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## **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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1 **Title**

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

4

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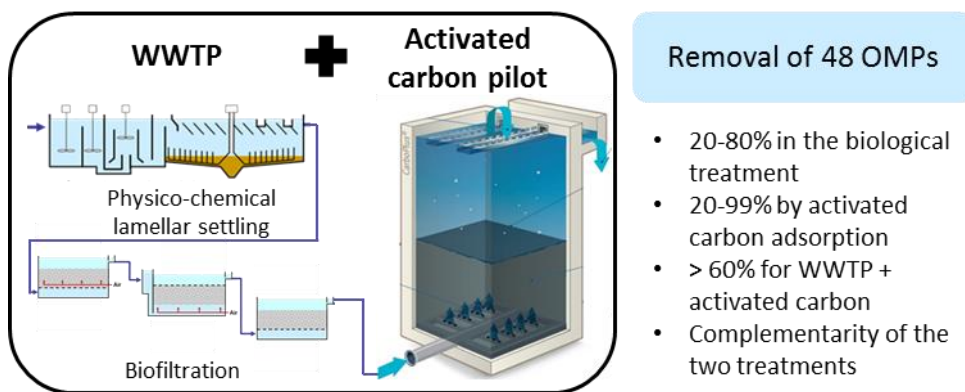
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## 26 Highlights

- 27 • Full-scale study of micropollutants removal in a WWTP with an advanced treatment.
- 28 • Most micropollutants were partially removed (20-80%) in the biological treatment.
- 29 • Micropollutants were removed (20-99%) by activated carbon adsorption.
- 30 • Compounds not removed in the WWTP (< 50%) were well removed by adsorption (>  
31 70%).
- 32 • Removals were higher than 60% over the upgraded WWTP.

## 33 Graphical abstract



## 35 Abstract

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

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

## 52 **Keywords**

53 Organic micropollutants; Wastewater treatment plant; Activated carbon adsorption

## 54 **Introduction**

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

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

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

81 The first regulation in Europe about OMPs removal (Office fédéral de l'environnement,  
82 Division Eaux, 2014) fixes a removal of 80% in comparison to the raw wastewater for at least  
83 6 compounds among 12 compounds selected based on their behavior in WWTP (amisulpride,  
84 benzotriazole, candesartan, carbamazepine, citalopram, clarithromycin, diclofenac,  
85 hydrochlorothiazide, irbesartan, mecoprop, metoprolol and venlafaxine). Nevertheless, to  
86 achieve such performances conventional installations should often be upgraded with an  
87 advanced treatment. Many studies were conducted on OMPs removal either in WWTP or in  
88 advanced treatments but only a few investigated an upgraded WWTP (Bourgin et al., 2017;  
89 Hollender et al., 2009; Margot et al., 2013).

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

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

## 96 **2. Materials and methods**

### 97 *2.1. Wastewater treatment plant and activated carbon advanced treatment descriptions*

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

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

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

## 117 *2.2. Sampling campaigns*

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

## 127 *2.3. Analytical procedures*

128 Conventional wastewater quality parameters were analyzed by the certified WWTP laboratory:  
129 pH, temperature, total organic carbon (TOC), dissolved organic carbon (DOC), chemical  
130 oxygen demand (COD), biological oxygen demand in 5 days (BOD<sub>5</sub>), total Kjeldahl nitrogen  
131 (TKN), NH<sub>4</sub><sup>+</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, total phosphorus (TP), PO<sub>4</sub><sup>3-</sup> and total suspended solids (TSS) (Table  
132 S2, SI). Results are given in the supporting material (Table S3, SI).

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

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

#### 143 *2.4. Data processing*

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

### 161 **3. Results and discussion**



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

### 171 *3.1. Elimination of OMPs in the WWTP*

172 4 compounds (imidacloprid, propranolol, roxithromycin and tetracycline) were not quantified  
173 in WWTP influent and the primary effluent due to interferences with other compounds eluting  
174 at the same retention time. 4 other compounds (cypermethrin, irbesartan, norfloxacin and  
175 sulfadiazine) were quantified in the WWTP influent, the primary and the WWTP effluent but  
176 their removal was not calculated because of high uncertainty (values close to their respective  
177 LOQ).

