



## Pilot trials with advanced water technologies to remove micropollutants from wastewater and assessment of the technologies for wastewater reuse in public blue-green solutions

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### ABSTRACT

Effluent wastewater contains organic micropollutants (OMPs) that harm aquatic ecosystems. Therefore, wastewater treatment plants (WWTPs) need to expand their facilities with advanced water treatment technologies. Water scarcity and improved effluent quality is encouraging the reuse of treated effluent. This study thus has a dual purpose: first, to evaluate OMP removal using two technique combinations – ultrafiltration and granulated activated carbon (UF:GAC), and ozone:GAC; second, to evaluate the potential of the technologies to produce water for non-potable use in public blue-green solutions, nature-based solutions for urban diversity and communities to thrive. The study assesses the effluent water quality against chemical status for reuse as surface water, bathing water quality, and irrigation quality. The results indicate that both technologies are applicable for wastewater reuse in public blue-green solutions. However, the UF:GAC combination demonstrated higher efficiency in micro-biological removal, achieving *E. coli* and Total coliforms concentrations below detection levels (<10 cfu/100 mL), compared to the ozone:GAC combination. Both technologies resulted in water quality suitable for irrigation class B, as chemical oxygen demand (COD) concentrations exceeded 10 mg/L. High OMP removal was observed with both technologies: UF:GAC showed over 70% removal at >55,000 EBV, while ozone:GAC showed more than 95% removal at 21,500 EBV.

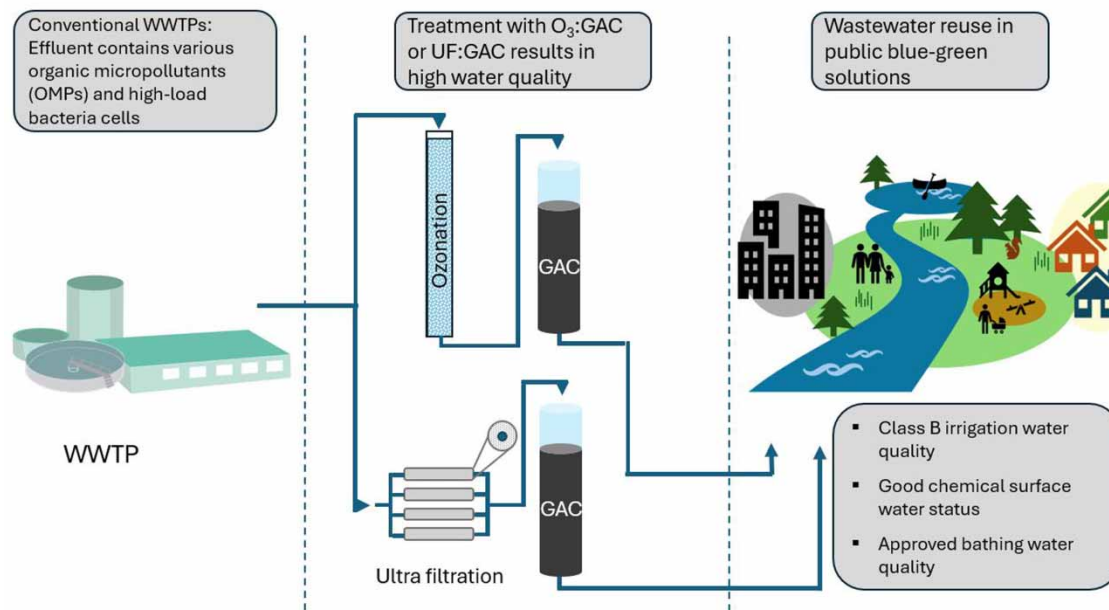
**Key words:** blue-green infrastructures, granular activated carbon (GAC), ozone, ultrafiltration (UF), wastewater treatment plants (WWTPs), water reuse

### HIGHLIGHTS

- Long-term trials for the removal of OMPs were performed over 2.5 years.
- Assessment of effluent water quality against Swedish and European directives.
- Assessment of reuse potential as surface water and bathing water.
- Assessment of reuse potential as irrigation in areas with high risk for aerosolization (*Legionella* spp.).
- This contribution will be of great interest to wastewater utilities in Europe.

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## GRAPHICAL ABSTRACT



## 1. INTRODUCTION

Climate change, population growth, and urbanisation increase the pressure on the available aquatic environment, including freshwater sources; there is thus an increased need for circular water management (Gilboa *et al.* 2023). Hence, water reuse potential has been an important field of research in recent years, aiming to reduce the use of conventional water sources and raising the issue of water quality requirements for different areas of use (Nkhoma *et al.* 2021; Takman *et al.* 2023). Water recycling takes place in several European countries and in other regions, depending on access to conventional water sources (Guo *et al.* 2022; Takman *et al.* 2023). An extensive European research project concluded that almost 70% of the European population was already experiencing water stress by 2006 (Bixio *et al.* 2006).

