Organic micropollutants in a large wastewater treatment plant: What are the benefits of an advanced treatment by activated carbon adsorption in comparison to conventional treatment?

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Title

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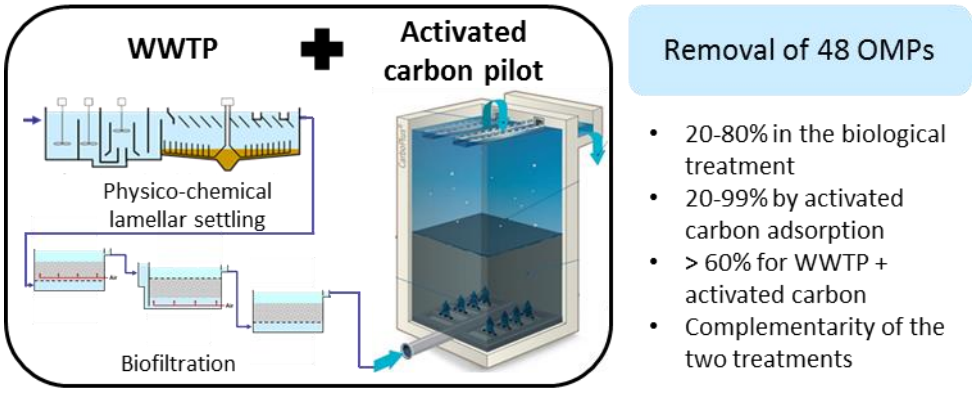
Highlights

- Full-scale study of micropollutants removal in a WWTP with an advanced treatment.
- Most micropollutants were partially removed (20-80%) in the biological treatment.
- Micropollutants were removed (20-99%) by activated carbon adsorption.
- Compounds not removed in the WWTP (< 50%) were well removed by adsorption (> 70%).
- Removals were higher than 60% over the upgraded WWTP.

Graphical abstract



Abstract

Several advanced treatments, such as ozonation or activated carbon adsorption, are currently considered for the removal of organic micropollutants (OMPs) in wastewater treatment plants (WWTP). However, little is known on the overall performances of a WWTP upgraded with those processes and the benefits provided regarding the elimination of multiple families of OMPs. In this study, 5 sampling campaigns were performed to determine the removal of 48 OMPs in a WWTP followed by an activated carbon pilot. The primary treatment had no effect on OMPs (removals < 20%), whereas the biological treatment removed OMPs that can be easily sorbed onto sludges or biodegraded (> 60%). The additional elimination provided by the advanced treatment was not significant (< 10%) for OMPs already well removed in the WWTP)

but was substantial (> 30%) for recalcitrant OMPs. Removals higher than 60% were obtained for all OMPs (except azithromycin and sulfamethoxazole) over the WWTP and the activated carbon pilot. The adsorption conditions (10 g/m^3 fresh activated carbon addition) were not sufficient to achieve the 80% removal targeted in Switzerland for compounds suggested as indicator substances for wastewater treatment. A higher dose of activated carbon or the combination with another advanced treatment should be used to achieve a satisfactory removal of those compounds.

Keywords

Organic micropollutants; Wastewater treatment plant; Activated carbon adsorption

Introduction

Many organic micropollutants (OMPs) are present in aquatic ecosystems and can lead to negative impacts (Luo et al., 2014). Treated effluents discharged by wastewater treatment plants (WWTP) are a major source for some OMPs families in aquatic ecosystems such as rivers, lakes and seas (Loos et al., 2013). WWTPs are designed to treat suspended particles and nutrients but not OMPs, which are found at concentrations ranging from ng/L to µg/L in raw wastewater. However, some removal can be achieved by conventional WWTPs for compounds exhibiting specific properties. In the primary treatment, hydrophobic compounds can be removed by sedimentation through sorption onto sludge particles and colloids (Alexander et al., 2012; Behera et al., 2011; Carballa et al., 2005). In biological processes, hydrophobic or positively charged compounds can be sorbed onto sludges, and biodegradable compounds can be degraded by metabolic or co-metabolic reactions (Falås et al., 2016; Mailler et al., 2014; Park et al., 2017; Verlicchi et al., 2012). To a lesser extent, volatile compounds can be transferred from liquid to gaseous phase by surface volatilization or stripping during aeration (Pomiès et al., 2013).

Hydrophilic and non-biodegradable OMPs, such as household chemicals, pesticides or pharmaceuticals, are partially eliminated (Margot et al., 2015; Yang et al., 2017). Complementary treatments must be performed to remove recalcitrant compounds.

To achieve OMPs removal, the most frequently used advanced technologies include activated carbon adsorption (Benstoem et al., 2017; Boehler et al., 2012; Meinel et al., 2015), advanced oxidation processes (Bourgin et al., 2017; Miklos et al., 2018; Yao et al., 2018) and membrane processes (Garcia-Ivars et al., 2017; Sahar et al., 2011; Snyder et al., 2007). Adsorption performances vary according to the type and dose of activated carbon (Kårelid et al., 2017), the operational conditions (Frank et al., 2015; Streicher et al., 2016) and the quality and composition of the water (Zietzschmann et al., 2014). Average removals range generally between 60% and 90% (Kårelid et al., 2017; Mailler et al., 2015; Margot et al., 2013) and are strongly dependent on the properties of molecules (e.g., charge, hydrophobicity or size) (Kovalova et al., 2013; Ridder et al., 2009).

The first regulation in Europe about OMPs removal (Office fédéral de l'environnement, Division Eaux, 2014) fixes a removal of 80% in comparison to the raw wastewater for at least 6 compounds among 12 compounds selected based on their behavior in WWTP (amisulpride, benzotriazole, candesartan, carbamazepine, citalopram, clarithromycin, diclofenac, hydrochlorothiazide, irbesartan, mecoprop, metoprolol and venlafaxine). Nevertheless, to achieve such performances conventional installations should often be upgraded with an advanced treatment. Many studies were conducted on OMPs removal either in WWTP or in advanced treatments but only a few investigated an upgraded WWTP (Bourgin et al., 2017; Hollender et al., 2009; Margot et al., 2013).

The aim of this study was to evaluate each treatment step in removing 48 OMPs of a large conventional WWTP upgraded with an activated carbon pilot. Five sampling campaigns were

conducted to (i) determine the efficiency of each treatment step to remove 48 OMPs, (ii) evaluate the benefits provided by activated carbon adsorption and (iii) assess if a WWTP upgraded with an advanced treatment could achieve a removal of 80% for substances suggested as indicators for regulation in Switzerland.

2. Materials and methods

2.1. Wastewater treatment plant and activated carbon advanced treatment descriptions

Seine Centre WWTP (Colombes, France), supervised by the Paris public sanitation service (SIAAP), treats wastewater of about 1,000,000 people equivalent with a flow of 240,000 m^3/day (Figure S1, SI). Screening, grit and oil removal units compose the pretreatment unit. A physico-chemical lamellar settling unit (Densadeg®) with addition of ferric chloride and anionic polymer removes particles and colloids. A first biofiltration stage (Biofor® filters – biolite media) in aerobic conditions treats the organic carbon, a second stage (Biostyr® filters – biostyrene media) ensures a total aerobic nitrification and a third stage (Biofor® filters – biolite media) in anoxic conditions allows a denitrification (Rocher et al., 2012). The WWTP switches to a degraded configuration with a flow of 405,000 m^3/day during rain events (wet weather flow from Parisian combined sewer) ensuring a partial nitrification and no denitrification. After treatment, the effluent is discharged into the Seine River.

The advanced treatment is an activated carbon adsorption pilot (CarboPlus®, Saur) which receives part of the WWTP effluent at a flow of 860 m^3/day. This process is based on a micro-grain activated carbon bed of 1000 kg which is continuously renewed by equivalent addition and extraction. A dose of 10 g of fresh activated carbon per m^3 of water is applied. The bed is fluidized in a 5 m high and 2.39 m^2 surface reactor with an ascendant fluid velocity of 15 m/h and a hydraulic retention time of about 20 min. The physical and chemical properties of the

micro-grain activated carbon (CycleCarb 305®, Chemviron Carbon) were previously determined and are listed in the supporting material (Table S1, SI).

2.2. Sampling campaigns

Five sampling campaigns were carried out between September and December 2017. WWTP influent, primary treatment effluent, WWTP effluent and activated carbon pilot effluent were sampled. Operating conditions were similar as a previous study (Mailler et al., 2016) and between each sampling campaign, except for the activated carbon pilot during the second campaign because the flow (ranging from 18 m^3/h to 24 m^3/h) and the flow velocity (10 m/h) were reduced after a technical problem on the pilot. Moreover, the WWTP operated in degraded configuration during the fourth campaign due to a rain event. For each campaign, 20 L of 24 h composite samples were collected by automatic samplers equipped with Teflon® pipes in cleaned glass bottles refrigerated at 4 °C.

2.3. Analytical procedures

Conventional wastewater quality parameters were analyzed by the certified WWTP laboratory: pH, temperature, total organic carbon (TOC), dissolved organic carbon (DOC), chemical oxygen demand (COD), biological oxygen demand in 5 days (BOD_5), total Kjeldahl nitrogen (TKN), NH_4^+, NO_2^-, NO_3^-, total phosphorus (TP), PO_4^{3-} and total suspended solids (TSS) (Table S2, SI). Results are given in the supporting material (Table S3, SI).

A total of 48 OMPs were studied. 28 micropollutants (19 pharmaceuticals, 6 pesticides, 2 hormones, and 1 perfluorinated acid) were analyzed within 48 h at the Institute of Analytical Sciences (ISA – Villeurbanne, France). Prior to analysis, all samples were homogenized and filtered on 0.7 µm GF/F glass filters (Whatman). More information on the analytical methods can be found in the literature (Barrek et al., 2009; Vulliet et al., 2011). 20 micropollutants (12

pesticides, 6 pharmaceuticals, 1 hormone and 1 degradation product) were analyzed at the CARSO laboratory (Vénissieux, France) and all samples were homogenized before analysis. The samples from the pilot effluent were filtered on a 2.7 µm GF/D glass filter (Whatman) to remove activated carbon particles and to prevent residual adsorption. Information about the analytical procedures are provided in the supporting material (Table S4, SI).