#### 178 3.1.1 OMPs concentration in the WWTP influent

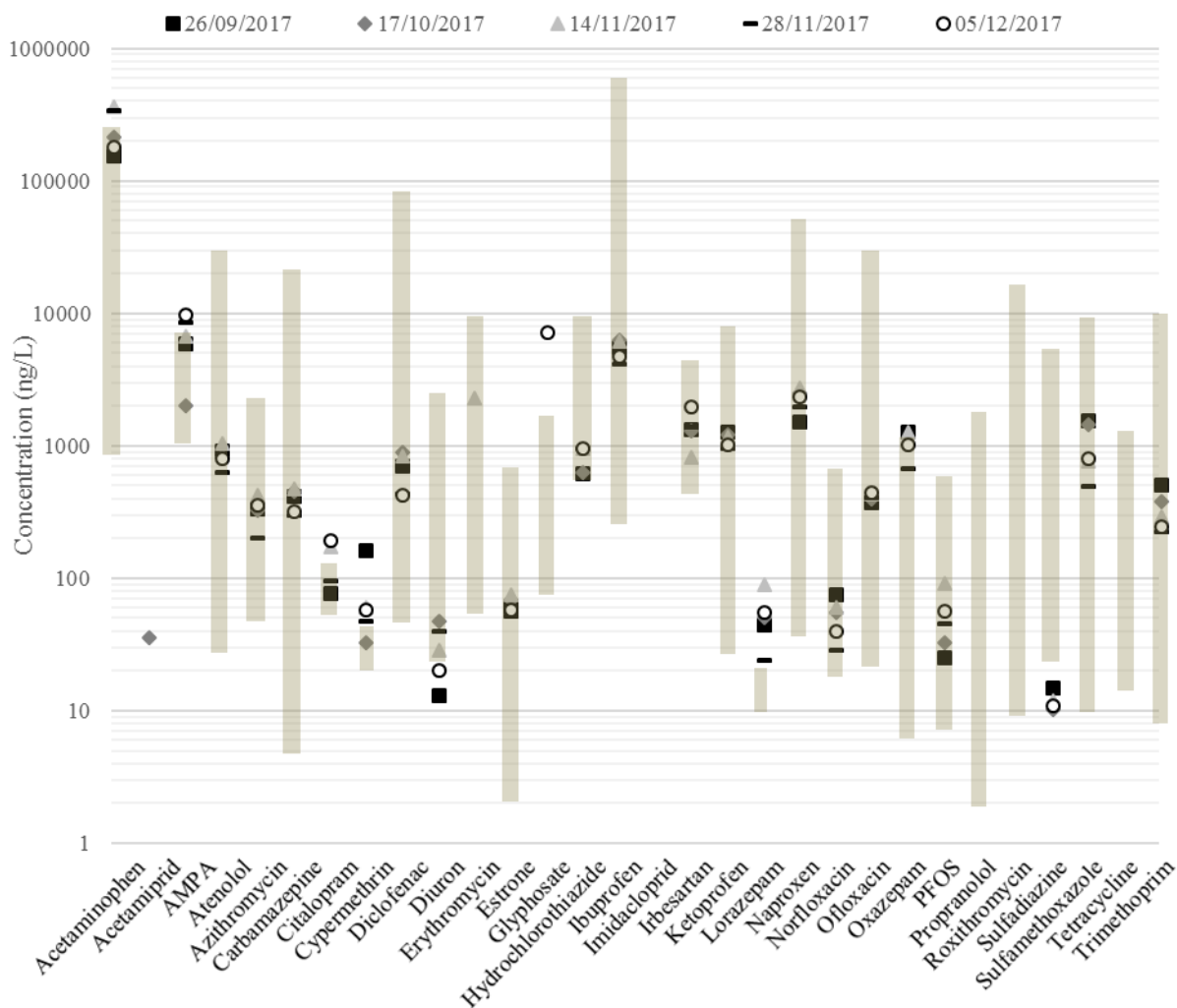
179 As presented in Figure 1 and Table 1, average concentrations of OMPs in the raw influent  
180 ranged between the minimal and the maximal value found in other studies for most compounds.  
181 Variations of concentrations in the influent can be substantial, depending on the country and  
182 seasonal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

