S6. Estimated annual loads (black filled circles, left axis) and THg concentrations (brown empty circles, right logarithmic axis), measured in the river Rhine at the station Weil from 1994-2017 by the Amt für Umwelt und Energie Basel-Stadt (AUE). The THg concentrations represent Hg bound to suspended solids. Concentrations of dissolved Hg were below the detection limits of 5 ng/L (1994-2012) and 2.5 ng/L (2012-2017). Yearly loads were estimated by multiplying the THg concentrations with the water flow at the time of the measurement and the number of days since the last measurement and subsequent summation over each year. 5-year averages of loads and concentrations are shown by black and brown lines. The means and standard deviations of the 5-year averages of loads and concentrations are 140 ± 42 kg/year and 3.35 ± 0.82 ng/L.

Table S7. WWTP information

Selected properties of the investigated wastewater treatment plants.

WWTP No.	WWTP Location*	WWTP Abbr.	Average						Treatment type**	Receiving water	water flow of receiving waters, Q347*** (m3/day)			
			daily waste- water inflow (m3/day)		Average daily sludge production (kg/day)		Connected population equivalents							
			inflow (m3/day)	production (kg/day)	sludge age (days)	population equivalents	Date of sampling							
2 Duebendorf	DUE		22'830	2'559	14	36'232	08.06.2017	Mech-biol with advanced P- elim, nitr and denitr	Glatt	177'785				
3 Winterthur	WIN		57'600	12'600	30	130'110	01.06.2017	Mech-biol with advanced P- elim, nitr and denitr	Töss	153'334				
5 Zurich (Werdhoelzli)	ZUE		219'512	25'712		429'371	05.06.2017	Mech-biol with advanced P-elim, nitr and denitr	Limmat	2'957'049				
8 Unterseen (Interlaken)	UNT		11'500	1'400	30	25'413	07.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Aare	884'779				
9 Opfikon-Kloten	OPF		18'840	2'879	6	37'167	06.06.2017	Mech-biol with advanced P-elim, nitr and denitr	Glatt	243'752				
11 Bruegg (Muera Biel)	BRUE		81'500	9'968	30	85'034	07.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Aare	9'155'514				
13 Uetendorf (Thun)	UET		42'834	5'240		123'223	07.06.2017	Mech-biol with P-elim, nitr and denitr	Aare	2'333'059				
25 Basel	BAS		90'787	36'255		268'317	06.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Rhein	39'116'071				
26 Birsfelden (Birs II)	BIRS		28'000	5'400	15	82'473	02.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Birs - Rhein	40'097'730				
29 Ramsen (Bibertal-Hegau)	RAM		35'274	5'098	35	97'591	20.06.2017	Mech-biol with P-elim, nitr and denitr	Rhein	14'443'194				
34 Buchs	BUCH		10'218	2'210	28	23'913	06.06.2017	Mech-biol with P-elim and nitr	Rhein	2'797'165				
35 Poschiavo	POSC		2'428	483	20	3'271	06.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Poschiavino	87'972				
38 Chur	CHUR		45'360	7'553	18	54'610	08.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Rhein	2'262'565				
40 Aarau	AAR		42'000	6'600	25	74'022	19.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Aare	12'057'837				
45 Reinach (Oberwynental)	REIN		11'500	1'750	25	17'395	20.06.2017	Mech-biol with P-elim, nitr and denitr	Wyna	25'047				
47 Weinfelden (Mittelthurgau)	WEIN		14'700	2'240	11	30'136	06.06.2017	Mech-biol with P-elim, nitr and denitr	Fabrikanal Thur	744'492				
50 Bioggio (Lugano)	BIO		96'000	13'350		124'990	06.06.2017	Mech-biol with advanced P-elim, nitr and denitr	Vedeggio	76'827				
51 Rancate/Mendrisio	RAN		18'000	4'265	25	29'009	07.06.2017	Mech-biol with advanced P-elim, nitr	Laveggio	19'380				
52 Lausanne	LAU		110'000	23'100		235'359	08.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Lac Léman ¹⁾	21'600'000				
55 Vevey/Aviron	VEV		30'000	5'544	35	51'937	06.06.2017	Mech-biol with P-elim	Lac Léman ¹⁾	21'600'000				
56 Yverdon	YVE		10'000	4'465	40	32'842	06.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Lac de Neuchâtel ²⁾	4'771'872				
57 Brig-Glis (Briglina)	BRI		20'000	3'300		27'702	06.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Rhône	326'566				
60 Visp/Lonza	VISP		16'500	18'607		12'985	26.02.2018	Mech-biol with P-elim (simultaneous precipitation)	Rhône	326'566				
61 La-Chaux-de-Fonds	ChdF		10'408	2'271	50	37'455	06.06.2017	Mech-biol with P-elim and nitr	Doubs ³⁾	16'156'800				
62 Neuchatel	NEU		31'000	3'760	39	40'567	07.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Lac de Neuchâtel ²⁾	4'771'872				
63 Vernier/Aire (Geneva)	VERN		175'000	24'500		440'750	06.06.2017	Mech-biol with P-elim, nitr and biofiltration	Rhône	11'911'830				
64 Porrentruy	POR		15'000	1'500		16'209	05.06.2017	Mech-biol with P-elim (simultaneous precipitation)	Allaine	28'901				
65 Vacallo/Chiasso	CHI		15'068	2'438		28'666	16.06.2017	Mech-biol with P-elim	Breggia	6'592				
Statistics			N	28	28	18	28							
			Min	2'428	483	6	3'271							
			Max	219'512	36'255	50	440'750							
			Average	45'781	8'395	26	92'741							
			Stdev			(11)								
			Median	25'415	4'782	27	39'011							
			Sum	1'281'859	235'048		2'596'749							

