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Investigations on the influence of short-term microplastics exposure on soluble micropollutants in municipal wastewater treatment plants

Michael Sturm, Dennis Schober, Katrin Schuhen

Wasser 3.0 / abcr GmbH

Abstract

The water entering sewage treatment plants increasingly poses challenges for operators. More and more often, there is a growing amount of microplastic in rainwater and wastewater for example due to fibers in synthetic clothing such as fleece jackets, tire abrasion or due to the input from industrial processes. The influences and effects of microplastic particles on the environment and humans have not yet been fully scientifically investigated. In a concept for removing microplastic particles from water presented in 2016, Herbort and Schuhen describe a new, adaptive and at the same time modular approach (agglomeration fixation process, AFP). During the pilot trials in a municipal wastewater treatment plant in Germany, an additional spiking with microplastic particles was used to test the process on large scale. In addition, the influence of this short-term microplastic exposure on dissolved micropollutants in municipal wastewater treatment plants was investigated. The results are presented in this publication.

Introduction

The contamination with micropollutants, which in addition to pharmaceuticals, pharmaceutical residues and pesticides also include plastics or microplastics, is currently not regulated on a case-by-case basis for municipal sewage treatment plants, although the statutory environmental quality standards are already being exceeded in many waters (Harmancioglu, Ozkul and Alpaslan 1998; Kim and Zoh 2016). For the reduction of dissolved micropollutants, both source-oriented and aftercare measures are available, each of which has its own specific advantages and disadvantages (ICPR 2012; Loucks and van Beek 2017; ICPR 2019). However, the measures have been developed to varying degrees and are currently far from being an economic standard solution. While the techniques for a 4th purification stage (e.g. use of activated charcoal, membrane filtration or ozone) are used for follow-up measures against dissolved micropollutants, but these have manifold limitations, the situation in the field of microplastics (plastic particles smaller than 5 mm) is hardly considered, especially in the area of the 4th purification stage (Rudloff *et al.* 2018).

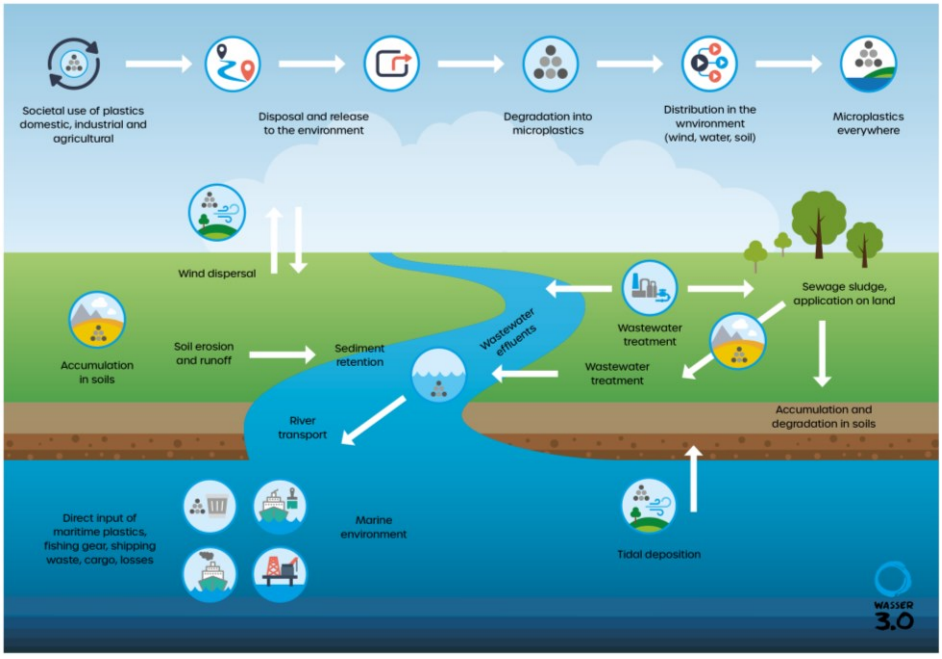

Fig. 1 Distribution ways of microplastics in the environment (C) Wasser 3.0

Increased microplastic contamination has already been detected at the outlet points of municipal sewage treatment plants (Browne *et al.* 2011; Mani *et al.* 2015). Also industrial wastewater treatment plants, especially from the plastic industries, can release microplastics into the environment (Lechner *et al.* 2014; Lechner and Ramler 2015). Primary and secondary microplastics (MP) enter the sewage treatment plants via wastewater. Within the clarification process a large proportion of the particles (usually particles with densities > 1 g/cm³) are transferred into the sewage sludge (Table 1).