185 were analgesics (acetaminophen, diclofenac, ibuprofen, ketoprofen and naproxen). Other  
186 commonly used pharmaceuticals (e.g., irbesartan, oxazepam, sulfamethoxazole) were found at  
187 concentrations higher than 1,000 ng/L. 7 other pharmaceuticals (atenolol, azithromycin,  
188 carbamazepine, citalopram, hydrochlorothiazide, ofloxacin and trimethoprim) had an average  
189 concentration ranging from 100 ng/L to 1,000 ng/L. 4 pharmaceuticals (estrone, lorazepam,  
190 norfloxacin and sulfadiazine) were found at concentrations below 100 ng/L, reflecting a lower  
191 consumption than other pharmaceuticals. Glyphosate was detected only during the fifth  
192 campaign at a high concentration (7,400 ng/L). Its degradation product, AMPA, was detected  
193 during each campaign with a high average concentration (6,840 ng/L) and a maximal value  
194 during the fifth campaign (10,100 ng/L). Despite the absence of agricultural land, one herbicide  
195 (diuron) and two insecticides (acetamiprid and cypermethrin) were found at trace levels (< 100  
196 ng/L). Diuron has been banned in France since 2003 for agricultural use but is still used as a  
197 biocide, especially in paints and plaster of facades (Bester et al., 2011; Schoknecht et al., 2016).  
198 Acetamiprid and cypermethrin are used in household chemicals and agriculture (Goulson,  
199 2013). The surfactant PFOS was also found at trace levels ( $52 \pm 24$  ng/L).

### 200 3.1.2. Primary treatment

201 OMPs average concentration in the WWTP influent and the primary effluent and their average  
202 removal are presented in Table 1. Despite the elimination of suspended solids ( $78 \pm 17\%$ , Table  
203 S2, SI), most OMPs were not affected by the primary treatment (removals between -10% and  
204 13% for 14 compounds). Therefore, sorption onto particles is not an important removal  
205 mechanism for most pharmaceuticals and pesticides. Gasperi et al., (2010) observed a trend  
206 between the removal of OMPs and their hydrophobicity in the same treatment unit, but here no  
207 correlation or trend were observed with the log D (corrected log  $K_{ow}$  at pH = 8). This different

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



210  
 211 Figure 1. Concentration of 30 OMPs in the WWTP influent for each sampling campaign and  
 212 range of values found in the literature (grey bars). Logarithmic-scale. References for literature  
 213 values are indicated in Table 1.

214 3 OMPs exhibited a moderate removal in the primary treatment with high variations among the  
 215 5 campaigns: azithromycin ( $27 \pm 31\%$ ), citalopram ( $31 \pm 14\%$ ) and glyphosate (58%, only in  
 216 the fifth campaign), but their removal remains unclear. 5 OMPs exhibited negative removals  
 217 with high variations (standard deviation SD between 10% and 48%): AMPA (-17%),

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

228 The removal of OMPs in the primary treatment was significantly higher (ANOVA, p-value <  
229 0.001, R) during the fourth (28/11/17, average removal = 6%) and the fifth sampling campaigns  
230 (05/12/17, average removal = 13%) as compared to the first campaign (26/09/17, average  
231 removal = - 24%) and the second campaign (17/10/17, average removal = - 19%). This indicates  
232 that the performances of the primary treatment depend on the period of the year, probably due  
233 to the seasonal variation of OMPs concentration levels in WWTP influent (Figure S3, SI).  
234 Moreover, the rain event that happened during the fourth campaign (28/11/17) didn't seem to  
235 have an impact, and removals were independent of the treated flow.

### 236 3.1.3. Biological treatment

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

240 6 pharmaceuticals (acetaminophen, erythromycin, estrone, ibuprofen, ketoprofen and  
241 naproxen) and 1 pesticide (glyphosate) exhibited average removals higher than 80% with low  
242 variations ( $SD < 6\%$ ). Average concentrations in the WWTP effluent were below 1,000 ng/L  
243 except for acetaminophen (2,600 ng/L). Similar concentrations and removals were found in  
244 other studies for those compounds (Table 1), and the removal of some analgesics (i.e.,  
245 ibuprofen, ketoprofen and naproxen) was generally higher in our study (Margot et al., 2013).  
246 Biodegradation is the main removal mechanism for these compounds (Jelic et al., 2011; Joss et  
247 al., 2005, 2004; Margot et al., 2015) but it remains unclear for glyphosate (no studies available  
248 about its biodegradation).

