

## Supplementary Information

### Combination of ozonation with GAC. AIX and biochar post-treatment for removal of pharmaceuticals and transformation products from municipal WWTP effluent

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**Table S1.** Characterization of the effluent wastewater used in the tests.

Parameter	Unit	Mean±standard deviation
pH	-	7.53±0.12
Temperature	°C	18.6±1.12
Dissolved organic carbon (DOC)	mg/L	9.42±3.70
Total organic carbon (TOC)	mg/L	9.98±4.12
Alkalinity	mg CaCO <sub>3</sub> /L	166±38
Total suspended solids (TSS)	mg/L	11.8±8.25
Chemical oxygen demand (COD)	mg/L	28.2±11.2
Nitrite nitrogen (NO <sub>2</sub> -N)	mg/L	0.01±0.01
Nitrate nitrogen (NO <sub>3</sub> -N)	mg/L	8.21±3.02
Ammonia nitrogen (NH <sub>3</sub> -N)	mg/L	0.11±0.13
UV <sub>254</sub>	cm <sup>-1</sup>	0.14±0.04
Bromide (Br <sup>-</sup> )	mg/L	0.53±0.41
Bromate (BrO <sub>3</sub> <sup>-</sup> )	µg/L	<5.0
Specific ultraviolet absorbance (SUVA <sub>254</sub> )	mg/L.cm	1.60±0.21

**Table S2.** LOQ values of all selected pharmaceuticals and their metabolites

<b>Pharmaceutical</b>	<b>LOQ</b>
Atenolol	< 7.2
Bicalutamide	< 8.6
Carbamazepine	< 5.4
Cetirizine	< 5.6
Citalopram	< 4.4
Clarithromycin	< 7.1
Clindamycin	< 7.7
Desvenlafaxine	< 5.1
Diclofenac	< 3.1
Erythromycin	< 12
Fexofenadine	< 8.7
Fluconazole	< 6.4
Furosemide	< 7.6
Irbesartan	< 7
Lamotrigine	< 5.5
Losartan	< 11
Metoprolol	< 7.1
Oxazepam	< 4.9
Propranolol	< 8.1
Sertraline	< 4.9
Sulfamethoxazole	< 11
Tramadol	< 6.5
Trimethoprim	< 11
Venlafaxine	< 4.2
<b>Metabolite</b>	<b>LOQ</b>
4 hydroxy diclofenac	< 2.6
4-chloro 5-sulfamoylanthranilic	< 75

acid	
Alpha hydroxymetoprolol	< 6.2
Citalopram related compound E	< 5.2
Fluconazole N-oxide	< 7.6
N-demethylerythromycin	< 11
Norsertaline	< 220

**Table S3.** Concentrations of the selected pharmaceuticals in the filtered effluent and after each treatment step

<b>Ozone dosage: 0.28 g O<sub>3</sub>/ g DOC - Day 1</b>							
Micropollutant	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Atenolol	175	147	< 7.2	146	149	< 7.2	31.9
Metoprolol	1773	1615	< 7.1	1637	1383	< 7.1	180
Carbamazepine	387	179	< 5.4	142	161	< 5.4	54.3
Cetirizine	240	114	< 5.6	56.4	94.1	< 5.6	29.6
Citalopram	179	58.8	< 4.4	55.1	48.7	< 4.4	< 4.4
Oxazepam	466	332	< 4.9	258	294	< 4.9	78.5
Lamotrigine	2005	1164	17.4	1068	1055	8.9	296
Bicalutamide	6517	2228	< 8.6	1085	1844	< 8.6	68.5
Clarithromycin	203	18.8	< 7.1	15.8	11.3	< 7.1	< 7.1
Clindamycin	630	74.5	< 7.7	70.5	56.0	< 7.7	8.2
Fexofenadine	8387	1868	< 8.7	1488	1332	< 8.7	255
Propranolol	638	118	< 8.1	100	81.2	< 8.1	< 8.1
Tramadol	502	593	< 6.5	519	450	< 6.5	166
Fluconazole	150	187	< 6.4	168	157	< 6.4	62.8
Diclofenac	540	198	< 3.1	71.8	163	< 3.1	44.9
Furosemide	302	69.5	< 7.6	26.2	67.6	< 7.6	29.3
Irbesartan	69	54.5	< 7	22.6	49.4	< 7	15.8
Losartan	558	344	< 11	144	288	< 11	80.5
Sulfamethoxazole	188	134	< 11	74.9	87.6	< 11	53.5