2.4. Data processing

No direct assessment of uncertainties was performed, but limit concentrations were defined for each OMP to calculate their removal. Such an approach has already been adopted by Choubert et al. (2017), which stated that the uncertainty of a micropollutant concentration in wastewater generally ranges between 30% and 100% when the measured value is lower than 2.5 to 10 times the limit of quantification (LOQ) of the compound, and lower than 30% at higher concentrations. Therefore, when a compound was quantified at concentrations above 10 times its LOQ in primary and biological influents and above the LOQ in effluents, its removal was conventionally calculated. When a compound was quantified at levels above 5 times the LOQ in WWTP effluent, and above the LOQ in the pilot effluent, the removal was also conventionally calculated. When a compound concentration was below the LOQ in effluents, its removal was estimated using LOQ/2. Removals were not calculated when concentrations of both influents and effluents were measured between the LOQ and 10 times the LOQ for the primary and biological treatment and 5 times the LOQ for the advanced treatment. The removal of each micropollutant was determined for the primary treatment, the biological treatment, the WWTP, the activated carbon advanced treatment and the combination of the WWTP and the advanced treatment. The contribution of each treatment step to the removal of each OMP was also assessed to compare their respective performances.

3. Results and discussion

Among the 48 selected OMPs, 18 compounds were not quantified at each sampling point (concentrations under their limit of quantification): 9 herbicides (aclonifen, chlorotoluron, diflufenican, 2,4-MCPA, mecoprop, metazachlor, oxadiazon, terbutryn and triallate), 3 insecticides (clothianidin, thiaclopride and thiamethoxam), 2 antibiotics (ciprofloxacin and clarithromycin), 2 hormones (17-beta-Estradiol and ethinyl estradiol), 1 molluscicide (metaldehyde) and 1 beta-blocker (metoprolol). For most of those compounds, this was the first time they were analyzed in a WWTP of the Paris conurbation. They are expected to be at trace levels (< 50 ng/L), for example herbicides that are forbidden in France, and were therefore not detected because their LOQ were equal or higher to 100 ng/L (Table S4, SI).

3.1. Elimination of OMPs in the WWTP

4 compounds (imidacloprid, propranolol, roxithromycin and tetracycline) were not quantified in WWTP influent and the primary effluent due to interferences with other compounds eluting at the same retention time. 4 other compounds (cypermethrin, irbesartan, norfloxacin and sulfadiazine) were quantified in the WWTP influent, the primary and the WWTP effluent but their removal was not calculated because of high uncertainty (values close to their respective LOQ).

3.1.1 OMPs concentration in the WWTP influent

As presented in Figure 1 and Table 1, average concentrations of OMPs in the raw influent ranged between the minimal and the maximal value found in other studies for most compounds. Variations of concentrations in the influent can be substantial, depending on the country and seasonal period. This is the first data reported for the concentrations of several molecules (acetamiprid, azithromycin, citalopram, cypermethrin, hydrochlorothiazide and irbesartan) in wastewaters of the Paris conurbation. The compounds with the highest average concentrations

were analgesics (acetaminophen, diclofenac, ibuprofen, ketoprofen and naproxen). Other commonly used pharmaceuticals (e.g., irbesartan, oxazepam, sulfamethoxazole) were found at concentrations higher than 1,000 ng/L. 7 other pharmaceuticals (atenolol, azithromycin, carbamazepine, citalopram, hydrochlorothiazide, ofloxacin and trimethoprim) had an average concentration ranging from 100 ng/L to 1,000 ng/L. 4 pharmaceuticals (estrone, lorazepam, norfloxacin and sulfadiazine) were found at concentrations below 100 ng/L, reflecting a lower consumption than other pharmaceuticals. Glyphosate was detected only during the fifth campaign at a high concentration (7,400 ng/L). Its degradation product, AMPA, was detected during each campaign with a high average concentration (6,840 ng/L) and a maximal value during the fifth campaign (10,100 ng/L). Despite the absence of agricultural land, one herbicide (diuron) and two insecticides (acetamiprid and cypermethrin) were found at trace levels (< 100 ng/L). Diuron has been banned in France since 2003 for agricultural use but is still used as a biocide, especially in paints and plaster of facades (Bester et al., 2011; Schoknecht et al., 2016). Acetamiprid and cypermethrin are used in household chemicals and agriculture (Goulson, 2013). The surfactant PFOS was also found at trace levels (52 ± 24 ng/L).

3.1.2. Primary treatment

OMPs average concentration in the WWTP influent and the primary effluent and their average removal are presented in Table 1. Despite the elimination of suspended solids (78 ± 17%, Table S2, SI), most OMPs were not affected by the primary treatment (removals between -10% and 13% for 14 compounds). Therefore, sorption onto particles is not an important removal mechanism for most pharmaceuticals and pesticides. Gasperi et al., (2010) observed a trend between the removal of OMPs and their hydrophobicity in the same treatment unit, but here no correlation or trend were observed with the log D (corrected log K_{ow} at pH = 8). This different

behavior is attributed to the difference in the type of OMPs analyzed, mostly strong hydrophobic molecules such as PAHs in the previous study.

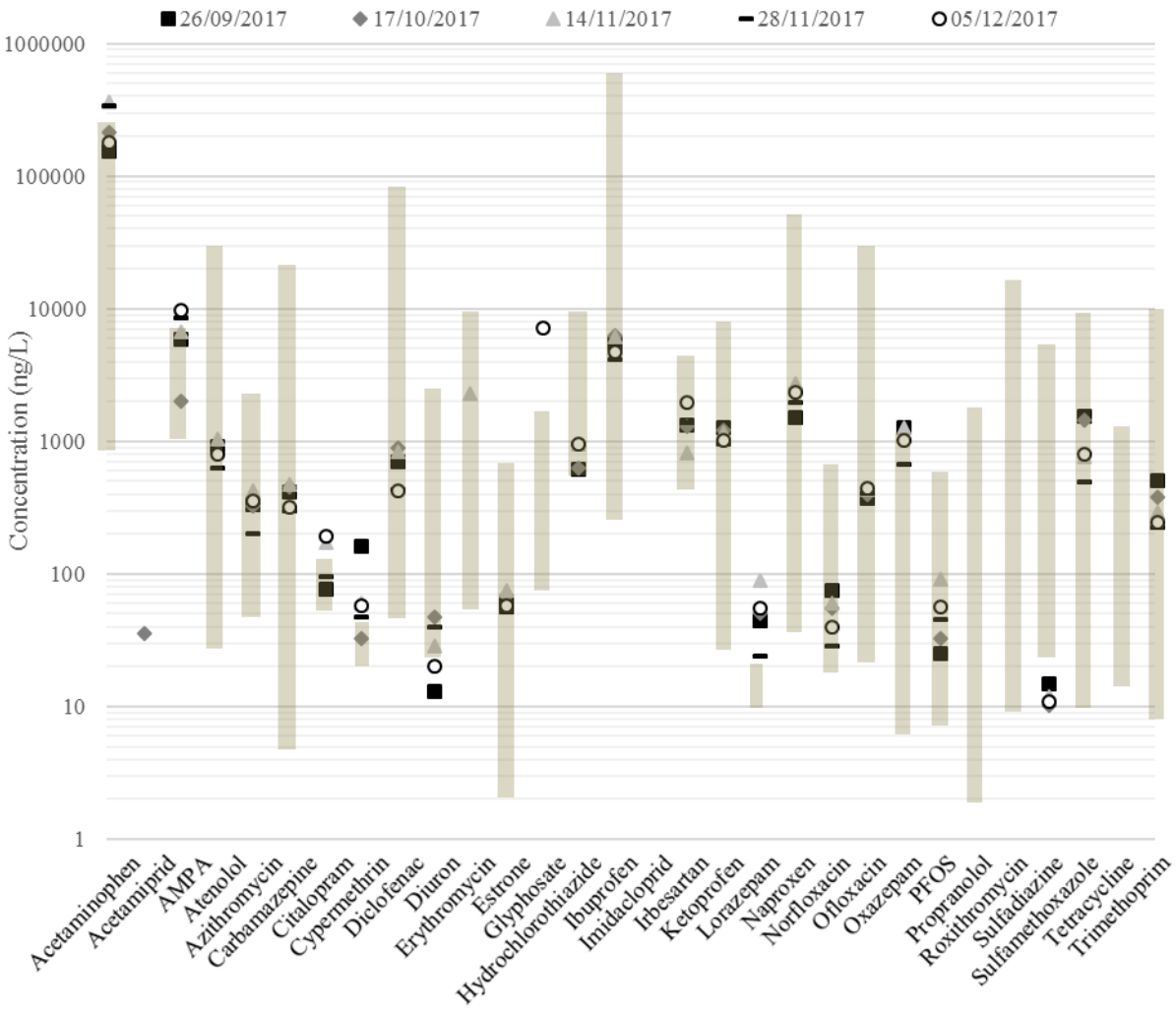


Figure 1. Concentration of 30 OMPs in the WWTP influent for each sampling campaign and range of values found in the literature (grey bars). Logarithmic-scale. References for literature values are indicated in Table 1.

3 OMPs exhibited a moderate removal in the primary treatment with high variations among the 5 campaigns: azithromycin (27 ± 31%), citalopram (31 ± 14%) and glyphosate (58%, only in the fifth campaign), but their removal remains unclear. 5 OMPs exhibited negative removals with high variations (standard deviation SD between 10% and 48%): AMPA (-17%),

erythromycin (-34%), PFOS (-32%), sulfamethoxazole (-34%) and trimethoprim (-19%). This increase of concentrations in the primary treatment has been previously observed for the same molecules (e.g., trimethoprim) and explained as a release from the particulate phase (fecal particles) when the physical and chemical properties of the wastewater varied after addition of coagulant and flocculant (Lindberg et al., 2006, 2005). Other authors suggested the transformation of metabolites or by-products to the original molecule. For example, the metabolite N^4-acetylsulfamethoxazole can be transformed back to sulfamethoxazole (Gobel et al., 2007). The increase in AMPA concentrations could be attributed to glyphosate degradation but also to the degradation of detergent components (i.e., phosphonic acids) (Kolpin et al., 2006; Skark et al., 1998).