* WWTP locations are shown in Figure 1, with their corresponding number indicated.

** Abbreviations: Mech-Biol: Mechanical-biological, P-elim: Phosphorus elimination, Nitr: nitrification, Denitr: denitrification.

*** FOEN, Information systems and methods: Q347 flow rate. www.bafu.admin.ch/bafu/de/home/themen/wasser/zustand/wasser--methoden.html

www.bafu.admin.ch/bafu/de/home/themen/wasser/zustand/wasser--methoden.html

1) FOEN, Swiss Federal Office for the Environment, Bern, Switzerland. Der Leman - Zustand bezüglich Wasserqualität, fact sheet July 2016.

2) FOEN, Swiss Federal Office for the Environment, Bern, Switzerland. Der Neuenburgersee - Zustand bezüglich Wasserqualität, fact sheet July 2016.

www.bafu.admin.ch/bafu/en/home/topics/water/info-specialists/state-of-waterbodies/state-of-lakes/water-quality-in-lakes.html

3) République et canton de Neuchâtel, Switzerland. Bassin Du Doubs Neuchâtelois, Rapport pincipale, August 2012.

www.ne.ch/autorites/DDTE/SENE/eaux/Documents/DoubsRapportSynthese.pdf

Table S8. Raw wastewater inflow

Concentrations and loads of mercury (Hg) and total dissolved solids (TSS) in the raw wastewater inflows of the WWTPs investigated in 2017.