Trimethoprim	176	62.4	< 11	60.5	52.4	< 11	< 11
Erythromycin	2094	834	< 12	850	800	< 12	342
Norsertaline	< 220	< 220	< 220	< 220	< 220	< 220	< 220
Sertraline	76.8	19.4	< 4.9	11.2	11.5	< 4.9	< 4.9
Venlafaxine	482	392	< 4.2	344	299	< 4.2	108
Desvenlafaxine	544	359	< 5.1	329	287	< 5.1	88.2

<b>Ozone dosage: 0.28 g O<sub>3</sub>/ g DOC - Day 5</b>							
Micropollutant	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Atenolol	153	124	< 7.2	138	146	< 7.2	37.6
Metoprolol	1876	1330	< 7.1	1771	1622	16.0	186
Carbamazepine	378	76.0	< 5.4	84.1	85.7	19.1	64.5
Cetirizine	261	36.0	< 5.6	22.0	39.3	< 5.6	17.9
Citalopram	181	23.7	< 4.4	25.9	25.3	< 4.4	< 4.4
Oxazepam	461	304	< 4.9	254	342	9.1	82.8
Lamotrigine	2388	1067	< 5.5	1118	1217	61.9	348
Bicalutamide	6828	1452	< 8.6	662	1418	< 8.6	62.3
Clarithromycin	543	12.2	< 7.1	11.1	9.8	< 7.1	< 7.1
Clindamycin	988	10.6	< 7.7	16.0	16.3	< 7.7	< 7.7
Fexofenadine	11154	760	< 8.7	668	730	16.4	209
Propranolol	775	25.6	< 8.1	28.9	22.9	< 8.1	< 8.1
Tramadol	474	290	< 6.5	255	342	25.9	165
Fluconazole	145	166	< 6.4	150	197	15.6	94.4
Diclofenac	538	41.8	< 3.1	11.1	61.9	5.7	34.2
Furosemide	245	16.0	< 7.6	8.7	14.9	< 7.6	< 7.6
Irbesartan	85	48.6	< 7	19.7	53.8	< 7	19.2
Losartan	842	216	< 11	89.9	261	12.1	85.5
Sulfamethoxazole	328	40.9	< 11	43.6	85.4	12.0	48.1
Trimethoprim	221	13.0	< 11	16.0	13.5	< 11	< 11
Erythromycin	1444	513	< 12	666	624	58.5	434



Sertraline	92.6	17.7	< 4.9	11.4	9.1	< 4.9	< 4.9
Venlafaxine	678	197	< 4.2	255	247	73.4	170
Desvenlafaxine	718	158	< 5.1	203	205	59.7	109