The removal of OMPs in the primary treatment was significantly higher (ANOVA, p-value < 0.001, R) during the fourth (28/11/17, average removal = 6%) and the fifth sampling campaigns (05/12/17, average removal = 13%) as compared to the first campaign (26/09/17, average removal = - 24%) and the second campaign (17/10/17, average removal = - 19%). This indicates that the performances of the primary treatment depend on the period of the year, probably due to the seasonal variation of OMPs concentration levels in WWTP influent (Figure S3, SI). Moreover, the rain event that happened during the fourth campaign (28/11/17) didn't seem to have an impact, and removals were independent of the treated flow.

3.1.3. Biological treatment

As presented in Table 1, OMPs showed various removals in the biological treatment step, depending on their capacity to be sorbed onto sludges or to be degraded by microorganisms. No correlation or trend were observed with their hydrophobicity.

6 pharmaceuticals (acetaminophen, erythromycin, estrone, ibuprofen, ketoprofen and naproxen) and 1 pesticide (glyphosate) exhibited average removals higher than 80% with low variations (SD < 6%). Average concentrations in the WWTP effluent were below 1,000 ng/L except for acetaminophen (2,600 ng/L). Similar concentrations and removals were found in other studies for those compounds (Table 1), and the removal of some analgesics (i.e., ibuprofen, ketoprofen and naproxen) was generally higher in our study (Margot et al., 2013). Biodegradation is the main removal mechanism for these compounds (Jelic et al., 2011; Joss et al., 2005, 2004; Margot et al., 2015) but it remains unclear for glyphosate (no studies available about its biodegradation).

5 OMPs (AMPA, atenolol, lorazepam, PFOS and trimethoprim) were moderately eliminated with average removals from 50% to 80%. Low variations were observed (average SD = 7%) except for AMPA (SD = ± 33%) and trimethoprim (SD = ± 18%). Removals of these compounds were more diverse in other studies, ranging from 0% to 80% (Bourgin et al., 2017; Hollender et al., 2009; Luo et al., 2014; Margot et al., 2013; Verlicchi et al., 2012). Contrary to our results, no elimination was previously reported for AMPA and PFOS (Bourgin et al., 2017; Gardner et al., 2013; Martin Ruel et al., 2010). Atenolol and trimethoprim are primarily biodegraded (Gobel et al., 2007; Jelic et al., 2011; Margot et al., 2015), while PFOS is the only substance preferentially removed by sorption onto sludges (Margot et al., 2015).

6 pharmaceuticals (acetamiprid, carbamazepine, citalopram, diclofenac, ofloxacin and oxazepam) were persistent with average removals lower than 30% and moderate variations (SD ranging from 10% to 20%) except for oxazepam (SD = 4%). Similar removals were reported for carbamazepine, diclofenac and oxazepam (Hollender et al., 2009; Luo et al., 2014; Margot et al., 2013; Ruel et al., 2011), but lower removals for citalopram (Bourgin et al., 2017; Subedi and Kannan, 2015) and higher ones for ofloxacin (Margot et al., 2013; Verlicchi et al., 2012;

264 Yan et al., 2014). Carbamazepine and diclofenac are known as recalcitrant OMPs in the
265 biological treatment but higher removals (i.e., 13-99%) were expected for ofloxacin via sorption
266 onto sludges (Jelic et al., 2011; Joss et al., 2005).

267 2 OMPs (diuron and sulfamethoxazole) were not removed in the biological process and
268 exhibited high variations (SD = 50%). Various removals were reported for sulfamethoxazole
269 (from 4% to 100%), and Gobel et al. (2007) suggested that removal variations were due to the
270 possible transformation of the metabolite N^4-acetylsulfamethoxazole to the parent molecule.

271 Negative removals were found for 2 OMPs during each campaign: the antibiotic azithromycin
272 (-213%) and to a lesser extent the diuretic hydrochlorothiazide (- 23%). This was consistent
273 with findings of Gobel et al. (2007) who observed a poor removal of azithromycin with lots of
274 variations between studied WWTPs (from - 26% to 55%). Hydrochlorothiazide was previously
275 found in sludges but not in the dissolved phase, despite its low hydrophobicity (logD = - 0.28)
276 (Jelic et al., 2011). Similar to the primary treatment, negatives removals could be attributed to
277 desorption from sludges or metabolites recombination. However, no studies are available to
278 support this hypothesis for these two compounds.

279 The differences in removal obtained for some OMPs between this study and the literature could
280 be due to the different biological processes used (i.e., biofilters in our study as compared to
281 conventional activated sludge or membrane bio-reactor in most studies from the literature). The
282 hydraulic retention time, the biomass structure (biodiversity and activity) and the biomass
283 sorption capacity are not the same and can have an impact on OMPs removal (Mailler et al.,
284 2014).

285 The removal of OMPs didn't seem to depend on the period of the year (i.e., removals among
286 sampling campaigns were generally less variable than in the primary treatment) (ANOVA, p-

287 value = 0.927, R) (Figure S3, SI). The rain event of the fourth campaign (28/11/17) didn't have
288 an impact either. Conventional water quality parameters did not show dramatic evolutions
289 during this campaign (Table S2, SI): NH_4^+, TKN and BOD_5 removals were 85%, 85%, and
290 87%, respectively, compared to an average of 98%, 96% and 96% for the four other campaigns.
291 This implies that the WWTP still achieved satisfactory performances (i.e., > 80%) despite an
292 increased flow and a degraded configuration and explains why the removal of OMPs did not
293 fluctuate significantly.

294 3.1.4. Overall WWTP

295 5 OMPs suggested as indicator substances in Switzerland (carbamazepine, citalopram,
296 diclofenac, hydrochlorothiazide, irbesartan, (Office fédéral de l'environnement, Division Eaux,
297 2014)) were quantified in the WWTP influent and effluent and their removal are presented in
298 Table 1 (except irbesartan due to values close to the LOQ). The Swiss regulation requires the
299 removal of at least 80% of 6 indicator substances among a list of 12 molecules. In our case,
300 removals below 50% were obtained for carbamazepine, citalopram and diclofenac. The average
301 concentration of hydrochlorothiazide and irbesartan was even higher in the WWTP effluent
302 than in the influent. The degree of nitrification was shown to be positively correlated with the
303 removal of some OMPs (Margot et al., 2013). Since the degree of nitrification was already high
304 in our study (average NH_4^+ removal = 95%), even higher OMPs removal seems hardly feasible.
305 Stable removals were observed for conventional wastewater quality parameters (e.g. nitrogen
306 parameters related to the degree of nitrification) in both the primary treatment (Gasperi et al.,
307 2010) and the biological treatment (Rocher et al., 2012) over a year. The same behavior can be
308 expected for OMPs since biodegradation (which is the main mechanism) is correlated with the
309 degree of nitrification. Therefore, the conventional WWTP processes were not adapted to treat
310 OMPs and would not comply with a similar regulation to the Swiss water act protection. An

311 upgrade with an advanced treatment is thus necessary to achieve removals higher than 80% and
312 to guarantee the protection of the receiving aquatic ecosystem.

Table 1. Concentration of 30 OMPs quantified in the WWTP influent and in the primary and WWTP effluents; removal rate achieved by primary treatment, biological treatment and the combination of both processes (WWTP removal); and values reported in the literature for similar processes. The OMPs used as indicator substances in Switzerland are in bold. [a] SD = standard deviation between 5 campaigns.

	This study						Literature			
	Concentrations (ng/L) (± SD [a])			Removal (%) (± SD [a])			Concentrations (ng/L)		Removal (%)	References
Molecule	WWTP influent	Primary effluent	WWTP effluent	Primary treatment	Biological treatment	WWTP	WWTP influent	WWTP effluent	WWTP	
Acetaminophen	260495 (± 89648)	25570 (± 59176)	2618 (± 4848)	-3 (± 16)	99 (± 2)	99 (± 1)	960-246000	2-20000	80-100	1, 2, 3, 4, 5, 6, 7, 8, 9, 10
Acetamiprid	37	32	22 (± 3)	13	18	29	-	-	-	
AMPA	6840 (± 2754)	7860 (± 3 037)	460 (± 543)	-17 (± 28)	62 (± 33)	62 (± 33)	1164-7143	735-7000	0	11, 12, 13, 14
Atenolol	873 (± 142)	942 (± 173)	288 (± 61)	-8 (± 10)	69 (± 7)	67 (± 6)	30-33100	5-7600	0-85	1, 3, 4, 7, 8, 9, 10, 15, 16
Azithromycin	342 (± 75)	258 (± 135)	698 (± 215)	27 (± 31)	-213 (± 126)	-100 (± 40)	56-2272	40-380	5-74	3, 4, 6, 9, 10, 16
Carbamazepine	410 (± 72)	391 (± 66)	337 (± 76)	4 (± 7)	14 (± 12)	19 (± 8)	5-21500	5-6300	0-97	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 16, 17, 18, 19
Citalopram	140 (± 51)	103 (± 34)	86 (± 21)	31 (± 14)	21 (± 12)	47 (± 3)	59-133	34-280	18	10, 15, 20, 21
Cypermethrin	75 (± 48)	37 (± 23)	20	-	-	-	21-44	-	-	22
Diclofenac	687 (± 209)	750 (± 260)	510 (± 128)	-8 (± 12)	29 (± 11)	24 (± 7)	50-94200	1-10670	0-81	1, 3, 5, 6, 7, 8, 9, 10, 13, 19
Diuron	31 (± 13)	36 (± 10)	42 (± 29)	-9 (± 21)	-2 (± 54)	-19 (± 85)	25-2530	2-2530	18-72	1, 4, 5, 10, 12
Erythromycin	2374	3191	245 (± 87)	-34	90	87	60-10020	9-2840	0-97	2, 3, 5, 6, 8, 16, 23
Estrone	65 (± 9)	66 (± 10)	5 (± 1)	-4 (± 13)	96 (± 2)	95 (± 3)	2-670	1-110	12-96	1, 3, 4, 5, 16, 18
Glyphosate	7400	3100	500	58	84	93	80-1 800	84-1380	0-40	11, 12, 14
Hydrochlorothiazide	856 (± 172)	886 (± 123)	1070 (± 124)	-6 (± 15)	-23 (± 20)	-31 (± 32)	610-10000	670-11000	-	3, 10
Ibuprofen	5 597 (± 851)	6127 (± 1439)	320 (± 213)	-8 (± 13)	95 (± 6)	95 (± 6)	268-603000	1-48240	52-100	1, 2, 3, 4, 6, 8, 10, 18, 19
Imidacloprid	-	-	25 (± 3)	-	-	-	-	3-166	-	24
Irbesartan	1412 (± 378)	1584 (± 529)	1792 (± 734)	-	-	-	490-4700	475-1700	79	4, 10, 20
Ketoprofen	1169 (± 121)	1261 (± 264)	240 (± 47)	-7 (± 14)	81 (± 3)	79 (± 4)	31-8560	3-3920	7-100	1, 3, 4, 5, 7, 8, 9, 10, 19,
Lorazepam	55 (± 22)	57 (± 22)	28 (± 9)	-10 (± 46)	54 (± 7)	48 (± 17)	10-20	11-196	-	3, 10, 15
Naproxen	2302 (± 450)	2259 (± 409)	205 (± 111)	0 (± 15)	91 (± 5)	91 (± 6)	40-52900	1-5200	35-100	1, 3, 4, 5, 7, 8, 9, 10, 18, 19
Norfloxacin	54 (± 17)	39 (± 82)	31 (± 10)	-	-	-	18-680	7-210	18-96	3, 4, 6, 9, 10, 16, 17, 23
Ofloxacin	429 (± 28)	402 (± 82)	333 (± 101)	5 (± 22)	18 (± 20)	21 (± 28)	22-31700	19-953	13-99	3, 4, 6, 16, 23
Oxazepam	1089 (± 226)	1135 (± 299)	844 (± 215)	-4 (± 12)	25 (± 4)	23 (± 10)	7-1200	8-499	13-25	4, 8, 10, 15
PFOS	52 (± 24)	67 (± 26)	16 (± 4)	-32 (± 10)	75 (± 6)	67 (± 9)	8-638	7-1057	-3	10, 20, 24
Propranolol	-	-	181 (± 45)	-	-	-	2-1900	2-560	0-78	1, 3, 4, 9, 10, 15, 16
Roxithromycin	-	-	157 (± 93)	-	-	-	10-17000	10-5000	0-75	1, 3, 6, 9, 16, 23
Sulfadiazine	13 (± 2)	13 (± 3)	11 (± 2)	-	-	-	25-5100	2-164	78-100	3, 6, 16
Sulfamethoxazole	1057 (± 432)	1581 (± 1079)	1312 (± 629)	-34 (± 48)	-4 (± 44)	-29 (± 53)	10-10000	3-5000	4-100	1, 2, 3, 4, 5, 6, 7, 10, 16, 18
Tetracycline	-	-	94 (± 53)	-	-	-	15-1344	2-5000	24-80	2, 3, 9, 16
Trimethoprim	344 (± 107)	429 (± 196)	118 (± 42)	-19 (± 25)	65 (± 18)	61 (± 17)	9-10500	10-6700	0-99	2, 3, 4, 5, 6, 7, 9, 10, 18, 23