The removal of most of microplastics can be explained by the specific behavior of the microplastics load within the clarification process as a function of particle size and density (Sturm and Schuhen 2019). However, the results of the current studies do not provide reliable and reproducible data and very diverse removal rates for microplastics within the three purification stages of the clarification process (Talvitie et al.

2017a, Mintenig et al. 2016, EUWID 2019, Murphy et al. 2016). Analytical standard methods are also lacking so far (Rocha-Santos and Duarte 2015).

In principle, plastic particles can be discharged into the environment from the wastewater via overflows in the sewer system (mixed water discharge), untreated water from the separating sewer system, residual contents in the treated wastewater and material sewage sludge recycling (Browne *et al.* 2011; Napper and Thompson 2016; Sturm and Schuhen 2019). The environmental behavior of microplastic particles has also not yet been clearly clarified. Various studies show that microplastics are able to adsorb and transport organic trace substances and heavy metals from water (Bakir, Rowland and Thompson 2012; Ziccardi *et al.* 2016). Thus, it can interact with the micropollutants in wastewater and act as transport vector. Additionally additives such as plasticizers or flame retardants can be released into the water via microplastics (Bergmann, Gutow and Klages 2015; Hahladakis *et al.* 2018). According to the current state of research, microplastics represents a burden on the environment that cannot be disregarded. (Mason *et al.* 2016; Talvitie *et al.* 2017b; Talvitie *et al.* 2017a). The effects and influence of microplastic particles on the concentrations of dissolved micropollutants within short-term exposures have not been investigated in real environments of a wastewater treatment plant. This publication closes this gap.

Tab. 1: Summary of various studies on the monitoring of microplastics in sewage sludge and wastewater.

Country	Sewage sludge [MP / kg dry]	Influent [MP / l]	Effluent [MP / l]	Removal [%]	Sources
Netherlands			9-91		(Leslie, et al. 2013)
Sweden	16.7 ± 1.96 x10^3	15.1±0.89·	8.3 ± 0.9 x10^{-3}	> 99,9	(Magnusson and Nóren 2014)
Finland		627	33	95	(Heinonen and Talvitie 2014)
Germany	1 – 24 x10^3		0,26-13,7		(Mintenig *et al.* 2016)
USA			0,004-0,2		(Mason *et al.* 2016)
USA		133	5,9	95,6	(Michielssen *et al.* 2016)
Denmark		18.3-2.2 x10^3	29-447	99,3	(Simon, et al. 2018)
China	22.7 ± 12.1 × 10^3				(Li *et al.* 2018)

Material and Methods

Chemicals

For the agglomeration of the microplastic, abcr eco Wasser 3.0 PE-X® wastewater (AB 930003) was acquired from abcr GmbH, Karlsruhe, Germany. This is an organosilane-based mixture of different chemicals, which was specially developed for the agglomeration of microplastics in water. The microplastic used is untreated plastic granulate from the plastics industry. Polyethylene (PE) and polypropylene (PP) were purchased from Lyondellbasell Rotterdam, Netherlands, and copolyester (CoPES) from EMS-Grilltech, Domat/Ems, Switzerland.

Technology

The reactor for microplastic removal is a fully automated pilot plant, which was developed by Zahnen Technik GmbH Arzfeld, Germany in cooperation with Wasser 3.0, Karlsruhe, Germany and installed in an ISO container. The core is a stainless steel cylinder with a maximum capacity of 1.2 m^3, which is equipped with a special agitator that can effectively collect plastic particles suspended in water. The cylinder can be filled and emptied by centrifugal pumps. The dosing of the agglomeration reagent is automated by a dosing pump (Pominent Gamma X). The entire pilot plant is controlled centrally via a control unit.