<b>Ozone dosage: 0.96 g O<sub>3</sub>/ g DOC - Day 1</b>							
Micropollutant	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Atenolol	346	65.0	< 7.2	61.7	61.8	< 7.2	11.5
Metoprolol	2379	487	< 7.1	561	487	< 7.1	58.9
Carbamazepine	4166	16.7	< 5.4	12.0	14.9	< 5.4	< 5.4
Cetirizine	2381	8.0	< 5.6	< 5.6	7.9	< 5.6	< 5.6
Citalopram	2845	15.2	< 4.4	12.3	13.1	< 4.4	< 4.4
Oxazepam	5152	179	< 4.9	84.3	177	< 4.9	42.8
Lamotrigine	25922	915	< 5.5	715	859	7.3	285
Bicalutamide	19659	350	< 8.6	18.0	322	< 8.6	23.3
Clarithromycin	2119	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1
Clindamycin	3280	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7
Fexofenadine	14250	84.8	< 8.7	52.4	81.3	< 8.7	22.9
Propranolol	2118	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1
Tramadol	4527	157	< 6.5	147	125	< 6.5	50.6
Fluconazole	465	82.5	< 6.4	72.7	68.3	< 6.4	30.7
Diclofenac	405	79.5	89.5	64.5	98.5	83.8	137
Furosemide	1434	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6
Irbesartan	803	22.2	< 7	< 7	24.4	< 7	< 7
Losartan	1322	15.3	< 11	< 11	19.1	< 11	< 11
Sulfamethoxazole	415	15.3	< 11	< 11	19.1	< 11	< 11
Trimethoprim	46.0	27.8	< 11	21.0	27.3	< 11	< 11
Erythromycin	156	< 12	< 12	< 12	< 12	< 12	< 12
Norsertaline	< 220	< 220	< 220	< 220	< 220	< 220	< 220
Sertraline	102	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9
Venlafaxine	13402	88.6	< 4.2	85.2	80.9	< 4.2	26.3

Desvenlafaxine	18508	71.5	< 5.1	68.7	66.1	< 5.1	22.6
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<b>Ozone dosage: 0.96 g O<sub>3</sub>/ g DOC - Day 5</b>							
Micropollutant	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Atenolol	335	204	< 7.2	48.0	47.9	< 7.2	< 7.2
Metoprolol	1542	1165	< 7.1	417	313	19.3	21.3
Carbamazepine	3185	41.2	< 5.4	< 5.4	6.2	< 5.4	< 5.4
Cetirizine	1833	27.3	< 5.6	< 5.6	< 5.6	< 5.6	< 5.6
Citalopram	1977	46.2	< 4.4	9.1	10.1	< 4.4	< 4.4
Oxazepam	3334	309	< 4.9	107	113	18.1	23.9
Lamotrigine	16776	1366	< 5.5	859	901	211	226
Bicalutamide	17000	609	< 8.6	189	293	17.0	9.2
Clarithromycin	1562	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1
Clindamycin	1979	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7
Fexofenadine	9242	277	< 8.7	73.4	75.2	11.7	14.3
Propranolol	1481	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1
Tramadol	2782	451	< 6.5	69.6	82.0	13.0	21.7
Fluconazole	340	117	< 6.4	52.9	62.7	11.8	20.0
Diclofenac	264	48.3	19.3	15.4	22.2	24.7	26.3
Furosemide	1278	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6
Irbesartan	627	16.3	< 7	13.3	17.8	< 7	< 7
Losartan	791	25.7	< 11	16.9	21.4	< 11	11.9
Sulfamethoxazole	388	25.7	< 11	16.9	21.4	< 11	11.9
Trimethoprim	32.3	46.5	< 11	18.3	17.4	< 11	< 11
Erythromycin	167	< 12	< 12	< 12	< 12	< 12	< 12
Norsertaline	< 220	< 220	< 220	< 220	< 220	< 220	< 220
Sertraline	89.8	20.1	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9
Venlafaxine	9103	288	< 4.2	59.3	57.3	10.8	19.1
Desvenlafaxine	12325	228	< 5.1	44.0	45.3	9.7	15.1