1: (Ruel et al., 2011); 2: (Yang et al., 2011); 3: (Verlicchi et al., 2012); 4: (Margot et al., 2013); 5: (Luo et al., 2014); 6: (Yan et al., 2014); 7: (Papageorgiou et al., 2016); 8: (Thiebault et al., 2017a, 2017b); 9: (Park et al., 2017); 10: (Bourgin et al., 2017); 11: (Popp et al., 2008); 12: (Martin Ruel et al., 2010); 13: (Gardner et al., 2013); 14: (Mailler et al., 2014); 15: (Subedi and Kannan, 2015); 16: (Ben et al., 2018); 17: (Kostich et al., 2014); 18:(Baalbaki et al., 2016); 19: (Martínez-Alcalá et al., 2017); 20: (Loos et al., 2013); 21: (Golovko et al., 2014); 22: (Weston Donald P. et al., 2013); 23: (Leung et al., 2012); 24: (Yu et al., 2009).

318 *3.2. Benefits provided by the advanced treatment*

319 3.2.1. Elimination of OMPs in the activated carbon adsorption pilot

320 Average concentrations of OMPs in the WWTP effluent and in the pilot effluent are presented
321 in Table 2 along with their average removal. The removal of three substances (cypermethrin,
322 estrone, glyphosate) was not calculated due to concentrations too close to their LOQ.

323 Concentrations in the WWTP effluent were similar to values reported in the literature (i.e.,
324 average concentration below 500 ng/L for 85% of OMPs) except for acetaminophen (2600 ng/L
325 compared to a maximum of 380 ng/L in the literature), irbesartan (1792 ng/L vs. 475 ng/L) and
326 oxazepam (844 ng/L vs. 239 ng/L) (Table 2).

327 Most OMPs were moderately or well removed in the advanced treatment, but no correlation or
328 trend were observed with the log D indicating that the hydrophobicity alone is not sufficient to
329 estimate OMPs removal. No trend was observed between OMPs removal and their physico-
330 chemical properties such as the size, the charge, the polarizability or some functional groups.
331 The adsorption process cannot be described by a single parameter, but rather by a variety of
332 physical and chemical interactions between OMPs, activated carbon and dissolved organic
333 matter (Mailler et al., 2016; Margot et al., 2013; Zietzschmann et al., 2014). Average removals
334 higher than 80% were obtained for 8 OMPs (acetaminophen, acetamiprid, diuron, ibuprofen,
335 imidacloprid, norfloxacin, propranolol and trimethoprim) with low variations among sampling
336 campaigns (SD < 6%). To the best of authors' knowledge, this is the first data reported for the
337 removal of the two insecticides acetamiprid and imidacloprid by activated carbon adsorption.
338 A substantial elimination was observed for 11 pharmaceuticals (atenolol, azithromycin,
339 carbamazepine, citalopram, diclofenac, hydrochlorothiazide, ketoprofen, naproxen, ofloxacin,
340 oxazepam, tetracycline) with average removals ranging from 50% to 80% and moderate

variations (SD ranging from 8% to 15%). Similar results were obtained for those 19 compounds (i.e., which were moderately or well removed) in different pilot configurations (powdered activated carbon in a contact reactor or a fluidized bed, or granular filters) (Beijer et al., 2017; Bourgin et al., 2017; Mailler et al., 2016; Margot et al., 2013). Those OMPs have a good affinity with all types of activated carbon and their removal is not affected by the process configuration.

A low elimination was observed for 7 OMPs (erythromycin, irbesartan, lorazepam, PFOS, roxithromycin, sulfadiazine and sulfamethoxazole) with average removals ranging from 30% to 50% and more variability between campaigns (SD from 11% to 19%). Similar results were reported in the literature for erythromycin and sulfamethoxazole (TableTable 2). Erythromycin and roxithromycin are large molecules (minimal projection radius of 0.67 nm and 0.91 nm, data from MarvinSketch, ChemAxon) that could not access to micropores (average size of 0.6 nm, Table S1, SI) but only to mesopores where a higher competition with the dissolved organic matter occurred (Zietzschmann et al., 2014). Higher removals of lorazepam, roxithromycin and sulfadiazine were found in a previous study with the same pilot, micro-grain activated carbon, same dose (10 g/m^3) and similar concentrations in the WWTP effluent (Mailler et al., 2016), and the explanation to those differences is currently investigated. Similar removals of irbesartan were obtained with a textile filter combined with a granular activated carbon filter (Bourgin et al., 2017) but higher performances were achieved with a powdered activated carbon reactor combined with ultrafiltration membranes (Margot et al., 2013) and granular activated carbon filters (Beijer et al., 2017). The by-product AMPA was the substance with the lowest removal in the pilot (13%), while a high removal (> 70%) was obtained by Ruel et al. (2011) with a granular activated carbon filter. The removal of those 7 OMPs appeared more variable and dependent on the type of activated carbon or process used.

OMPs removal were significantly higher (ANOVA, p-value < 0.01, R) during the first (26/09/17: total average removal = 75%) and second campaigns (17/10/17: 75%) than during the 3 others (63%, 56% and 56%) (Figure S4, SI). A first hypothesis would be that the performances of the pilot depend on the period of the year, mainly under the influence of the water temperature. However, the activated carbon bed mass was higher during the first campaign (1200 kg) and a reduced flow occurred during the second campaign, increasing contact time and the dose of activated carbon, which explains the higher removals. Moreover, the rain even that happened during the fourth campaign (28/11) had an effect on the removal of most OMPs. Lower removals were achieved in comparison to the other sampling campaigns, even if hydraulic conditions and activated carbon doses were identical. Similar results about the effect of rain events (i.e., degraded WWTP configuration) were found in a previous study (Mailler et al., 2015), explained by the hypothesis of higher levels of organic matter and higher concentrations of OMPs. However, the quality of the WWTP effluent in our study (i.e., dissolved organic carbon, total nitrogen and OMPs) didn't vary statistically from one campaign to another (ANOVA, p-value = 0.804, R) (Table S3, SI). In the previous study (Mailler et al., 2016), the removal of 13 pharmaceuticals was less impacted by the period (± 5 to 25%) than by the treatment rate (20 g/m^3 vs 10 g/m^3). Moreover, the UV_{254} removal, used as surrogate for the removals of the 13 pharmaceuticals, was not affected by the period of the year (standard deviation < ± 2%). Therefore, the studied period can be considered representative of the performance of the process over the year.

3.2.2. Relative contribution of advanced treatment compared to the conventional process

The average concentration of OMPs in the WWTP influent and the pilot effluent are presented in Table 3, as well as their overall removal. No data are presented for cypermethrin due to low concentrations in all samples. The benefits provided by the activated carbon adsorption for the

388 elimination of OMPs were minor for 7 compounds that were mostly eliminated (>80%) in the
389 WWTP (acetaminophen, erythromycin, estrone, glyphosate, ibuprofen, ketoprofen and
390 naproxen) (Figure 2). As an example, acetaminophen was removed at 99% overall and the
391 advanced treatment only contributed to 1% of its elimination. An advanced treatment was thus
392 not necessary in terms of removal for those OMPs but was however effective at decreasing their
393 concentration (e.g., 320 ng/L to 3 ng/L for ibuprofen) (Table 2). 4 OMPs (atenolol, lorazepam,
394 PFOS, trimethoprim) that were moderately removed in the WWTP (from 50% to 80%) were
395 further removed by the advanced treatment (additional removal ranging between 13% and
396 29%).

Table 2. Concentration of 30 OMPs quantified in the effluents of the WWTP and of the pilot, removal rate achieved within the advanced treatment, and values reported in the literature for similar processes. The OMPs used as indicator substances in Switzerland are in bold. [a] SD = standard deviation between 5 campaigns.