Particle counting via FlowCam®

For particle counting a FlowCam® 8400 was used, which is equipped with a 300 µm flow cell and a 4x objective. The minimum particle size was set to 20 µm. A sample volume of 10 ml was measured at a flow rate of 3 ml/min and a frame rate of 21.82 fps. The data were analyzed with the VisualSpreadsheet 4.10.8 software.

Sampling for particle counting is performed with a 20 ml syringe with which a sample was taken from a valve in the middle height of the reactor with the agitator switched on. The sampling and measurements were carried out five times.

Tab. 2: Measured wastewater parameters and test methods

Wastewater parameter	Test method
pH-value	DIN EN ISO 10523:2012-04
Measuring temperature pH value	DIN 38404-4:1976-12
Electrical conductivity (25°C)	DIN EN 27888:1993-11
TOC	DIN EN 1484:1997-08
DOC	DIN EN 1484:1997-08
CSB	DIN 38409-41:1980-12
BSB5	DIN EN 1899-2:1998-05
Ammonium-N	DIN 38406-5:1983-10
Nitrite-N	DIN EN 26777:1993-04
Nitrate-N	DIN EN ISO 10304-1:2009-07
Total inorganic nitrogen	Calculated

Determination of further water parameters

Table 2 lists the other water parameters determined by Limbach Analytics GmbH, Mannheim, Germany.

In addition

- **Heavy metals:** lead, cadmium, chrome, total copper, nickel, zinc: DIN EN ISO 17294-2:2005-02. mercury: DIN EN 1483:2007-07
- **Phosphorous, total:** DIN EN ISO 17294-2:2005-02
- **Pharmaceutical residues:** 10,11-Dihydro-10,11-dihydroxy carbamazepine, Azithromycin, Bezafibrate, Candesartan, Carbamazepine, Ciprofloxacin, Clarithromycin, Dehydrato-

Erythromycin A (= anhydroerythromycin), Diclofenac, Erythromycin A, Gabapentin, Hydrochlorothiazide, Ibuprofen, Irbesartan, Metoprolol Sulfamethoxazole: LAM-MLC.M.0051; Metformin, Guanylurea LAM-MLC.M.0051 / Hypercarb.

- **Radiocontrast agents:** Amidotrizoic acid / diatrizoate, Iohexol, Iomeprol, Iopamidol, Iopromide: LAM-MLC.M.0051 / LV.
- **Pesticides:** Carbendazim, DEET, Mecoprop, Terbutryn: DIN 38407-35:2010-10.
- **Corrosion inhibitors:** Benzotriazole LAM-MLC.M.0050 Σ 4- and 5-Methylbenzotriazole LAM-MLC.M.0050.
- **Chelating agents:** DTPA, EDTA, NTA: DIN EN ISO 16588:2004-02.
- **Other chemicals:** Melamine: LAM-MLC.M.0051.
- **Perfluorinated surfactants:** PFBA, PFBS, PFOA, PFOS: DIN 38407-42:2011-03.
- **Synthetic fragrances:** AHTN, HHCB: LAM-MGC.M.0003.
- **Flame retardants:** TCEP, TCPP: LAM-MGC.M.0003.
- **Artificial sweeteners:** Acesulfame, Cyclamate: LAM-MLC.M.0050, Sucralose LAM-MLC.M.0050/LV.

were determined.

Experimental procedure:

The tests were carried out in a municipal sewage treatment plant with three purification stages in southwest Germany. The reactor was filled with 1.2 m³ purified wastewater from the secondary sedimentation tank effluent. A defined amount of microplastic (spiking) was then added.

The advantage of microplastic spiking is that the removal efficiency can be determined directly during the experiment by a simple particle count using FlowCam®.

In experiment 1 1.5g PE and 1.5g CoPES were added. In experiment 2 and 3 1.5g CoPES, 2.5g PE and 2.5g PP were added. After the addition of the microplastic, the removal process was started.