<b>Ozone dosage: 0.96 g O<sub>3</sub>/ g DOC - Day 12</b>							
Micropollutant	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Atenolol	387	25.9	< 7.2	15.8	16.9	< 7.2	< 7.2
Metoprolol	1481	127	< 7.1	89.1	104	< 7.1	< 7.1
Carbamazepine	3724	< 5.4	< 5.4	< 5.4	< 5.4	< 5.4	< 5.4
Cetirizine	2335	< 5.6	< 5.6	< 5.6	< 5.6	< 5.6	< 5.6
Citalopram	2446	< 4.4	< 4.4	< 4.4	< 4.4	< 4.4	< 4.4
Oxazepam	3734	93.9	< 4.9	70.2	68.8	< 4.9	13.2
Lamotrigine	17861	840	< 5.5	784	715	35.7	147
Bicalutamide	12520	192	< 8.6	115	161	< 8.6	< 8.6
Clarithromycin	2031	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1
Clindamycin	1581	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7
Fexofenadine	7578	21.2	< 8.7	18.0	17.7	< 8.7	< 8.7
Propranolol	1458	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1
Tramadol	3049	14.9	< 6.5	11.4	12.5	< 6.5	8.6
Fluconazole	344	40.3	< 6.4	28.7	30.7	< 6.4	14.3
Diclofenac	263	117	41.6	29.9	25.4	129	79.1
Furosemide	1212	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6
Irbesartan	777	13.0	< 7	8.0	10.5	< 7	< 7
Losartan	1101	16.4	< 11	< 11	11.9	< 11	< 11
Sulfamethoxazole	110.	< 11	< 11	< 11	< 11	< 11	< 11
Trimethoprim	173	< 11	< 11	< 11	< 11	< 11	< 11
Erythromycin	212	< 12	< 12	< 12	< 12	< 12	< 12
Norsertaline	< 220	< 220	< 220	< 220	< 220	< 220	< 220
Sertraline	139	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9
Venlafaxine	10190	18.5	< 4.2	13.5	14.8	< 4.2	12.4
Desvenlafaxine	13438	11.0	< 5.1	8.6	9.4	< 5.1	8.1

<b>Ozone dosage: 2.17 g O<sub>3</sub>/ g DOC - Day 1</b>							
Micropollutant	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2



Atenolol	304	23.1	< 7.2	21.9	21.8	< 7.2	< 7.2
Metoprolol	1014	97.2	< 7.1	76.2	70.9	< 7.1	< 7.1
Carbamazepine	4548	< 5.4	< 5.4	< 5.4	< 5.4	< 5.4	< 5.4
Cetirizine	2552	< 5.6	< 5.6	< 5.6	< 5.6	< 5.6	< 5.6
Citalopram	2537	< 4.4	< 4.4	< 4.4	< 4.4	< 4.4	< 4.4
Oxazepam	2856	65.9	< 4.9	49.4	67.6	< 4.9	< 4.9
Lamotrigine	12315	660	< 5.5	569	535	< 5.5	40.5
Bicalutamide	10386	136	< 8.6	15.4	185	< 8.6	< 8.6
Clarithromycin	1640	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1
Clindamycin	1573	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7
Fexofenadine	8520	16.5	< 8.7	10.9	16.0	< 8.7	< 8.7
Propranolol	1731	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1
Tramadol	1490	13.9	< 6.5	13.0	12.3	< 6.5	< 6.5
Fluconazole	231	26.6	< 6.4	26.0	24.4	< 6.4	< 6.4
Diclofenac	135	50.6	159	61.0	59.1	92.8	101
Furosemide	551	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6
Irbesartan	939	8.6	< 7	< 7	7.4	< 7	< 7
Losartan	1481	< 11	< 11	< 11	< 11	< 11	< 11
Sulfamethoxazole	119	< 11	< 11	< 11	< 11	< 11	< 11
Trimethoprim	147	< 11	< 11	< 11	< 11	< 11	< 11
Erythromycin	454	19.0	< 12	23.1	19.8	< 12	< 12
Norsertraline	< 220	< 220	< 220	< 220	< 220	< 220	< 220
Sertraline	110	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9
Venlafaxine	7629	19.3	< 4.2	18.9	20.7	< 4.2	< 4.2
Desvenlafaxine	7759	10.8	< 5.1	9.6	10.3	< 5.1	< 5.1

<b>Ozone dosage: 2.17 g O<sub>3</sub>/ g DOC - Day 5</b>							
Micropollutant	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Atenolol	310	20.7	< 7.2	20.3	19.0	< 7.2	< 7.2
Metoprolol	922	72.3	< 7.1	67.0	62.6	< 7.1	< 7.1