	This study			Literature			
	Concentrations (ng/L) (± SD [a])		Removal (%) (± SD [a])	Concentrations (ng/L)		Removal (%)	References
Molecule	WWTP effluent	Pilot effluent	Pilot	WWTP effluent	Tertiary effluent	Tertiary	
Acetaminophen	2618 (± 4848)	363 (± 683)	85 (± 7)	33-380	32	99	(Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011; Yang et al., 2011)
Acetamiprid	22 (± 3)	1	98 (± 0)	-	-	-	
AMPA	460 (± 543)	420 (± 466)	13	360-5200	720	>70	(Ruel et al., 2011)
Atenolol	288 (± 61)	73 (± 21)	73 (± 10)	102-448	<LQ-90	70-99	(Beijer et al., 2017; Bourgin et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011)
Azithromycin	698 (± 215)	262 (± 142)	62 (± 15)	140-935	51	63-76	(Bourgin et al., 2017; Margot et al., 2013)
Carbamazepine	337 (± 76)	119 (± 27)	63 (± 11)	66-640	< LQ-67	23-99	(Beijer et al., 2017; Bourgin et al., 2017; Grover et al., 2011; Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011; Yang et al., 2011)
Citalopram	86 (± 21)	19 (± 10)	69 (± 1)	30-212	<LQ-16	85-99	(Beijer et al., 2017; Bourgin et al., 2017)
Cypermethrin	20	20	-	-	-	-	
Diclofenac	510 (± 128)	235 (± 35)	51 (± 13)	13-1350	<LQ-480	63-98	(Beijer et al., 2017; Bourgin et al., 2017; Grover et al., 2011; Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011; Yang et al., 2011)
Diuron	42 (± 29)	4 (± 5)	92 (± 6)	17-524	<LQ	70-91	(Bourgin et al., 2017; Margot et al., 2013; Ruel et al., 2011)
Erythromycin	245 (± 87)	151 (± 49)	35 (± 11)	124-270	28-156	24-90	(Mailler et al., 2016, 2015; Yang et al., 2011)
Estrone	5 (± 1)	5 (± 1)	-	6-20	<LQ-9	45-92	(Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011)
Glyphosate	500	500	-	84-1100	<LQ	>70	(Ruel et al., 2011)
Hydrochlorothiazide	1070 (± 124)	378 (± 94)	64 (± 9)	930	185	80	(Bourgin et al., 2017)
Ibuprofen	320 (± 213)	3	99 (± 1)	9-2500	9	38-95	(Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011; Yang et al., 2011)
Imidacloprid	25 (± 3)	3 (± 2)	89 (± 7)	-	-	-	
Irbesartan	1792 (± 734)	1102 (± 395)	36	54-475	<LQ-230	51-98	(Beijer et al., 2017; Bourgin et al., 2017; Margot et al., 2013)
Ketoprofen	240 (± 47)	83 (± 26)	66 (± 8)	34-640	<LQ-16	50-99	(Beijer et al., 2017; Bourgin et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011)
Lorazepam	28 (± 9)	13 (± 4)	49 (± 19)	11-17	<LQ-6	63-97	(Bourgin et al., 2017; Mailler et al., 2016, 2015)
Naproxen	205 (± 111)	80 (± 59)	62 (± 10)	11-449	<LQ-47	67-81	(Beijer et al., 2017; Bourgin et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011)
Norfloxacin	31 (± 10)	3	92 (± 2)	67-98	10-17	60-98	(Bourgin et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013)
Ofloxacin	333 (± 101)	89 (± 20)	71 (± 8)	3-953	<LQ-363	63-94	(Beijer et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013)
Oxazepam	844 (± 215)	362 (± 91)	54 (± 15)	20-257	<LQ-159	39-99	(Beijer et al., 2017; Bourgin et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013)
PFOS	16 (± 4)	9 (± 4)	45 (± 11)	8-55	5	37-88	(Bourgin et al., 2017; Mailler et al., 2016)
Propranolol	181 (± 45)	14 (± 6)	91 (± 5)	37-160	4-40	17-99	(Grover et al., 2011; Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011)
Roxithromycin	157 (± 93)	93 (± 46)	34 (± 18)	10-464	35	60-99	(Mailler et al., 2016, 2015; Ruel et al., 2011)
Sulfadiazine	11 (± 2)	6 (± 2)	47 (± 6)	10-83	4	60-95	(Mailler et al., 2016, 2015)
Sulfamethoxazole	1312 (± 629)	758 (± 227)	34 (± 19)	27-1430	<LQ-670	0-64	(Beijer et al., 2017; Bourgin et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013; Yang et al., 2011)
Tetracycline	94 (± 53)	30 (± 25)	59	47	33	61	(Mailler et al., 2016)
Trimethoprim	118 (± 42)	19 (± 9)	85 (± 6)	61-181	<LQ-9	87-94	(Beijer et al., 2017; Bourgin et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013)

Their overall removal reached values higher than 80%. The activated carbon adsorption had a more important impact on 7 substances (acetamiprid, AMPA, carbamazepine, citalopram, diclofenac, ofloxacin, oxazepam) that were partially eliminated in the WWTP (from 13% to 40%): their additional removal ranged from 38% to 59% in the advanced treatment, with a high overall removal (from 62% to 99%). 4 OMPs were not quantified in the WWTP influent (imidacloprid, propranolol, roxithromycin and tetracycline) and the overall removal of 3 other substances (irbesartan, norfloxacin and sulfadiazine) was not calculated due to low concentrations in the primary treatment effluent. Therefore, only the pilot contributed to the elimination (34% to 92%) of those 7 OMPs. Finally, 4 OMPs were negatively removed in the WWTP (azithromycin, diuron, hydrochlorothiazide and sulfamethoxazole) but the activated carbon adsorption was sufficient to obtain positive overall removals (18% to 87%).

High eliminations were also achieved in studies using conventional activated sludge combined with ozonation or granular activated carbon filters for 8 OMPs present in our study (acetaminophen, atenolol, citalopram, erythromycin, ibuprofen, ketoprofen, naproxen and trimethoprim) (Bourgin et al., 2017; Hollender et al., 2009). Therefore, those compounds can be qualified as easily eliminated in conventional WWTP and advanced treatment. The same statement could be made for 6 other OMPs (hydrochlorothiazide, norfloxacin, propranolol, roxithromycin, sulfadiazine and tetracycline) based on the literature, but their elimination by biological treatment could not be estimated in this study. 6 molecules (carbamazepine, diclofenac, oxazepam, irbesartan, diuron, sulfamethoxazole) are generally well eliminated in upgraded WWTPs but show important variations depending mostly on the advanced treatment used. For example, diclofenac is well removed in WWTPs upgraded with activated carbon (around 65%) and very well removed by ozonation (> 90%) (Bourgin et al., 2017; Hollender et al., 2009; Yang et al., 2011). No data was available in the literature for others OMPs (acetamiprid, estrone, glyphosate, imidacloprid, lorazepam, ofloxacin) to suggest a general

behavior in WWTPs combined with advanced treatment, but some assumptions can be made based on removals in WWTPs and advanced treatments separately. Glyphosate, lorazepam and ofloxacin were poorly eliminated in WWTPs (Bourgin et al., 2017; Margot et al., 2013; Martin Ruel et al., 2010; Verlicchi et al., 2012) but were well removed by activated carbon adsorption (granular, micro-grain and powdered) (Bourgin et al., 2017; Mailler et al., 2016, 2015; Margot et al., 2013; Ruel et al., 2011), therefore good removals can be expect for these OMPs in a WWTP upgraded with an activated carbon treatment.

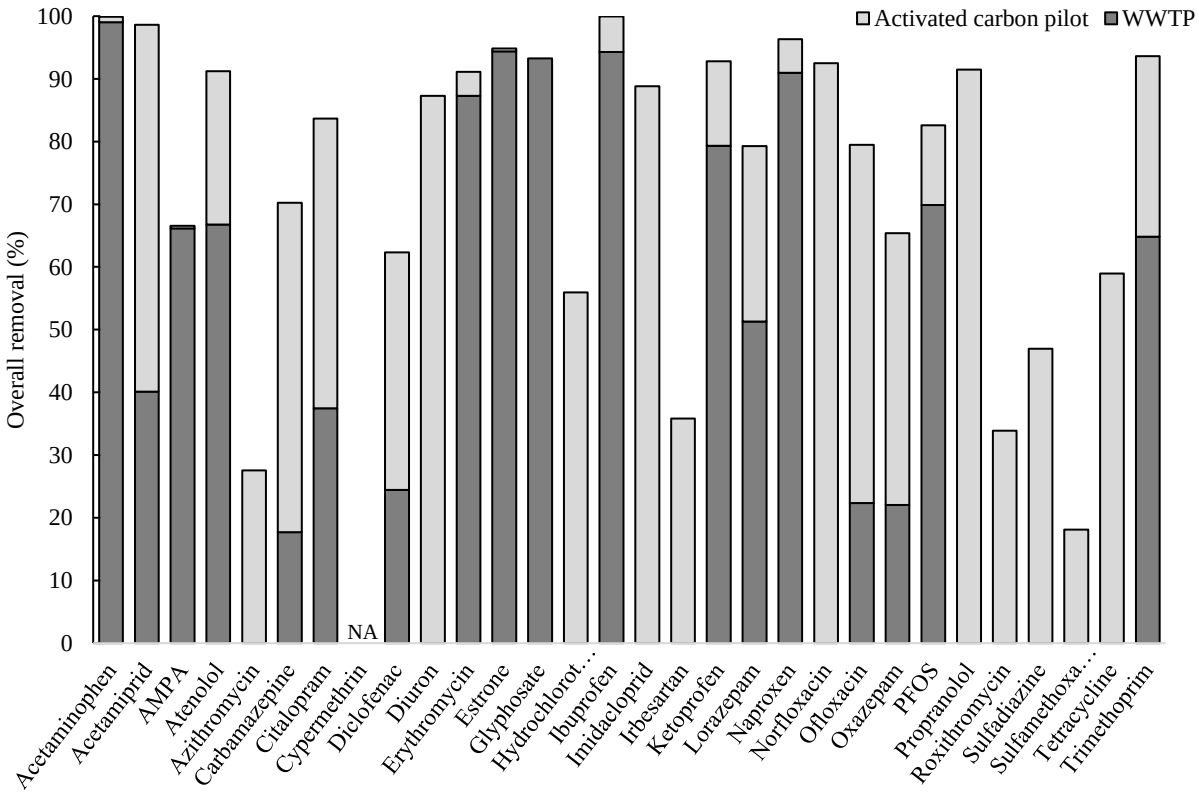


Figure 2. Overall removal of 30 OMPs over the WWTP and the activated carbon advanced treatment. The contribution of the WWTP (dark grey) and the activated carbon advanced treatment (light grey) to the overall removal are displayed for each OMPs. NA: no data are presented due to low concentrations in all samples (< 5 times LOQ).