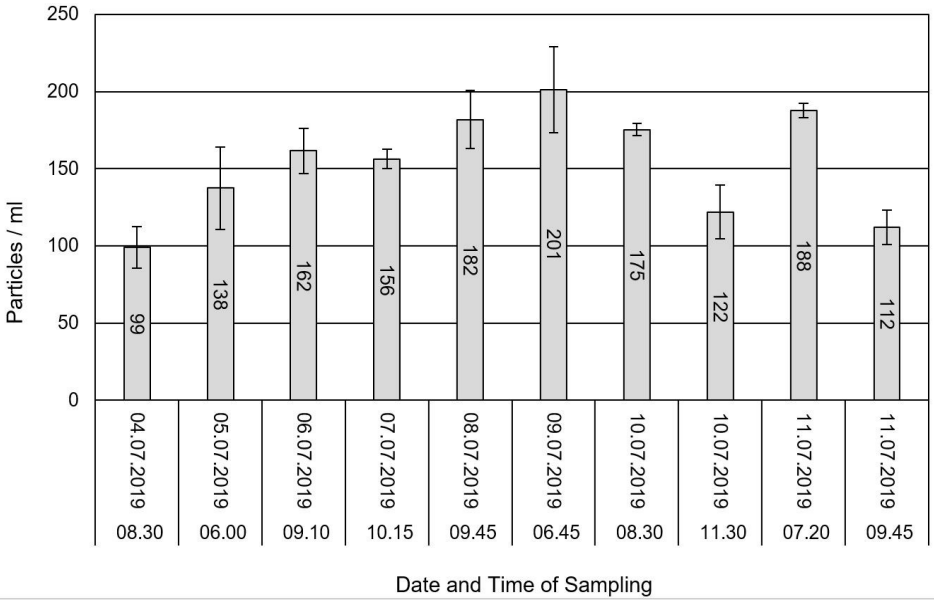

Fig.2: Weekly course of the total particle load (microplastics and natural organic materials) in the treated wastewater of the effluent secondary sedimentation tank.

The added amount of Wasser 3.0 PE-X® was 3 ml per batch. The test lasted until the agglomerates were removed T = 150 sec. The agglomerates are continuously removed by means of a removal technique.

The samples for the water parameters were taken after filling the reactor and after completion of the removal process. In addition, samples were taken during the pilot week (04-Jul-2019 – 11-Jul-2019) for particle load and general water parameters.

Results and discussion

Wastewater parameters and particle contamination over the course of a week

The particle load in the effluent of the wastewater treatment plant (Figure 2) shows a clear variance between the different samples and ranges between 99 and 201 particles/ml on average. The measured values from 10-Jul-2019 and 11-Jul-2019 show that there are also clear differences in the particle load when sampling on the same day. The particle contamination is due to residues of the organic substance degraded in the clarification process and microorganisms such as algae and microplastics.

In a sampling of several German sewage treatment plants, microplastic concentrations of 0.26 - 13.7 microplastic particles (MP) / liter (l) were found in the sewage treatment plant effluent (Mintenig *et al.* 2016). The comparison with the total particle load shows that the load with natural particles from the clarification process exceeds that with microplastics by several orders of magnitude.

Calculated for the capacity of the reactor of the pilot plant of 1.2 m³, this would result in a number of 312 - 16,440 microplastic particles per test run. This high background contamination with natural particles is the reason why the selective analysis of microplastics in environmental and wastewater samples is so far very complex and not suitable for continuous process control within the purification stages of the sewage treatment plant.

In order to distinguish microplastics from natural particles, the number of natural particles must first be greatly reduced in the first processing step by density separation and selective decomposition of natural organic particles (Rocha-Santos and Duarte 2015; Mintenig *et al.* 2016). Since not all natural particles can be removed here, a selective analysis with chemical characterization of the remaining

particles is required to detect specific plastic particles. In addition, special precautions are required to reduce contamination with microplastics from ambient air or laboratory equipment and materials during sampling and processing to a minimum. The analysis of microplastics is therefore very time-consuming, error-prone and requires special measuring instruments which are not available at many research institutions. For this reason, we decided to work with microplastic spiking to test the functionality of the process on large scale and to additionally evaluate the influence of microplastics on soluble organic chemical compounds.