WWTP No.	WWTP Location	WWTP Abbr.	THg conc.		Hg-dissolved conc.		percentage dissolved (%)	THg load (g/day)		THg per-capita load (µg/capita/day)		TSS conc. (mg/L)	TSS load (kg/day)
			(ng/L)	SD (ng/L)	(ng/L)	SD (ng/L)		(g/day)	SD (g/day)	(µg/capita/day)	SD		
2 Duebendorf	DUE		23 (3)		1.2 (0.1)		5.2	0.53 (0.07)		14 (2)		103	2'340
3 Winterthur	WIN		61 (7)		2.9 (0.4)		4.8	3.5 (0.40)		27 (3)		150	8'640
5 Zurich (Werdhoelzli)	ZUE		54 (9)		0.7 (0.3)		1.3	11.9 (1.98)		28 (5)		218	47'700
8 Unterseen (Interlaken)	UNT		107 (50)		11 (4.7)		10.3	1.2 (0.58)		48 (23)		205	2'360
9 Opfikon-Kloten	OPF		52 (17)		1.2 (0.3)		2.3	0.98 (0.32)		26 (9)		392	7'390
11 Bruegg (Muera Biel)	BRUE		46 (4)		5.4 (0.2)		11.7	3.7 (0.33)		44 (4)		30	2'440
13 Uetendorf (Thun)	UET		90 (20)		5.1 (0.8)		5.7	3.9 (0.86)		31 (7)		132	5'680
25 Basel	BAS		122 (54)		1.7 (0.1)		1.4	11 (4.90)		41 (18)		313	28'400
26 Birsfelden (Birs II)	BIRS		60 (10)		5.1 (2.1)		8.5	1.7 (0.28)		20 (3)		192	5'390
29 Ramsen (Bibertal-Hegau)	RAM		48 (18)		1.6 (0.1)		3.3	1.7 (0.63)		17 (7)		220	7'760
34 Buchs	BUCH		48 (4)		9.6 (6.7)		20.0	0.49 (0.04)		21 (2)		182	1'860
35 Poschiavo	POSC		21 (4)		2.2 (0.1)		10.5	0.05 (0.01)		16 (3)		60	150
38 Chur	CHUR		122 (44)		2.0 (0.1)		1.6	5.5 (2.00)		100 (37)		343	15'500
40 Aarau	AAR		30 (3)		0.6 (0.3)		2.0	1.3 (0.13)		17 (2)		202	8'500
45 Reinach (Oberwynental)	REIN		27 (2)		0.8 (0.1)		3.0	0.31 (0.02)		18 (1)		177	2'040
47 Weinfelden (Mittelthurgau)	WEIN		9 (1)		0.7 (0.1)		7.8	0.13 (0.01)		4.4 0.5		55	810
50 Bioggio (Lugano)	BIO		44 (6)		1.2 (0.1)		2.7	4.2 (0.58)		34 (5)		102	9'840
51 Rancate/Mendrisio	RAN		46 (7)		0.9 (0.1)		2.0	0.83 (0.13)		29 (4)		180	3'240
52 Lausanne	LAU		118 (8)		0.9 (0.2)		0.8	13 (0.88)		55 (4)		245	27'000
55 Vevey/Aviron	VEV		200 (20)		30 (0.1)		15.0	6.0 (0.60)		120 (12)		115	3'450
56 Yverdon	YVE		81 (46)		6.0 (1.2)		7.4	0.81 (0.46)		25 (14)		205	2'050
57 Brig-Glis (Briglina)	BRI		44 (23)		2.0 (0.5)		4.5	0.88 (0.46)		32 (17)		207	4'150
60 Visp/Lonza	VISP		490 (35)		59 (10)		12.0	8.1 (0.58)		620 (44)			
61 La-Chaux-de-Fonds	ChdF		750 (60)		19 (0.6)		2.5	7.8 (0.62)		210 (17)		277	2'890
62 Neuchatel	NEU		108 (20)		2.1 (0.3)		1.9	3.3 (0.62)		83 (15)		177	5'500
63 Vernier/Aire (Geneva)	VERN		225 (102)		9.2 (0.6)		4.1	39 (17.85)		89 (40)		297	52'100
64 Porrentruy	POR		63 (10)		17 (0.4)		27.0	0.95 (0.15)		58 (9)		67	1'010
65 Vacallo/Chiasso	CHI		43 (6)		5.7 (2.0)		13.3	0.65 (0.09)		23 (3)			
Statistics		N	28		28		28	28		28		26	26
		Min	9		0.6		0.8	0.05		4.4		30	150
		Max	750		59		27	39		620		392	52'100
		Average	110		7.3		6.9	4.8		66		187	9'930
		Stdev	(150)		(12)		(6.3)	7.7		(117)		(91)	(13'755)
		Median	57		2.2		4.6			30		187	4'770
Weighted mean*		104	(28)										

* Weighted mean (= sum of THg loads in raw wastewater/sum of wastewater flow).

Table S9. Treated wastewater outflow

Total mercury (THg) concentrations and loads in the treated wastewater outflows of the WWTPs investigated in 2017.