438 Table 3. Concentration of 30 OMPs found in the WWTP influent and the pilot effluent; removal rate achieved within the WWTP and the advanced treatment combined; and values reported in the
439 literature for similar processes. The OMPs used as indicator substances in Switzerland are in bold. [a] SD = standard deviation between 5 campaigns.

	This study			Literature			
	Concentrations (ng/L) (± SD [a])		Removal (%) (± SD [a])	Concentrations (ng/L)		Removal (%)	References
Molecule	WWTP influent	Pilot effluent	WWTP + pilot	WWTP influent	Tertiary effluent	WWTP + tertiary	
Acetaminophen	260495 (± 89648)	363 (± 683)	100 (± 0)	33764-80000	<LQ	100	(Bourgin et al., 2017; Hollender et al., 2009; Yang et al., 2011)
Acetamiprid	37	1	99	-	-	-	
AMPA	6840 (± 2754)	420 (± 466)	67 (± 28)	-	-	-	
Atenolol	873 (± 142)	73 (± 21)	91 (± 4)	860-2519	11-276	89-99	(Bourgin et al., 2017; Hollender et al., 2009)
Azithromycin	342 (± 75)	262 (± 142)	28 (± 27)	-	51	-	
Carbamazepine	410 (± 72)	119 (± 27)	70 (± 8)	150-509	1-67	63-99	(Bourgin et al., 2017; Hollender et al., 2009; Yang et al., 2011)
Citalopram	140 (± 51)	19 (± 10)	84 (± 0)	113	1-16	86-99	(Bourgin et al., 2017)
Cypermethrin	75 (± 48)	20 (± 0)	-	-	-	-	
Diclofenac	687 (± 209)	235 (± 35)	62 (± 13)	220-1466	7-480	67-99	(Bourgin et al., 2017; Hollender et al., 2009; Yang et al., 2011)
Diuron	31 (± 13)	4 (± 5)	87 (± 13)	111	2-54	67-93	(Bourgin et al., 2017; Hollender et al., 2009)
Erythromycin	2374	151 (± 49)	91	340	28	92	(Yang et al., 2011)
Estrone	65 (± 9)	5 (± 1)	93 (± 2)	-	-	-	
Glyphosate	7400	500 (± 0)	93	-	-	-	
Hydrochlorothiazide	856 (± 172)	378 (± 94)	56 (± 5)	980	13-185	81-99	(Bourgin et al., 2017)
Ibuprofen	5597 (± 851)	3 (± 0)	100	3860-11000	<LQ	100	(Bourgin et al., 2017; Hollender et al., 2009; Yang et al., 2011)
Imidacloprid	-	3 (± 2)	-	-	-	-	
Irbesartan	1412 (± 378)	1102 (± 395)	-	490	50-230	53-90	(Bourgin et al., 2017)
Ketoprofen	1169 (± 121)	83 (± 26)	93 (± 3)	275	16-50	82-94	(Bourgin et al., 2017)
Lorazepam	55 (± 22)	13 (± 4)	79 (± 1)	-	6	-	
Naproxen	2302 (± 450)	80 (± 59)	96 (± 3)	587-1035	47-79	95-98	(Bourgin et al., 2017; Hollender et al., 2009)
Norfloxacin	54 (± 17)	3 (± 0)	-	185	10	95	(Bourgin et al., 2017)
Ofloxacin	429 (± 28)	89 (± 20)	79 (± 4)	-	-	-	
Oxazepam	1089 (± 226)	362 (± 91)	65 (± 10)	102	11-45	56-90	(Bourgin et al., 2017)
PFOS	52 (± 24)	9 (± 4)	83 (± 4)	8	4-8	1-47	(Bourgin et al., 2017)
Propranolol	-	14 (± 6)	-	31-107	2-26	87-94	(Bourgin et al., 2017; Hollender et al., 2009)
Roxithromycin	-	93 (± 46)	-	21	-	91	(Hollender et al., 2009)
Sulfadiazine	13 (± 2)	6 (± 2)	-	130	-	80	(Hollender et al., 2009)
Sulfamethoxazole	1057 (± 432)	758 (± 227)	18 (± 34)	200-292	5-102	49-96	(Bourgin et al., 2017; Hollender et al., 2009)
Tetracycline	-	30 (± 25)	-	160	<LQ	97	(Yang et al., 2011)
Trimethoprim	344 (± 107)	19 (± 9)	94 (± 4)	91-610	9-31	90-98	(Bourgin et al., 2017; Hollender et al., 2009; Yang et al., 2011)

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3.2.3. Relevance of the advanced treatment

Among the 5 molecules classified in Switzerland and found in this study, the targeted 80% removal was only obtained for citalopram (84%). In other studies, satisfactory removals were generally obtained for carbamazepine and citalopram (Table 3). A higher dose of activated carbon than that used in our study (i.e., 10 g/m^3) should be employed in order to reach an average removal higher than 80% and comply with the Swiss water protection act. Removals higher than 80% were obtained for carbamazepine with an activated carbon dose of 20 g/m^3 in a previous study using the same process (Mailler et al., 2016). Another solution would be to use ozonation or membrane filtration prior to the activated carbon adsorption process (Bourgin et al., 2017; Margot et al., 2013; Yang et al., 2011).

The advanced treatment improved the quality of the effluent and most OMPs were moderately or substantially removed (final concentrations < 200 ng/L for 70% of OMPs). A reduction in toxic impacts can be expected for the receiving aquatic ecosystem, especially because of the low residuals of compounds usually found at concentrations higher than 1000 ng/L in the treated effluent (e.g., acetaminophen, hydrochlorothiazide, irbesartan and sulfamethoxazole).

Ecotoxicological and economical studies as well as life-cycle assessment should be conducted to get an overall view of the benefits provided by the advanced treatment but also of its costs.

Conclusions

- Most OMPs were not affected by the primary treatment, but 3 compounds were moderately removed. However, 5 compounds exhibited negative removals, possibly due to a release from fecal particles or transformation of metabolites and by-products.

- Various removals were observed in the biological treatment, depending on OMPs capacity to be sorbed onto sludges or to be biodegraded. 11 compounds were removed by more than 50%, while 10 compounds were not significantly eliminated.
- Activated carbon adsorption was used as an advanced treatment and substantially improved the quality of the WWTP effluent. Most compounds were removed by more than 50% and average concentrations in the pilot effluent were below 200 ng/L for 70% of OMPs.
- The combination of the WWTP and the advanced treatment exhibited removals higher than 60% for all compounds except sulfamethoxazole. One molecule classified in Switzerland regulation (citalopram) was removed by more than 80% but not the 4 others included in our study (carbamazepine, citalopram, hydrochlorothiazide, irbesartan). A higher dose of activated carbon or a combination with another advanced treatment should be applied to achieve the targeted 80% removal.
- The impact of the advanced treatment was low for OMPs well removed in the WWTP. However, it improved the removal of the OMPs moderately eliminated in the WWTP to an overall removal higher than 80% and it removed (> 50%) the OMPs that were not eliminated in the WWTP.

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References

Alexander, J.T., Hai, F.I., Al-aboud, T.M., 2012. Chemical coagulation-based processes for trace organic contaminant removal: Current state and future potential. Journal of

Environmental Management 111, 195–207. https://doi.org/10.1016/j.jenvman.2012.07.023

Baalbaki, Z., Sultana, T., Maere, T., Vanrolleghem, P.A., Metcalfe, C.D., Yargeau, V., 2016. Fate and mass balance of contaminants of emerging concern during wastewater treatment determined using the fractionated approach. Science of The Total Environment 573, 1147–1158. https://doi.org/10.1016/j.scitotenv.2016.08.073

Barrek, S., Cren-Olivé, C., Wiest, L., Baudot, R., Arnaudguilhem, C., Grenier-Loustalot, M.-F., 2009. Multi-residue analysis and ultra-trace quantification of 36 priority substances from the European Water Framework Directive by GC–MS and LC-FLD-MS/MS in surface waters. Talanta 79, 712–722. https://doi.org/10.1016/j.talanta.2009.04.058

Behera, S.K., Kim, H.W., Oh, J.-E., Park, H.-S., 2011. Occurrence and removal of antibiotics, hormones and several other pharmaceuticals in wastewater treatment plants of the largest industrial city of Korea. Science of The Total Environment 409, 4351–4360. https://doi.org/10.1016/j.scitotenv.2011.07.015

Beijer, K., Björlenius, B., Shaik, S., Lindberg, R.H., Brunström, B., Brandt, I., 2017. Removal of pharmaceuticals and unspecified contaminants in sewage treatment effluents by activated carbon filtration and ozonation: Evaluation using biomarker responses and chemical analysis. Chemosphere 176, 342–351. https://doi.org/10.1016/j.chemosphere.2017.02.127

Ben, W., Zhu, B., Yuan, X., Zhang, Y., Yang, M., Qiang, Z., 2018. Occurrence, removal and risk of organic micropollutants in wastewater treatment plants across China: Comparison of wastewater treatment processes. Water Research 130, 38–46. https://doi.org/10.1016/j.watres.2017.11.057

Benstoem, F., Nahrstedt, A., Boehler, M., Knopp, G., Montag, D., Siegrist, H., Pinnekamp, J., 2017. Performance of granular activated carbon to remove micropollutants from municipal wastewater—A meta-analysis of pilot- and large-scale studies. Chemosphere 185, 105–118. https://doi.org/10.1016/j.chemosphere.2017.06.118

Bester, K., Banzhaf, S., Burkhardt, M., Janzen, N., Niederstrasser, B., Scheytt, T., 2011. Activated soil filters for removal of biocides from contaminated run-off and waste-waters. Chemosphere 85, 1233–1240. https://doi.org/10.1016/j.chemosphere.2011.07.017