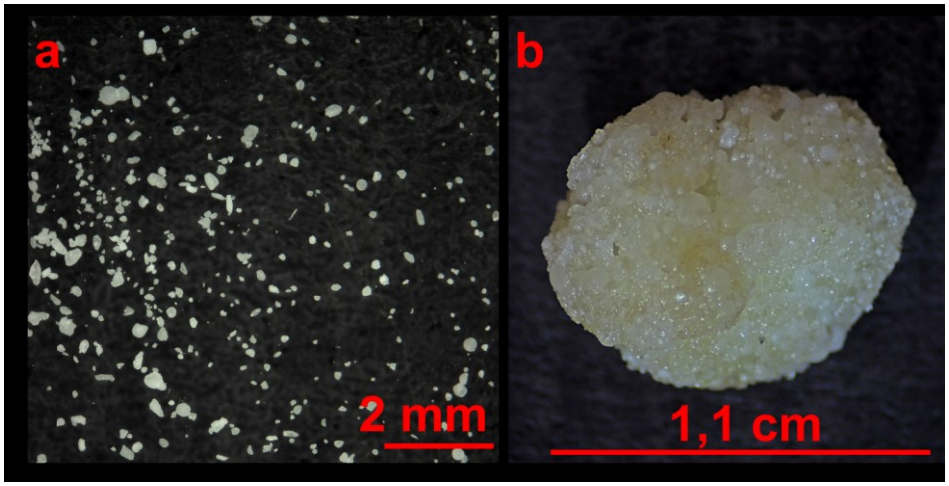

Fig.3: Stereolupe picture of (a) a mixture of microplastic particles (PE, PP and CoPES 1:1:1); (b) an agglomerate of microplastics and Wasser 3.0 PE-X® after removal process.

Agglomeration fixation process for microplastic elimination

After addition of the microplastics (Figure 3a), PE and PP floats up, CoPEs sink due to the higher density. After switching on the agitator, the microplastic is first distributed in the water and then collected. After the addition of Wasser 3.0 PE-X®, the first agglomerates form within a few seconds. During the further 150 seconds of stirring, they increase in size and absorb the free microplastic. After the stirrer is switched off, numerous agglomerates in the size range from 1 to 3 cm float up. These agglomerates are continuously removed during the process (Figure 3b). The physical-chemical processes taking place during the agglomeration fixation were described in previous studies (Herbort and Schuhen 2016; Herbort *et al.* 2018; Herbort, Sturm and Schuhen 2018).

Investigation of changes in the load values of dissolved organic-chemical micropollutants before and after microplastic elimination

The other wastewater parameters measured (Table 4, see appendix) show no noticeable fluctuations over the course of the week. Short-term spiking of the wastewater sample with microplastic particles has no significant influence. The general wastewater parameters are typical for purified municipal wastewater. The pH value is in the neutral range and the conductivity is not significantly increased with 792-861 µS/cm. The values of TOC (7.7 - 9.3 mg/l), DOC (6.9 - 7.6 mg/l) and COD (19 - 25 mg/l) indicate a good purification performance of the sewage treatment plant. The values for nitrogen and phosphorus are also well below the limits applicable in Germany (AbwV 2004). Wastewater decomposition shows that there is no heavy metal contamination. The pharmaceutical residues are within the normal range for purified municipal wastewater (Sacher 2014; Rößler, Metzger and Rau 2018).

Only guanylurea, with values between 47-69 µg/l, is slightly elevated. Radio-contrast agents were detected in all samples and are presumably being introduced by two hospitals in the catchment area. These are the only substance classes that show significant variations between the different samples. As the catchment area is characterized by a high level of economic activity, pesticides can also be found in all samples. Corrosion inhibitors, chelating agents, synthetic fragrances, flame-retardants and artificial sweeteners are found in a typical range for purified municipal wastewater and show no abnormalities. Perfluorinated surfactants could not be detected in any of the samples.

The consideration of the other water parameters (Table 3 and Table 4, see appendix) before and after microplastic removal does not distinguish any regular differences. The differences in the general water parameters and organic trace substances are within the range of normal measurement fluctuations. All heavy metals were below the detection limit.