WWTP No.	WWTP Location	WWTP Abbr.	THg conc. (ng/L)	THg load (mg/day)	THg per-capita load ($\mu\text{g}/\text{capita/day}$)	THg removal efficiency (%)	Estimated THg contribution to receiving waters (level 95% of the time (Q347)) (ng/L)
2	Duebendorf	DUE	0.35 (0.05)	8.0 (1.1)	0.2 (0.03)	98	0.04
3	Winterthur	WIN	0.54 (0.15)	31 (8.6)	0.2 (0.07)	99	0.20
5	Zurich (Werdhoelzli)	ZUE	0.32 (0.02)	70 (4.4)	0.2 (0.01)	99	0.02
8	Unterseen (Interlaken)	UNT	0.75 (0.08)	8.6 (0.9)	0.3 (0.04)	99	0.01
9	Opfikon-Kloten	OPF	0.75 (0.36)	14 (6.8)	0.4 (0.18)	99	0.06
11	Bruegg (Muera Biel)	BRUE	2.3 (0.09)	190 (7.3)	2.2 (0.09)	95	0.02
13	Uetendorf (Thun)	UET	3.5 (0.12)	150 (5.1)	1.2 (0.04)	96	0.06
25	Basel	BAS	5.0 (2.6)	450 (232)	1.7 (0.87)	96	0.01
26	Birsfelden (Birs II)	BIRS	<0.3*	<5.9*	<0.1*	>99.7*	<0.01
29	Ramsen (Bibertal-Hegau)	RAM	1.0 (0.17)	36 (6.0)	0.4 (0.06)	98	<0.01
34	Buchs	BUCH	0.49 (0.20)	5.0 (2.0)	0.2 (0.09)	99	<0.01
35	Poschiavo	POSC	1.4 (0.13)	3.4 (0.3)	1.0 (0.10)	93	0.04
38	Chur	CHUR	0.40 (0.13)	18 (5.9)	0.3 (0.11)	99.7	0.01
40	Aarau	AAR	0.74 (0.03)	31 (1.3)	0.4 (0.02)	98	<0.01
45	Reinach (Oberwynental)	REIN	0.42 (0.17)	4.8 (2.0)	0.3 (0.11)	98	0.19
47	Weinfelden (Mittelthurgau)	WEIN	0.43 (0.05)	6.3 (0.7)	0.2 (0.02)	95	0.01
50	Bioggio (Lugano)	BIO	0.38 (0.12)	36 (12)	0.3 (0.09)	99.1	0.47
51	Rancate/Mendrisio	RAN	0.42 (0.08)	7.6 (1.4)	0.3 (0.05)	99.1	0.39
52	Lausanne	LAU	1.6 (0.03)	180 (3.3)	0.8 (0.01)	99	<0.01
55	Vevey/Aviron	VEV	30 (0.27)	910 (8.1)	18 (0.16)	85	0.04
56	Yverdon	YVE	0.41 (0.06)	4.1 (0.6)	0.1 (0.02)	99.5	<0.01
57	Brig-Glis (Briglina)	BRI	1.7 (0.02)	33 (0.4)	1.2 (0.01)	96	0.10
60	Visp/Lonza	VISP	57 (6.0)	940 (99)	72 (7.6)	88	2.9
61	La-Chaux-de-Fonds	ChdF	92 (6.1)	950 (64)	25 (1.7)	88	0.06
62	Neuchatel	NEU	1.2 (0.07)	37 (2.2)	0.9 (0.05)	99	<0.01
63	Vernier/Aire (Geneva)	VERN	3.3 (0.16)	580 (28)	1.3 (0.06)	99	0.05
64	Porrentruy	POR	8.3 (0.30)	120 (4.5)	7.4 (0.28)	87	4.3
65	Vacallo/Chiasso	CHI	3.7 (0.32)	56 (4.8)	2.0 (0.17)	91	8.5
Statistics		N	28	28	28	28	28
Min		<0.3	3	0.1	85	<0.01	
Max		92	950	72	>99.7	8.5	
Average		7.8	175	5	96	0.6	
Stdev		(20)	(299)	(14)	(4)	(1.8)	
Median		0.9	35	0.4	98	0.04	
Weighted mean**		3.8 (0.4)					

* THg concentration in treated wastewater was below LOD of 0.3 ng/L and was replaced by $\sqrt{(\text{LOD})} = 0.21 \text{ ng/L}$ in the statistics.

** Weighted mean (= sum of THg loads in treated wastewater/sum of wastewater flow).

Table S10. Sewage sludge 2017

Concentrations and loads of inorganic and methylated mercury species (THg, IHg, MMHg) in the sewage sludges investigated in 2017.