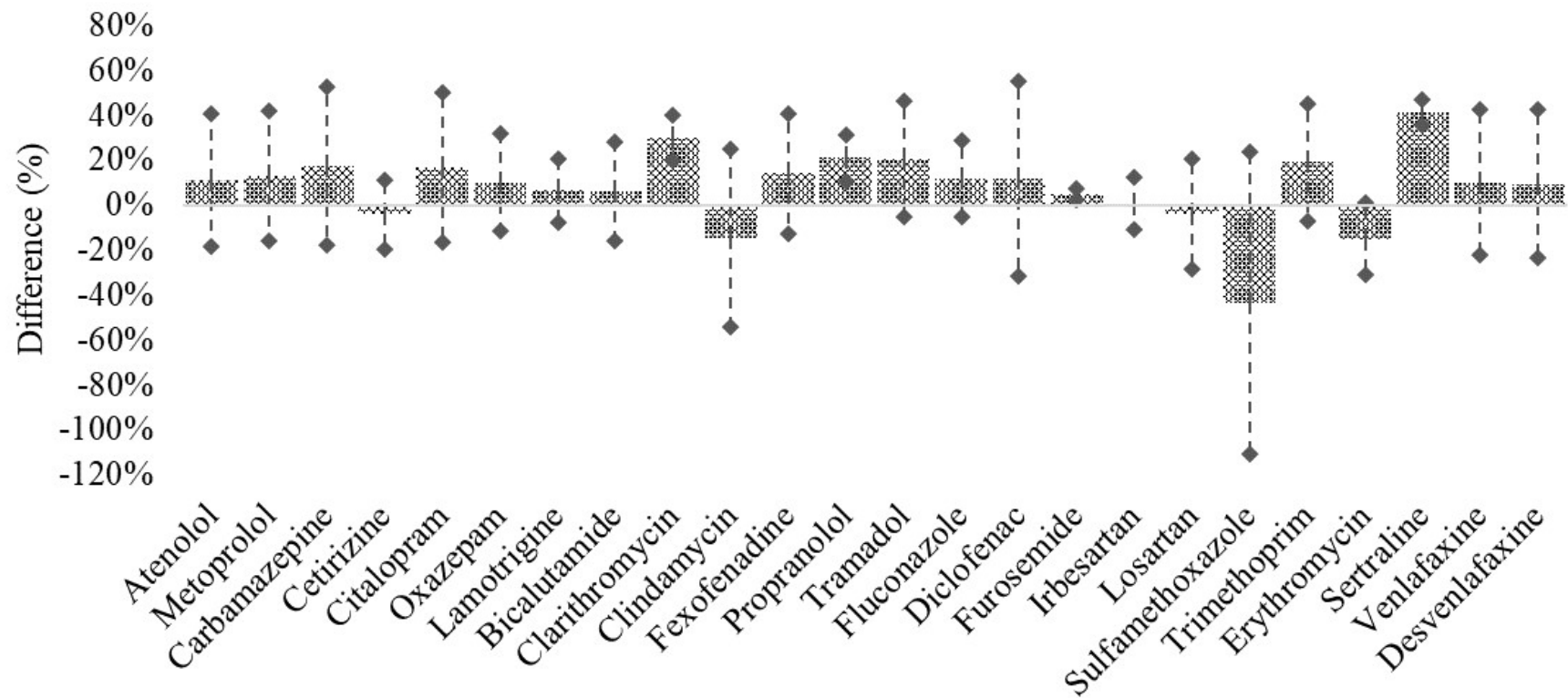


Citalopram	926	< 4.4	< 4.4	< 4.4	< 4.4	< 4.4	< 4.4
Oxazepam	1046	34.8	< 4.9	34.0	34.6	5.2	8.3
Lamotrigine	6826	442	< 5.5	457	428	111	152
Bicalutamide	6022	93.4	< 8.6	98.6	90.6	9.7	10.3
Clarithromycin	775	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1	< 7.1
Clindamycin	803	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7	< 7.7
Fexofenadine	3032	< 8.7	< 8.7	< 8.7	< 8.7	< 8.7	< 8.7
Propranolol	654	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1	< 8.1
Tramadol	655	< 6.5	< 6.5	< 6.5	< 6.5	< 6.5	< 6.5
Fluconazole	149	15.2	< 6.4	17.0	15.2	< 6.4	7.8
Diclofenac	67.4	50.9	12.8	42.0	19.3	84.3	115
Furosemide	303	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6	< 7.6
Irbesartan	203	< 7	< 7	< 7	< 7	< 7	< 7
Losartan	610	< 11	< 11	< 11	< 11	< 11	< 11
Sulfamethoxazole	316	< 11	< 11	< 11	< 11	< 11	< 11
Trimethoprim	11.5	< 11	< 11	< 11	< 11	< 11	< 11
Erythromycin	132	< 12	< 12	< 12	< 12	< 12	< 12
Norsertaline	< 220	< 220	< 220	< 220	< 220	< 220	< 220
Sertraline	70.8	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9	< 4.9
Venlafaxine	2212	4.3	< 4.2	5.0	< 4.2	< 4.2	4.7
Desvenlafaxine	3733	< 5.1	< 5.1	< 5.1	< 5.1	< 5.1	5.8

**Table S4.** Concentrations of transformation products in the filtered effluent and after each treatment step (Specific ozone dosage : 0.28 g O<sub>3</sub>/ g DOC)

Ozone dosage: 0.28 g O <sub>3</sub> / g DOC - Day 1							
Transformation Products	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Alpha hydroxymetoprolol	128	74.3	<6.2	65.1	64.7	<6.2	14.7