Boehler, M., Zwickenpflug, B., Hollender, J., Ternes, T., Joss, A., Siegrist, H., 2012. Removal of micropollutants in municipal wastewater treatment plants by powder-activated carbon. Water Science & Technology 66, 2115. https://doi.org/10.2166/wst.2012.353

Bourgin, M., Beck, B., Boehler, M., Borowska, E., Fleiner, J., Salhi, E., Teichler, R., von Gunten, U., Siegrist, H., McArdell, C.S., 2017. Evaluation of a full-scale wastewater treatment plant upgraded with ozonation and biological post-treatments: Abatement of micropollutants, formation of transformation products and oxidation by-products. Water Research. https://doi.org/10.1016/j.watres.2017.10.036

Carballa, M., Omil, F., Lema, J.M., 2005. Removal of cosmetic ingredients and pharmaceuticals in sewage primary treatment. Water Research 39, 4790–4796. https://doi.org/10.1016/j.watres.2005.09.018

Choubert, J.-M., Martin Ruel, S., Miege, C., Coquery, M., 2017. Rethinking micropollutant removal assessment methods for wastewater treatment plants – how to get more robust

data? Water Science and Technology 75, 2964–2972. https://doi.org/10.2166/wst.2017.181

Falås, P., Wick, A., Castronovo, S., Habermacher, J., Ternes, T.A., Joss, A., 2016. Tracing the limits of organic micropollutant removal in biological wastewater treatment. Water Research 95, 240–249. https://doi.org/10.1016/j.watres.2016.03.009

Frank, J., Ruhl, A.S., Jekel, M., 2015. Impacts of backwashing on granular activated carbon filters for advanced wastewater treatment. Water Research 87, 166–174. https://doi.org/10.1016/j.watres.2015.09.020

Garcia-Ivars, J., Martella, L., Massella, M., Carbonell-Alcaina, C., Alcaina-Miranda, M.-I., Iborra-Clar, M.-I., 2017. Nanofiltration as tertiary treatment method for removing trace pharmaceutically active compounds in wastewater from wastewater treatment plants. Water Research 125, 360–373. https://doi.org/10.1016/j.watres.2017.08.070

Gardner, M., Jones, V., Comber, S., Scrimshaw, M.D., Coello - Garcia, T., Cartmell, E., Lester, J., Ellor, B., 2013. Performance of UK wastewater treatment works with respect to trace contaminants. Science of The Total Environment 456–457, 359–369. https://doi.org/10.1016/j.scitotenv.2013.03.088

Gasperi, J., Rocher, V., Gilbert, S., Azimi, S., Chebbo, G., 2010. Occurrence and removal of priority pollutants by lamella clarification and biofiltration. Water Research 44, 3065–3076. https://doi.org/10.1016/j.watres.2010.02.035

Gobel, A., Mcardell, C., Joss, A., Siegrist, H., Giger, W., 2007. Fate of sulfonamides, macrolides, and trimethoprim in different wastewater treatment technologies. Science of The Total Environment 372, 361–371. https://doi.org/10.1016/j.scitotenv.2006.07.039

Golovko, O., Kumar, V., Fedorova, G., Randak, T., Grabic, R., 2014. Seasonal changes in antibiotics, antidepressants/psychiatric drugs, antihistamines and lipid regulators in a wastewater treatment plant. Chemosphere 111, 418–426. https://doi.org/10.1016/j.chemosphere.2014.03.132

Goulson, D., 2013. An overview of the environmental risks posed by neonicotinoid insecticides. Journal of Applied Ecology 50, 977–987. https://doi.org/10.1111/1365-2664.12111

Grover, D.P., Zhou, J.L., Frickers, P.E., Readman, J.W., 2011. Improved removal of estrogenic and pharmaceutical compounds in sewage effluent by full scale granular activated carbon: Impact on receiving river water. Journal of Hazardous Materials 185, 1005–1011. https://doi.org/10.1016/j.jhazmat.2010.10.005

Hollender, J., Zimmermann, S.G., Koepke, S., Krauss, M., McArdell, C.S., Ort, C., Singer, H., von Gunten, U., Siegrist, H., 2009. Elimination of Organic Micropollutants in a Municipal Wastewater Treatment Plant Upgraded with a Full-Scale Post-Ozonation Followed by Sand Filtration. Environmental Science & Technology 43, 7862–7869. https://doi.org/10.1021/es9014629

Jelic, A., Gros, M., Ginebreda, A., Cespedes-Sánchez, R., Ventura, F., Petrovic, M., Barcelo, D., 2011. Occurrence, partition and removal of pharmaceuticals in sewage water and sludge during wastewater treatment. Water Research 45, 1165–1176. https://doi.org/10.1016/j.watres.2010.11.010

Joss, A., Andersen, H., Ternes, T., Richle, P.R., Siegrist, H., 2004. Removal of Estrogens in Municipal Wastewater Treatment under Aerobic and Anaerobic Conditions:

574 Consequences for Plant Optimization. Environmental Science & Technology 38, 3047–
575 3055. https://doi.org/10.1021/es0351488

576 Joss, A., Keller, E., Alder, A.C., Göbel, A., McArdell, C.S., Ternes, T., Siegrist, H., 2005.
577 Removal of pharmaceuticals and fragrances in biological wastewater treatment. Water
578 Research 39, 3139–3152. https://doi.org/10.1016/j.watres.2005.05.031

579 Kårelid, V., Larsson, G., Björlenius, B., 2017. Pilot-scale removal of pharmaceuticals in
580 municipal wastewater: Comparison of granular and powdered activated carbon
581 treatment at three wastewater treatment plants. Journal of Environmental Management
582 193, 491–502. https://doi.org/10.1016/j.jenvman.2017.02.042

583 Kolpin, D.W., Thurman, E.M., Lee, E.A., Meyer, M.T., Furlong, E.T., Glassmeyer, S.T., 2006.
584 Urban contributions of glyphosate and its degradate AMPA to streams in the United
585 States. Science of The Total Environment 354, 191–197.
586 https://doi.org/10.1016/j.scitotenv.2005.01.028

587 Kostich, M.S., Batt, A.L., Lazorchak, J.M., 2014. Concentrations of prioritized pharmaceuticals
588 in effluents from 50 large wastewater treatment plants in the US and implications for
589 risk estimation. Environmental Pollution 184, 354–359.
590 https://doi.org/10.1016/j.envpol.2013.09.013

591 Kovalova, L., Siegrist, H., von Gunten, U., Eugster, J., Hagenbuch, M., Wittmer, A., Moser,
592 R., McArdell, C.S., 2013. Elimination of Micropollutants during Post-Treatment of
593 Hospital Wastewater with Powdered Activated Carbon, Ozone, and UV. Environ. Sci.
594 Technol. 47, 7899–7908. https://doi.org/10.1021/es400708w

595 Leung, H.W., Minh, T.B., Murphy, M.B., Lam, J.C.W., So, M.K., Martin, M., Lam, P.K.S.,
596 Richardson, B.J., 2012. Distribution, fate and risk assessment of antibiotics in sewage
597 treatment plants in Hong Kong, South China. Environment International, Emerging
598 Environmental Health Issues in Modern China 42, 1–9.
599 https://doi.org/10.1016/j.envint.2011.03.004

600 Lindberg, R.H., Olofsson, U., Rendahl, P., Johansson, M.I., Tysklind, M., Andersson, B.A.V.,
601 2006. Behavior of Fluoroquinolones and Trimethoprim during Mechanical, Chemical,
602 and Active Sludge Treatment of Sewage Water and Digestion of Sludge. Environ. Sci.
603 Technol. 40, 1042–1048. https://doi.org/10.1021/es0516211

604 Lindberg, R.H., Wennberg, P., Johansson, M.I., Tysklind, M., Andersson, B.A.V., 2005.
605 Screening of Human Antibiotic Substances and Determination of Weekly Mass Flows
606 in Five Sewage Treatment Plants in Sweden. Environ. Sci. Technol. 39, 3421–3429.
607 https://doi.org/10.1021/es048143z

608 Loos, R., Carvalho, R., António, D.C., Comero, S., Locoro, G., Tavazzi, S., Paracchini, B.,
609 Ghiani, M., Lettieri, T., Blaha, L., Jarosova, B., Voorspoels, S., Servaes, K., Haglund,
610 P., Fick, J., Lindberg, R.H., Schwesig, D., Gawlik, B.M., 2013. EU-wide monitoring
611 survey on emerging polar organic contaminants in wastewater treatment plant effluents.
612 Water Research 47, 6475–6487. https://doi.org/10.1016/j.watres.2013.08.024

613 Luo, Y., Guo, W., Ngo, H.H., Nghiem, L.D., Hai, F.I., Zhang, J., Liang, S., Wang, X.C., 2014.
614 A review on the occurrence of micropollutants in the aquatic environment and their fate
615 and removal during wastewater treatment. Science of The Total Environment 473–474,
616 619–641. https://doi.org/10.1016/j.scitotenv.2013.12.065

617 Mailler, R., Gasperi, J., Coquet, Y., Buleté, A., Vulliet, E., Deshayes, S., Zedek, S., Mirande-
618 Bret, C., Eudes, V., Bressy, A., Caupos, E., Moilleron, R., Chebbo, G., Rocher, V.,

2016. Removal of a wide range of emerging pollutants from wastewater treatment plant discharges by micro-grain activated carbon in fluidized bed as tertiary treatment at large pilot scale. Science of The Total Environment 542, Part A, 983–996. http://dx.doi.org/10.1016/j.scitotenv.2015.10.153

Mailler, R., Gasperi, J., Coquet, Y., Deshayes, S., Zedek, S., Cren-Olivé, C., Cartiser, N., Eudes, V., Bressy, A., Caupos, E., Moilleron, R., Chebbo, G., Rocher, V., 2015. Study of a large scale powdered activated carbon pilot: Removals of a wide range of emerging and priority micropollutants from wastewater treatment plant effluents. Water Research 72, 315–330. https://doi.org/10.1016/j.watres.2014.10.047

Mailler, R., Gasperi, J., Rocher, V., Gilbert-Pawlik, S., Geara-Matta, D., Moilleron, R., Chebbo, G., 2014. Biofiltration vs conventional activated sludge plants: what about priority and emerging pollutants removal? Environmental Science and Pollution Research 21, 5379–5390. https://doi.org/10.1007/s11356-013-2388-0