WWTP No.	WWTP Location	WWTP Abbr.	THg conc.		IHg conc. percentage of THg			THg load			THg per-capita load			MMHg conc.		MMHg load		MMHg percentage of THg	
			(ng/g)	SD (ng/g)	(ng/g)	SD (ng/g)	(%)	(mg/day)	SD (mg/day)	(µg/capita/day)	SD	(ng/g)	SD (ng/g)	(mg/day)	SD (mg/day)	(%)	(mg/day)	SD (mg/day)	(%)
2 Duebendorf	DUE		390	(50)	278	(3.4)	71	998	(128)	28	(4)	<0.11*		<0.28		<0.28		0.03	
3 Winterthur	WIN		680	(90)	566	(5.7)	83	8'568	(1'134)	66	(9)	2.5	(0.08)	32	(1.0)	32	(1.0)	0.37	
5 Zurich (Werdhoelzli)	ZUE		510	(140)	647	(51)	127	13'113	(3'600)	31	(8)	0.56	(0.78)	14	(20)	14	(20)	0.11	
8 Unterseen (Interlaken)	UNT		1'040	(120)	678	(6.3)	65	1'456	(168)	57	(7)	1.4	(0.15)	2.0	(0.2)	2.0	(0.2)	0.14	
9 Optikon-Kloten	OPF		760	(420)	384	(3.9)	51	2'188	(1'209)	59	(33)	3.0	(0.17)	8.5	(0.5)	8.5	(0.5)	0.39	
11 Bruegg (Muera Biel)	BRUE		1'090	(310)	702	(6.7)	64	10'865	(3'090)	128	(36)	8.6	(0.26)	86	(2.5)	86	(2.5)	0.79	
13 Uetendorf (Thun)	UET		1'400	(80)	1'260	(19)	90	7'336	(419)	60	(3)	3.0	(0.20)	16	(1.0)	16	(1.0)	0.21	
25 Basel	BAS		410	(70)	352	(40)	86	14'865	(2'538)	55	(9)	2.3	(0.55)	85	(20)	85	(20)	0.57	
26 Birsfelden (Birs II)	BIRS		540	(220)	379	(2.9)	70	2'916	(1'188)	35	(14)	1.9	(0.07)	10	(0.4)	10	(0.4)	0.35	
29 Ramsen (Bibertal-Hegau)	RAM		470	(140)	330	(3.8)	70	2'396	(714)	25	(7)	<0.11*		<0.56		<0.56		0.02	
34 Buchs	BUCH		870	(10)	732	(2.7)	84	1'923	(22)	80	(1)	7.2	(0.50)	16	(1.1)	16	(1.1)	0.82	
35 Poschiavo	POSC		460	(50)	446	(1.6)	97	222	(24)	68	(7)	<0.11*		<0.05		<0.05		0.02	
38 Chur	CHUR		590	(50)	362	(4.9)	61	4'456	(378)	82	(7)	2.2	(0.01)	17	(0.1)	17	(0.1)	0.37	
40 Aarau	AAR		540	(30)	465	(6.5)	86	3'564	(198)	48	(3)	<0.11*		<0.73		<0.73		0.02	
45 Reinach (Oberwlynental)	REIN		430	(90)	616	(1.2)	143	753	(158)	43	(9)	3.4	(0.08)	5.9	(0.1)	5.9	(0.1)	0.79	
47 Weinfelden (Mittelthurgau)	WEIN		320	(30)	253	(1.0)	79	717	(67)	24	(2)	<0.11*		<0.25		<0.25		0.03	
50 Bioggio (Lugano)	BIO		600	(40)	528	(2.7)	88	8'010	(534)	64	(4)	<0.11*		<1.47		<1.47		0.02	
51 Rancate/Mendrisio	RAN		710	(40)	701	(3.7)	99	3'028	(171)	104	(6)	<0.11*		<0.47		<0.47		0.02	
52 Lausanne	LAU		540	(140)	411	(5.0)	76	12'474	(3'234)	53	(14)	2.1	(0.17)	50	(3.9)	50	(3.9)	0.40	
55 Vevey/Aviron	VEV		1'000	(70)	769	(4.9)	77	5'544	(388)	107	(7)	0.81	(0.91)	4.5	(5.1)	4.5	(5.1)	0.08	
56 Yverdon	YVE		1'090	(130)	954	(7.9)	88	4'867	(580)	148	(18)	1.7	(0.11)	7.4	(0.5)	7.4	(0.5)	0.15	
57 Brig-Glis (Briglina)	BRI		470	(70)	691	(11)	147	1'551	(231)	56	(8)	<0.11*		<0.36		<0.36		0.02	
60 Visp/Lonza	VISP		740	(100)				13'769	(1'861)	1'060	(143)								
61 La-Chaux-de-Fonds	ChdF		1'190	(60)	967	(14)	81	2'702	(136)	72	(4)	<0.11*		<0.25		<0.25		0.01	
62 Neuchatel	NEU		670	(110)	776	(5.0)	116	2'519	(414)	62	(10)	<0.11*		<0.41		<0.41		0.02	
63 Vernier/Aire (Geneva)	VERN		810	(100)	896	(6.6)	111	19'845	(2'450)	45	(6)	<0.11*		<2.70		<2.70		0.01	
64 Porrentruy	POR		1'030	(210)	815	(12)	79	1'545	(315)	95	(19)	<0.11*		<0.17		<0.17		0.01	
65 Vacallo/Chiasso	CHI		790	(40)				1'926	(98)	67	(3)								
Statistics			N	28		26		26		28		26		26		26		26	
			Min	320		253		51	222		24		<0.11		<0.05		0.01		
			Max	1'400		1'260		147	19'845		1'060		8.6		86		0.82		
			Average	720		610		88	5'504		101		1.6		14		0.22		
			Stdev	(280)		(250)		(24)	(5'229)		(190)		2.2		24		0.27		
			Median	675		631		84	2'972		61		0.7		4		0.09		
			Weighted mean**	656	(108)							1.5	(0.2)						