249 5 OMPs (AMPA, atenolol, lorazepam, PFOS and trimethoprim) were moderately eliminated  
250 with average removals from 50% to 80%. Low variations were observed (average  $SD = 7\%$ )  
251 except for AMPA ( $SD = \pm 33\%$ ) and trimethoprim ( $SD = \pm 18\%$ ). Removals of these  
252 compounds were more diverse in other studies, ranging from 0% to 80% (Bourgin et al., 2017;  
253 Hollender et al., 2009; Luo et al., 2014; Margot et al., 2013; Verlicchi et al., 2012). Contrary to  
254 our results, no elimination was previously reported for AMPA and PFOS (Bourgin et al., 2017;  
255 Gardner et al., 2013; Martin Ruel et al., 2010). Atenolol and trimethoprim are primarily  
256 biodegraded (Gobel et al., 2007; Jelic et al., 2011; Margot et al., 2015), while PFOS is the only  
257 substance preferentially removed by sorption onto sludges (Margot et al., 2015).

258 6 pharmaceuticals (acetamiprid, carbamazepine, citalopram, diclofenac, ofloxacin and  
259 oxazepam) were persistent with average removals lower than 30% and moderate variations ( $SD$   
260 ranging from 10% to 20%) except for oxazepam ( $SD = 4\%$ ). Similar removals were reported  
261 for carbamazepine, diclofenac and oxazepam (Hollender et al., 2009; Luo et al., 2014; Margot  
262 et al., 2013; Ruel et al., 2011), but lower removals for citalopram (Bourgin et al., 2017; Subedi  
263 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<sup>4</sup>-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):  $\text{NH}_4^+$ , TKN and  $\text{BOD}_5$  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  $\text{NH}_4^+$  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.



313 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  
 314 (WWTP removal); and values reported in the literature for similar processes. The OMPs used as indicator substances in Switzerland are in bold. \*SD = standard deviation between 5 campaigns.

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

315 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.,  
 316 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.,  
 317 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

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

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

364 OMPs removal were significantly higher (ANOVA, p-value < 0.01, R) during the first  
365 (26/09/17: total average removal = 75%) and second campaigns (17/10/17: 75%) than during  
366 the 3 others (63%, 56% and 56%) (Figure S4, SI). A first hypothesis would be that the  
367 performances of the pilot depend on the period of the year, mainly under the influence of the  
368 water temperature. However, the activated carbon bed mass was higher during the first  
369 campaign (1200 kg) and a reduced flow occurred during the second campaign, increasing  
370 contact time and the dose of activated carbon, which explains the higher removals. Moreover,  
371 the rain even that happened during the fourth campaign (28/11) had an effect on the removal of  
372 most OMPs. Lower removals were achieved in comparison to the other sampling campaigns,  
373 even if hydraulic conditions and activated carbon doses were identical. Similar results about  
374 the effect of rain events (i.e., degraded WWTP configuration) were found in a previous study  
375 (Mailler et al., 2015), explained by the hypothesis of higher levels of organic matter and higher  
376 concentrations of OMPs. However, the quality of the WWTP effluent in our study (i.e.,  
377 dissolved organic carbon, total nitrogen and OMPs) didn't vary statistically from one campaign  
378 to another (ANOVA, p-value = 0.804, R) (Table S3, SI). In the previous study (Mailler et al.,  
379 2016), the removal of 13 pharmaceuticals was less impacted by the period ( $\pm 5$  to 25%) than by  
380 the treatment rate (20 g/m<sup>3</sup> vs 10 g/m<sup>3</sup>). Moreover, the UV<sub>254</sub> removal, used as surrogate for the  
381 removals of the 13 pharmaceuticals, was not affected by the period of the year (standard  
382 deviation <  $\pm 2\%$ ). Therefore, the studied period can be considered representative of the  
383 performance of the process over the year.

### 384 3.2.2. Relative contribution of advanced treatment compared to the conventional process

385 The average concentration of OMPs in the WWTP influent and the pilot effluent are presented  
386 in Table 3, as well as their overall removal. No data are presented for cypermethrin due to low  
387 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%).