The parameters that appear to be increased after the removal of microplastics are TOC (inlet 7.7-8.7 mg/l; outlet 8.3-8.8 mg/l), DOC (inlet 6.9-7.5 mg/l; outlet 6.7-7.4 mg/l) and COD (inlet 19-20 mg/l; outlet 22-25 mg/l). However, since the differences are in the range of the measurement fluctuations and therefore cannot be attributed directly to the microplastic removal process and influence of Wasser 3.0 PE-X®.

Thus, the removal process and the added microplastics have no measurable effect on the wastewater parameters and the dissolved organic chemical micropollutants contained. Different laboratories studies suggest time frames of 24 h up to several weeks for reaching the equilibrium concentration (Teuten *et al.* 2007; Velzeboer, Kwadijk, C J A F and Koelmans 2014; Zhan *et al.* 2016). Despite the high microplastic concentration, the short contact time avoids a detectable sorption of micropollutants to the microplastics.

Conclusion

(1) The results show that the sewage treatment plant investigated has a good purification performance and a typical load of organic trace substances in the wastewater.

(2) The contact with microplastics and the experiments on microplastic elimination had no effect on the contamination of the wastewater with organic trace substances, as the contact time was too short to adsorb them from the water.

(3) The agglomeration fixation process for the removal of microplastics from wastewater could be successfully performed in fully automated pilot plant scale.

(4) High load of natural particles in the effluent of the secondary sedimentation tank exceed microplastics concentrations by sveral orers of magnitude wherby a comlex and time intesive analyticy for seletice detection of microplastics is necesarry.

Wasser 3.0 PE-X Application in municipal wastewater treatment plants

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Apendix

Tab. 3: Measured general water parameters before (influent) and after (effluent) removal of the microplastics

Wastewater parameter	Unit	Exp. 1 influent	Exp. 1 effluent	Exp. 2 influent	Exp. 2 effluent	Exp. 3 influent	Exp. 3 effluent
Color (visual)		pale yellow	pale yellow	pale yellow	pale yellow	pale yellow	pale yellow
Turbidity (visual)		Clear	clear	clear	clear	clear	clear
Odor (qualitative)		slightly musty	slightly musty	slightly musty	slightly musty	slightly musty	slightly musty
pH value		7.39	7.32	7.47	7.32	7.31	7.41
Measuring temperature pH value	°C	17.7	20.2	18	19.6	20.3	20.3
Electrical conductivity (25 °C)	µS/cm	848	820	862	824	825	826
TOC	mg/l	7.7	8.8	8.7	8.7	8.3	8.5
DOC	mg/l	7.4	7.5	6.9	6.7	7.5	7.4
COD	mg/l	19	25	19	25	20	22
BOD5	mg/l	< 3	< 3	< 3	< 3	< 3	< 3
Ammonium-N	mg/l	0.28	0.29	0.31	0.32	0.14	0.14
Nitrite-N	mg/l	0.15	0.15	0.18	0.17	0.12	0.12
Nitrate-N	mg/l	5.4	5.3	5.5	5.2	5.9	5.9
Total inorganic nitrogen	mg/l	5.83	5.74	5.99	5.69	6.16	6.16

Tab. 4: Measured contamination with organic trace substances and heavy metals before (inlet) and after (outlet) removal of the microplastics

Wastewater parameter	Unit	Exp. 1 influent	Exp. 1 effluent	Exp. 2 influent	Exp. 2 effluent	Exp. 3 influent	Exp. 3 effluent
Wastewater Digestion							
Lead	mg/l	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Cadmium	mg/l	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001
Chrome, total	mg/l	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Coppwe	mg/l	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Nickel	mg/l	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Mercury	mg/l	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001
Zinc	mg/l	0.02	0.02	0.02	0.02	0.02	0.02
Total phosphorus	mg/l	0.5	0.48	0.54	0.55	0.49	0.49