* MMHg concentration in sludge was below LOD of 0.11 µg/g and was replaced by $\sqrt{LOD} = 0.08 \mu\text{g/g}$ in the statistics.

** Weighted mean (= sum of THg loads in sludge/sum of sludge production).

Table S11. Detailed study of the WWTP Zurich-Werdhoelzli: Temporal variations

Total mercury (THg) concentrations and loads in the inflow and the outflow of the WWTP Zurich-Werdhoelzli over the course of 8 days (15–22 August 2017).

WWTP Zurich-Werdhoelzli	Date of sampling	WW flow (m ³ /day)	Raw wastewater		Raw wastewater		Raw wastewater		Treated wastewater	Treated wastewater	Treated wastewater	THg removal efficiency
			THg conc. (ng/L)	SD (ng/L)	THg load (g/day)	SD (ng/L)	THg per-capita load (g/day)	SD (ng/L)	THg conc. (ng/L)	THg load (g/day)	THg per-capita load (μg/capita/day)	(%)
Day 1	Tue 15.08.17	150'577	73 (20)		11.0 (3.1)		25.6 (7.1)		<0.30*	<0.03	<0.07	>99.6
Day 2	Wed 16.08.17	168'226	45 (1.3)		7.5 (0.2)		17.6 (0.5)		<0.30*	<0.04	<0.08	>99.3
Day 3	Thu 17.08.17	146'438	47 (5.2)		6.9 (0.8)		16.2 (1.8)		<0.30*	<0.03	<0.07	>99.4
Day 4	Fri 18.08.17	179'071	62 (6.5)		11.1 (1.2)		25.8 (2.7)		<0.30*	<0.04	<0.09	>99.5
Day 5	Sat 19.08.17	173'261	29 (1.5)		5.0 (0.3)		11.5 (0.6)		<0.30*	<0.04	<0.08	>99.0
Day 6	Sun 20.08.17	138'893	33 (9.1)		4.5 (1.3)		10.5 (3.0)		<0.30*	<0.03	<0.07	>99.1
Day 7	Mon 21.08.17	148'973	65 (19)		9.6 (2.8)		22.5 (6.5)		<0.30*	<0.03	<0.07	>99.5
Day 8	Tue 22.08.17	148'575	61 (20)		9.0 (2.9)		21.0 (6.9)		<0.30*	<0.03	<0.07	>99.5
Average on weekend	19.+20.08.2017	156'077	31 (2.8)		4.8 (0.4)		11.1 (1.0)		<0.30*	<0.03	<0.08	>99.0
Statistics		N	8	8	8		8		8	8	8	8
Min		138'893	29		5		11		<0.30	<0.03	<0.07	>99.0
Max		179'071	73		11		26		<0.30	<0.04	<0.09	>99.6
Average		156'752	52		8		19		<0.30	<0.03	<0.08	>99.4
Stdev		(14'605)	(16)		(2.5)		(5.9)					(0.2)
Median		149'775	54		8		19		<0.30	<0.03	<0.07	>99.4
Weighted mean**			52						<0.03			

* THg concentration in treated wastewater was below LOD of 0.3 ng/L and was replaced by $\sqrt{LOD} = 0.21$ ng/L in the calculations.

** Weighted mean (= sum of THg loads in raw wastewater/sum of wastewater flow).

Table S12. Detailed study of the WWTP Zurich-Werdhoelzli: Following the treatment stages

THg and MMHg concentrations in wastewater and sludge along the treatment stages of the WWTP Zurich-Werdhoelzli.