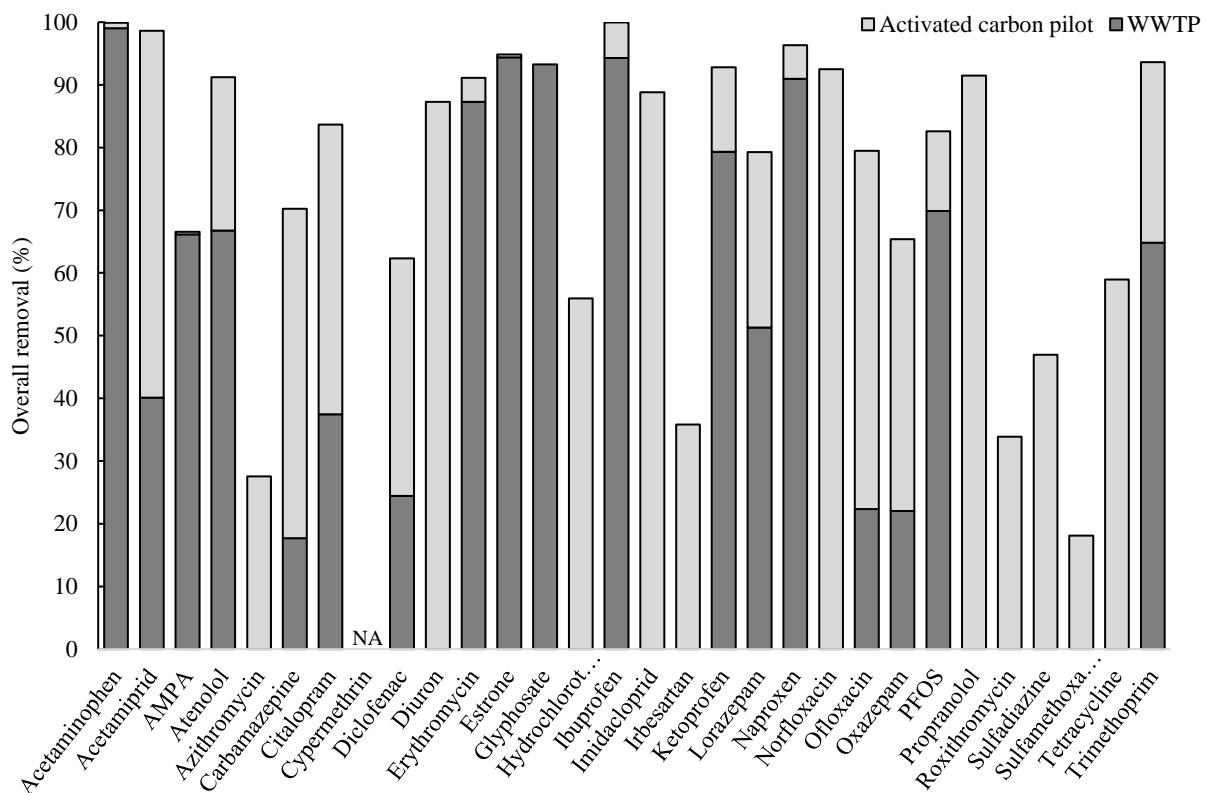
Table 2. Concentration of 30 OMPs quantified in the effluents of the WWTP and of the pilot, removal rate achieved within the advanced treatment, and values reported in the literature for similar processes. The OMPs used as indicator substances in Switzerland are in bold. <sup>a</sup>SD = standard deviation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

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

400 Their overall removal reached values higher than 80%. The activated carbon adsorption had a  
401 more important impact on 7 substances (acetamiprid, AMPA, carbamazepine, citalopram,  
402 diclofenac, ofloxacin, oxazepam) that were partially eliminated in the WWTP (from 13% to  
403 40%): their additional removal ranged from 38% to 59% in the advanced treatment, with a high  
404 overall removal (from 62% to 99%). 4 OMPs were not quantified in the WWTP influent  
405 (imidacloprid, propranolol, roxithromycin and tetracycline) and the overall removal of 3 other  
406 substances (irbesartan, norfloxacin and sulfadiazine) was not calculated due to low  
407 concentrations in the primary treatment effluent. Therefore, only the pilot contributed to the  
408 elimination (34% to 92%) of those 7 OMPs. Finally, 4 OMPs were negatively removed in the  
409 WWTP (azithromycin, diuron, hydrochlorothiazide and sulfamethoxazole) but the activated  
410 carbon adsorption was sufficient to obtain positive overall removals (18% to 87%).

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

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



432  
 433 Figure 2. Overall removal of 30 OMPs over the WWTP and the activated carbon advanced  
 434 treatment. The contribution of the WWTP (dark grey) and the activated carbon advanced  
 435 treatment (light grey) to the overall removal are displayed for each OMPs. NA: no data are  
 436 presented due to low concentrations in all samples (< 5 times LOQ).

437



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. <sup>a</sup>SD = standard deviation between 5 campaigns.

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

### 441 3.2.3. Relevance of the advanced treatment

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

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

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

### 458 **Conclusions**

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

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

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