Wastewater parameter	Unit	Exp. 1 influent	Exp. 1 effluent	Exp. 2 influent	Exp. 2 effluent	Exp. 3 influent	Exp. 3 effluent
Pharmaceutical residues							
10,11- Dihydro- 10,11- dihydroxy carbamazepine	µg/l	21	21	17	19	20	20
Azithromycin	µg/l	0.46	0.43	0.39	0.37	0.49	0.5
Bezafibrate	µg/l	0.28	0.3	0.26	0.31	0.33	0.36
Candesartan	µg/l	2.4	1.6	2.1	2.3	1.6	1.3
Carbamazepine	µg/l	0.84	0.89	0.85	0.76	1.1	1.2
Ciprofloxacin	µg/l	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05
Clarithromycin	µg/l	0.19	0.19	0.22	0.21	0.14	0.09
Dehydrato-Erythromycin A (= anhydro-erythromycin)	µg/l	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05
Diclofenac	µg/l	2.7	2.6	2.4	2.4	2.4	2.3
Erythromycin A	µg/l	< 0.05	< 0.05	< 0.05	< 0.05	0.05	< 0.05
Gabapentin	µg/l	10	9.7	9.9	9.1	10	11
Guanylurea	µg/l	57	66	62	61	53	56
Hydrochlorothiazide	µg/l	1.1	1.2	1.2	0.98	1.1	1.3
Ibuprofen	µg/l	< 1	< 1	< 1	< 1	< 1	< 1
Irbesartan	µg/l	0.54	0.42	0.59	0.57	0.48	0.5
Metformin	µg/l	0.35	0.35	0.36	0.37	0.31	0.24
Metoprolol	µg/l	1.5	1.4	1.5	1.5	1.5	1.4
Sulfamethoxazole	µg/l	0.36	0.47	0.44	0.45	0.45	0.42
Radiocontrast agents							
Amidotrizoic acid / diatrizoate	µg/l	< 0.2	< 0.2	< 0.2	< 0.2	1	0.4
Iohexol	µg/l	0.6	0.6	0.3	0.3	3.1	2.7
Iomeprol	µg/l	6.7	6.8	2.2	2.3	12	11
Iopamidol	µg/l	< 0.2	< 0.2	9.4	11	< 0.2	< 0.2
Iopromide	µg/l	< 0.2	< 0.2	< 0.2	< 0.2	4.6	1.8
Pesticides							
Carbendazim	µg/l	< 0.025	< 0.025	< 0.025	< 0.025	< 0.025	< 0.025
DEET	µg/l	1.4	1.4	1.5	1.2	1.4	1.3

Wastewater parameter	Unit	Exp. 1 influent	Exp. 1 effluent	Exp. 2 influent	Exp. 2 effluent	Exp. 3 influent	Exp. 3 effluent
Mecoprop	µg/l	0.044	0.057	0.026	0.032	0.072	0.067
Terbutryn	µg/l	0.1	0.18	0.1	0.11	0.11	0.13
Corrosion inhibitors							
Benzotriazole	µg/l	15	14	10	11	13	13
Σ 4- und 5-Methylbenzotriazole	µg/l	6.6	6.5	4.5	5.1	5.4	5
Chelating agents							
DTPA	µg/l	< 10	< 10	< 10	< 10	< 10	< 10
EDTA	µg/l	35	34	38	38	27	28
NTA	µg/l	< 10	< 10	< 10	< 10	< 10	< 10
Other chemicals							
Melamine	µg/l	3.4	2.3	3.9	3.8	2	2.2
Perfluorinated tensides							
PFBA	µg/l	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
PFBS	µg/l	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
PFOA	µg/l	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
PFOS	µg/l	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
Synthetic fragrances							
AHTN	µg/l	< 0.025	< 0.025	< 0.025	< 0.025	< 0.025	< 0.025
HHCB	µg/l	0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05
Flame retardants							
TCEP	µg/l	< 0.05	< 0.05	< 0.05	< 0.05	0.06	< 0.05
TCPP	µg/l	< 1	< 1	< 1	< 1	< 1	< 1
Artificial sweeteners							
Acesulfame	µg/l	0.5	0.5	0.5	0.5	0.3	0.3
Cyclamat	µg/l	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Sucralose	µg/l	8.1	8.9	8.3	8.7	9.1	8.9