(22 August 2017)

Date of sampling	Location along treatment steps	Liquid phase		Sludge		Sludge MMHg conc.		MMHg percentage of THg	
		THg conc. (ng/L)	SD (ng/L)	THg conc. (ng/g)	SD (ng/g)	(ng/g)	SD (ng/g)	(%)	SD (%)
Tue 22.08.17	raw wastewater	52	(16)						
Tue 22.08.17	primary clarifier	22	(2)	416	(28)	1.0	(0.03)	0.2	(0.1)
Tue 22.08.17	bioreactor	103	(3)	163	(4)	0.2		0.1	
Tue 22.08.17	return sludge	203	(56)						
Tue 22.08.17	after secondary clarifier	9	(2)						
Tue 22.08.17	treated wastewater	>99.5	(<0.3)						
Tue 22.08.17	fat from the fat filters			72	(11)	0.6	(0.03)	0.9	(0.3)
Tue 22.08.17	predigestion			479	(132)	1.6	(0.3)	0.3	(0.2)
Tue 22.08.17	digestion			491	(61)	1.1	(0.1)	0.2	(0.2)
Tue 22.08.17	dewatered sludge			465	(40)	1.5	(0.1)	0.3	(0.3)
Tue 22.08.17	return water	6750	(5270)						

* MMHg concentration in sludge was below LOD of 0.11 µg/g.

Table S13. Sewage sludge 2016

THg concentrations and loads in the sewage sludges investigated in 2016.