Citalopram related compound E	25.0	13.0	<5.2	19.5	25.4	<5.2	7.3
Fluconazole N-oxide	60.5	<7.6	8.9	<7.6	<7.6	13.1	34.7
4 hydroxy diclofenac	4.6	3.0	<2.6	<2.6	<2.6	<2.6	2.6

4-chloro 5-sulfamoylanthranilic acid	497	390	< 75	364	502	< 75	241
N-demethylerythromycin	< 11	< 11	< 11	< 11	< 11	< 11	< 11
Desvenlafaxine	2665	149	<5.1	142	163	<5.1	48.0
<b>Ozone dosage: 0.28 g O<sub>3</sub>/ g DOC - Day 5</b>							
	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Alpha hydroxymetoprolol	137	57.6	<6.2	56.2	60.2	6.2	17.4
Citalopram related compound E	23.1	24.5	<5.2	29.4	39.4	<5.2	11.8
Fluconazole N-oxide	126	<7.6	<7.6	9.8	19.8	31.5	<7.6
4 hydroxy diclofenac	4.6	<2.6	<2.6	<2.6	<2.6	<2.6	<2.6
4-chloro 5-sulfamoylanthranilic acid	713	329	< 75	328	430	188	377
N-demethylerythromycin	< 11	< 11	< 11	< 11	< 11	< 11	< 11
Desvenlafaxine	2945	13.2	<5.1	8.4	12.9	6.2	15.5
<b>Ozone dosage: 0.28 g O<sub>3</sub>/ g DOC - Day 12</b>							
	Filtered Effluent	Equalization Tank	GAC	AIX	Reference	Biochar 1	Biochar 2
Alpha hydroxymetoprolol	128	46.3	<6.2	41.0	46.6	10.2	21.1
Citalopram related compound E	22.9	35.1	5.2	29.5	31.0	10.7	15.3
Fluconazole N-oxide	57.1	14.9	<7.6	21.3	9.9	<7.6	<7.6
4 hydroxy diclofenac	5.5	2.8	<2.6	<2.6	<2.6	<2.6	<2.6
4-chloro 5-sulfamoylanthranilic acid	691	346	< 75	282	306	240	240
N-demethylerythromycin	< 11	< 11	< 11	< 11	< 11	< 11	< 11
Desvenlafaxine	3330	6.6	<5.1	8.1	11.2	9.7	15.1



**Figure S1.** The concentration difference between the equalization tank and the reference column as percentage in all samples (n=2-9)