Margot, J., Kienle, C., Magnet, A., Weil, M., Rossi, L., de Alencastro, L.F., Abegglen, C., Thonney, D., Chèvre, N., Schärer, M., Barry, D.A., 2013. Treatment of micropollutants in municipal wastewater: Ozone or powdered activated carbon? Science of The Total Environment 461–462, 480–498. https://doi.org/10.1016/j.scitotenv.2013.05.034

Margot, J., Rossi, L., Barry, D.A., Holliger, C., 2015. A review of the fate of micropollutants in wastewater treatment plants: Micropollutants in wastewater treatment plants. Wiley Interdisciplinary Reviews: Water 2, 457–487. https://doi.org/10.1002/wat2.1090

Martin Ruel, S., Esperanza, M., Choubert, J.-M., Valor, I., Budzinski, H., Coquery, M., 2010. On-site evaluation of the efficiency of conventional and advanced secondary processes for the removal of 60 organic micropollutants. Water Science & Technology 62, 2970. https://doi.org/10.2166/wst.2010.989

Martínez-Alcalá, I., Guillén-Navarro, J.M., Fernández-López, C., 2017. Pharmaceutical biological degradation, sorption and mass balance determination in a conventional activated-sludge wastewater treatment plant from Murcia, Spain. Chemical Engineering Journal 316, 332–340. https://doi.org/10.1016/j.cej.2017.01.048

Meinel, F., Ruhl, A.S., Sperlich, A., Zietzschmann, F., Jekel, M., 2015. Pilot-Scale Investigation of Micropollutant Removal with Granular and Powdered Activated Carbon. Water, Air, & Soil Pollution 226. https://doi.org/10.1007/s11270-014-2260-y

Miklos, D.B., Hartl, R., Michel, P., Linden, K.G., Drewes, J.E., Hübner, U., 2018. UV/H2O2 process stability and pilot-scale validation for trace organic chemical removal from wastewater treatment plant effluents. Water Research 136, 169–179. https://doi.org/10.1016/j.watres.2018.02.044

Office fédéral de l'environnement, Division Eaux, 2014. Rapport explificatif concernant la modification de l'ordonnance sur la protection des eaux.

Papageorgiou, M., Kosma, C., Lambropoulou, D., 2016. Seasonal occurrence, removal, mass loading and environmental risk assessment of 55 pharmaceuticals and personal care products in a municipal wastewater treatment plant in Central Greece. Science of The Total Environment 543, 547–569. https://doi.org/10.1016/j.scitotenv.2015.11.047

Park, J., Yamashita, N., Park, C., Shimono, T., Takeuchi, D.M., Tanaka, H., 2017. Removal characteristics of pharmaceuticals and personal care products: Comparison between membrane bioreactor and various biological treatment processes. Chemosphere 179, 347–358. https://doi.org/10.1016/j.chemosphere.2017.03.135

664 Pomiès, M., Choubert, J.-M., Wisniewski, C., Coquery, M., 2013. Modelling of micropollutant
665 removal in biological wastewater treatments: A review. Science of The Total
666 Environment 443, 733–748. https://doi.org/10.1016/j.scitotenv.2012.11.037

667 Popp, M., Hann, S., Mentler, A., Fuerhacker, M., Stingeder, G., Koellensperger, G., 2008.
668 Determination of glyphosate and AMPA in surface and waste water using high-
669 performance ion chromatography coupled to inductively coupled plasma dynamic
670 reaction cell mass spectrometry (HPIC–ICP–DRC–MS). Analytical and Bioanalytical
671 Chemistry 391, 695–699. https://doi.org/10.1007/s00216-008-2037-5

672 Ridder, D.J. de, McConville, M., Verliefde, A.R.D., Aa, L.T.J. van der, Heijman, S.G.J.,
673 Verberk, J.Q.J.C., Rietveld, L.C., Dijk, J.C. van, 2009. Development of a predictive
674 model to determine micropollutant removal using granular activated carbon. Drinking
675 Water Engineering and Science 2, 57–62. https://doi.org/10.5194/dwes-2-57-2009

676 Rocher, V., Paffoni, C., Gonçalves, A., Guérin, S., Azimi, S., Gasperi, J., Moilleron, R., Pauss,
677 A., 2012. Municipal wastewater treatment by biofiltration: comparisons of various
678 treatment layouts. Part 1: assessment of carbon and nitrogen removal. Water Science &
679 Technology 65, 1705. https://doi.org/10.2166/wst.2012.105

680 Ruel, S.M., Choubert, J.M., Esperanza, M., Miège, C., Madrigal, P.N., Budzinski, H., Le
681 Ménach, K., Lazarova, V., Coquery, M., 2011. On-site evaluation of the removal of 100
682 micro-pollutants through advanced wastewater treatment processes for reuse
683 applications. Water Science & Technology 63, 2486.
684 https://doi.org/10.2166/wst.2011.470

685 Sahar, E., David, I., Gelman, Y., Chikurel, H., Aharoni, A., Messalem, R., Brenner, A., 2011.
686 The use of RO to remove emerging micropollutants following CAS/UF or MBR
687 treatment of municipal wastewater. Desalination, Special issue to mark the 45th
688 Anniversary of launching Desalination journal and to honour Professor David Hasson
689 for his enormous support and contribution to Desalination 273, 142–147.
690 https://doi.org/10.1016/j.desal.2010.11.004

691 Schoknecht, U., Mathies, H., Wegner, R., 2016. Biocide leaching during field experiments on
692 treated articles. Environmental Sciences Europe 28. https://doi.org/10.1186/s12302-
693 016-0074-9

694 Skark, C., Zullei-seibert, N., Schöttler, U., Schlett, C., 1998. The Occurrence of Glyphosate in
695 Surface Water. International Journal of Environmental Analytical Chemistry 70, 93–
696 104. https://doi.org/10.1080/03067319808032607

697 Snyder, S.A., Adham, S., Redding, A.M., Cannon, F.S., DeCarolis, J., Oppenheimer, J., Wert,
698 E.C., Yoon, Y., 2007. Role of membranes and activated carbon in the removal of
699 endocrine disruptors and pharmaceuticals. Desalination 202, 156–181.
700 https://doi.org/10.1016/j.desal.2005.12.052

701 Streicher, J., Ruhl, A.S., Gnirß, R., Jekel, M., 2016. Where to dose powdered activated carbon
702 in a wastewater treatment plant for organic micro-pollutant removal. Chemosphere 156,
703 88–94. https://doi.org/10.1016/j.chemosphere.2016.04.123

704 Subedi, B., Kannan, K., 2015. Occurrence and fate of select psychoactive pharmaceuticals and
705 antihypertensives in two wastewater treatment plants in New York State, USA. Science
706 of The Total Environment 514, 273–280.
707 https://doi.org/10.1016/j.scitotenv.2015.01.098

Thiebault, T., Boussafir, M., Le Milbeau, C., 2017a. Occurrence and removal efficiency of pharmaceuticals in an urban wastewater treatment plant: Mass balance, fate and consumption assessment. Journal of Environmental Chemical Engineering 5, 2894–2902. https://doi.org/10.1016/j.jece.2017.05.039

Thiebault, T., Fougère, L., Destandau, E., Réty, M., Jacob, J., 2017b. Temporal dynamics of human-excreted pollutants in wastewater treatment plant influents: Toward a better knowledge of mass load fluctuations. Science of The Total Environment 596–597, 246–255. https://doi.org/10.1016/j.scitotenv.2017.04.130

Verlicchi, P., Al Aukidy, M., Zambello, E., 2012. Occurrence of pharmaceutical compounds in urban wastewater: Removal, mass load and environmental risk after a secondary treatment—A review. Science of The Total Environment, Special Section - Arsenic in Latin America, An Unrevealed Continent: Occurence, Health Effects and Mitigation 429, 123–155. https://doi.org/10.1016/j.scitotenv.2012.04.028

Vulliet, E., Cren-Olivé, C., Grenier-Loustalot, M.-F., 2011. Occurrence of pharmaceuticals and hormones in drinking water treated from surface waters. Environ Chem Lett 9, 103–114. https://doi.org/10.1007/s10311-009-0253-7

Weston Donald P., Ramil Heather L., Lydy Michael J., 2013. Pyrethroid insecticides in municipal wastewater. Environmental Toxicology and Chemistry 32, 2460–2468. https://doi.org/10.1002/etc.2338

Yan, Q., Gao, X., Huang, L., Gan, X.-M., Zhang, Y.-X., Chen, Y.-P., Peng, X.-Y., Guo, J.-S., 2014. Occurrence and fate of pharmaceutically active compounds in the largest municipal wastewater treatment plant in Southwest China: Mass balance analysis and consumption back-calculated model. Chemosphere 99, 160–170. https://doi.org/10.1016/j.chemosphere.2013.10.062

Yang, X., Flowers, R.C., Weinberg, H.S., Singer, P.C., 2011. Occurrence and removal of pharmaceuticals and personal care products (PPCPs) in an advanced wastewater reclamation plant. Water Research 45, 5218–5228. https://doi.org/10.1016/j.watres.2011.07.026

Yang, Y., Ok, Y.S., Kim, K.-H., Kwon, E.E., Tsang, Y.F., 2017. Occurrences and removal of pharmaceuticals and personal care products (PPCPs) in drinking water and water/sewage treatment plants: A review. Science of The Total Environment 596–597, 303–320. https://doi.org/10.1016/j.scitotenv.2017.04.102

Yao, W., Ur Rehman, S.W., Wang, H., Yang, H., Yu, G., Wang, Y., 2018. Pilot-scale evaluation of micropollutant abatements by conventional ozonation, UV/O3, and an electro-peroxone process. Water Research 138, 106–117. https://doi.org/10.1016/j.watres.2018.03.044

Yu, J., Hu, J., Tanaka, S., Fujii, S., 2009. Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in sewage treatment plants. Water Research 43, 2399–2408. https://doi.org/10.1016/j.watres.2009.03.009

Zietzschmann, F., Worch, E., Altmann, J., Ruhl, A.S., Sperlich, A., Meinel, F., Jekel, M., 2014. Impact of EfOM size on competition in activated carbon adsorption of organic micro-pollutants from treated wastewater. Water Research 65, 297–306. https://doi.org/10.1016/j.watres.2014.07.043