WWTP No.	WWTP Location	Average daily sludge production (kg/day)	Connected population equivalents	THg conc.* (ng/g)	SD (ng/g)	THg load (mg/day)	SD (mg/day)	THg per-capita load (µg/capita/day)	SD
		(kg/day)		(ng/g)		(mg/day)	SD (mg/day)	(µg/capita/day)	SD
1	Richterswil	900	13'900	160 (8)		144 (7)		10 (0.5)	
2	Duebendorf	855	36'232	310 (13)		265 (11)		7 (0.3)	
3	Winterthur	12'600	112'327	580 (155)		7'308 (1'953)		65 (17)	
4	Dietikon (Limmattal)	4'400	77'656	600 (29)		2'640 (127)		34 (1.6)	
5	Zuerich (Werdhoelzli)	27'500	394'812	2'600 (67)		71'500 (1'834)		181 (4.6)	
6	ARA Region Bern AG	26'950	202'653	570 (28)		15'362 (747)		76 (3.7)	
7	Ittigen (Worbletal)	7'500	63'958	480 (23)		3'600 (175)		56 (2.7)	
8	Unterseen (Interlaken)	2'237	50'727	780 (33)		1'745 (74)		34 (1.5)	
9	Opfikon-Kloten	2'400	30'855	370 (16)		888 (38)		29 (1.2)	
10	Laupen (Sensetal)	5'600	56'894	230 (11)		1'288 (61)		23 (1.1)	
11	Bruegg (Muera Biel)	9'968	74'814	980 (42)		9'769 (416)		131 (5.6)	
12	Innertkirchen	52	1'375	180 (7)		9 (0)		7 (0.3)	
13	Uetendorf (Thun)	5'625	118'000	820 (35)		4'613 (197)		39 (1.7)	
14	Emmen (Buchholz)	9'597	162'369	580 (28)		5'566 (269)		34 (1.7)	
15	Dagmersellen (Oberes Wiggertal)	1'148	37'739	250 (12)		287 (14)		8 (0.4)	
16	Altdorf	2'900	31'600	470 (23)		1'363 (66)		43 (2.1)	
17	Schwyz	2'800	27'548	250 (12)		700 (34)		25 (1.2)	
18	Bilten	3'990	41'639	410 (20)		1'636 (78)		39 (1.9)	
19	Cham (Schoenau)	9'500	124'533	410 (20)		3'895 (188)		31 (1.5)	
20	Ecubens (Vog)	1'280	20'740	270 (13)		346 (16)		17 (0.8)	
21	Vuippens (Ais)	2'549	23'963	340 (16)		867 (42)		36 (1.7)	
22	Fribourg	5'013	40'000	380 (18)		1'905 (91)		48 (2.3)	
23	Winznau (ZV Olten)	9'100	51'320	500 (24)		4'550 (218)		89 (4.3)	
24	Zuchwil	5'730	90'006	400 (19)		2'292 (109)		25 (1.2)	
25	Basel	20'000	246'042	600 (25)		12'000 (506)		49 (2.1)	
26	Birsfelden (Birs II)	5'600	30'680	350 (15)		1'960 (83)		64 (2.7)	
27	Zwingen (Laufental)	3'585	21'630	720 (35)		2'581 (125)		119 (5.8)	
28	Neuhausen (am Rheinfall)	3'041	53'734	530 (25)		1'612 (78)		30 (1.4)	
29	Ramsen (Bibertal-Hegau)	19'000	94'000	330 (14)		6'270 (268)		67 (2.9)	
30	Appenzell (Boedeli)	660	12'894	240 (12)		158 (8)		12 (0.6)	
31	St. Gallen	13'948	37'057	310 (15)		4'324 (209)		117 (5.7)	
32	SG-Hofen (Wittenbach)	6'637	52'006	740 (36)		4'912 (238)		94 (4.6)	
33	Thal/Altrhein	7'000	52'900	270 (13)		1'890 (91)		36 (1.7)	
34	Buchs	1'750	24'000	890 (38)		1'558 (66)		65 (2.8)	
35	Poschiavo	483	3'320	280 (14)		135 (7)		41 (2.0)	
36	Scoul (Sot Ruinas)	432	8'507	300 (14)		130 (6)		15 (0.7)	
37	Davos	600	10'577	460 (22)		276 (13)		26 (1.2)	
38	Chur	7'553	43'717	530 (26)		4'003 (195)		92 (4.5)	
39	Disentis/Muster	787	3'799	160 (8)		126 (6)		33 (1.6)	
40	Aarau	6'600	70'000	280 (12)		1'848 (79)		26 (1.1)	
41	Turgi (Baden)	4'050	54'133	350 (17)		1'418 (69)		26 (1.3)	
42	Unterehrendingen	700	10'299	290 (14)		203 (10)		20 (1.0)	
43	Wohlen	1'550	28'800	580 (25)		899 (38)		31 (1.3)	
44	Windisch (Brugg)	2'400	29'806	510 (22)		1'224 (52)		41 (1.7)	
45	Reinach (Oberwynental)	1'750	17'395	330 (14)		578 (24)		33 (1.4)	
46	Kaisten	120	20'633	240 (11)		29 (1.4)		1 (0.1)	
47	Winfelden (Mittelthurgau)	2'240	29'500	320 (14)		717 (30)		24 (1.0)	
48	Giubiasco (Bellinzona)	7'700	50'000	550 (27)		4'235 (204)		85 (4.1)	
49	Locarno (Maggia-CDL)	3'350	40'914	320 (15)		1'072 (52)		26 (1.3)	
50	Bioggio (Lugano)	13'350	92'097	560 (24)		7'476 (315)		81 (3.4)	
51	Rancate/Mendrisio	4'265	24'958	520 (22)		2'218 (95)		89 (3.8)	
52	Lausanne	23'100	204'080	670 (32)		15'477 (747)		76 (3.7)	
53	Gland VD	1'150	33'079	430 (21)		495 (24)		15 (0.7)	
54	Nyon	1'900	23'026	540 (23)		1'026 (44)		45 (1.9)	
55	Vevey/Aviron	5'544	42'951	510 (22)		2'827 (120)		66 (2.8)	
56	Yverdon	3'200	10'000	690 (29)		2'208 (93)		221 (9.3)	
57	Brig-Glis (Briglina)	3'300	25'129	350 (15)		1'155 (49)		46 (2.0)	
58	Leuk (Radet)	1'790	13'034	320 (15)		573 (28)		44 (2.1)	
59	Evionnaz	350	3'744	350 (17)		123 (6)		33 (1.6)	
60	Visp (Lonza)	18'607	11'361	750 (32)		13'955 (589)		1'228 (52)	
61	La-Chaux-de-Fonds	2'946	40'000	1'960 (83)		5'774 (245)		144 (6.1)	
62	Neuchatel	3'760	38'345	830 (35)		3'121 (132)		81 (3.4)	
63	Vernier/Aire	24'500	425'000	710 (30)		17'395 (739)		41 (1.7)	
64	Porrentruy	1'400	17'500	680 (29)		952 (40)		54 (2.3)	
Statistics	N	64	64	64		64		64	
	Min	52	1'375	160		9		1.4	
	Max	27'500	425'000	2'600		71'500		1'228	
	Average	6'108	61'505	520		4'242		71	
	Stdev	(6'833)	(80'040)	(380)		(9'446)		(153)	
	Median	3'468	37'398	445		1'690		40	
	Weighted mean**			694 (32)					

* THg concentrations in sludge samples from 2016 were determined by direct combustion with an advanced mercury analyzer (AMA).

** Weighted mean (= sum of THg loads in sludge/sum of sludge production).

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