A complicating factor for water reuse is the fact that urban wastewater treatment plants (WWTPs) are not designed to produce water of the quality required for most water reuse applications; the effluent water contains various organic micropollutants (OMPs) and high-load bacteria cells, including pathogens and other microorganisms (Ferreiro *et al.* 2021; Lind *et al.* 2024). Advanced treatment technologies are thus necessary for increasing the quality of the produced water (Guo *et al.* 2022; Takman *et al.* 2023). To address this problem and constrain WWTPs to reduce OMPs in the treated water, the European Parliament commission proposed a revision of the Urban Wastewater Treatment Directive (UWWTD) (EU 2022). To fulfil the requirements proposed by the UWWTD, conventional WWTPs will need to extend their facilities with advanced water treatment technologies if they fall within the selection criteria.

The new requirements for quaternary treatment at WWTPs and the need for circular water management necessitate applied long-term studies, such as the one presented in this paper, to evaluate the performance of reduction of OMPs. Evaluation of current technologies is a crucial first step in meeting the demand for advanced techniques. Several technologies are of interest; one of them is to combine separation and adsorption techniques with ultrafiltration (UF) and granulated active carbon (GAC). In such a combination, UF serves as a pretreatment for subsequent GAC. As presented in Edefell *et al.* (2022), pretreatment with UF increases the efficiency and life-time of the activated carbon, since it reduces the dissolved organic carbon (DOC) content and the concentration of suspended solids (SS) (Edefell *et al.* 2022). However, previous studies have demonstrated that UF is ineffective for the removal of most MPs due to its relatively high molecular weight cut-off, while adsorption of MPs occurs primarily during filtration with GAC (Kennedy *et al.* 2015; Sheng *et al.* 2016; Mohammad-pajooh *et al.* 2018). Another promising combination is that of oxidation technique with ozone prior to adsorption with GAC (Baresel *et al.* 2015; Baresel *et al.* 2019). Ozone acts as a chemical oxidant, effective in the oxidation of OMPs, and has been demonstrated effective for inactivating higher dosages of microorganisms (Spit *et al.* 2022). The literature

shows that ozonation contributes to by- and transformation products, and it should therefore be followed by a filtration step in order to eliminate potentially toxic products from the effluent water (Jennings *et al.* 2023).

The studies mentioned above all have a limited focus, centring either on a single combination at a time or on short-term trial periods in which primarily a single filter with virgin GAC is evaluated (Luo *et al.* 2014; Jin *et al.* 2019; Fundneider *et al.* 2021; Shi *et al.* 2022; Zhang *et al.* 2023). To provide more relevant and applicable data, this study evaluates the performance of technique combinations with both virgin and reactivated GAC, based on well-founded data collected over 2.5 years.

Given the improved effluent quality provided by the above-mentioned advanced water techniques, WWTPs may benefit from considering the reuse of treated wastewater for non-potable uses (Mottaghi *et al.* 2020; Takman *et al.* 2023). A potential use of such 'technical water' could be public blue-green solutions with which to address the effects of urbanization and climate change. Public blue-green solutions are nature-based installations combining urban water (blue) systems and urban vegetated (green) areas increasing urban diversity, providing ecosystem services, mitigating climate change, and sustainable urban development (Mottaghi *et al.* 2020). Assessments performed by Takman *et al.* (2023) demonstrated that water resulting from UF with a membrane bioreactor followed by GAC is class B irrigation quality (EU 2022), but it does not fulfil drinking water quality requirements (Dias *et al.* 2023; Takman *et al.* 2023). However, previously performed assessments for irrigation purposes do not address the risk for aerosolization, which may arise in public solutions, and *Legionella spp.* is thus not addressed.

The WWTP Getteröverket in Varberg, Sweden is planning an expansion that includes opening specific parts of the facility to the public. This initiative aims to implement a blue-green infrastructure solution, featuring recreational areas with water bodies such as surface water, water fountains, and green spaces. The municipally-owned company that operates Getteröverket, Vivab, intends to utilize treated effluent water for these purposes, promoting sustainable water reuse practices. Public blue-green solutions of this kind are a valuable asset for the city's residents, offering an attractive landscape and recreation and leisure opportunities, acting as social gathering points, and attracting diverse forms of wildlife (Mottaghi *et al.* 2020, 2023; Kaur & Gupta 2022; Kabisch *et al.* 2018).

The aim of this study was thus twofold: both to evaluate the performance of two technique combinations for OMP removal at municipal WWTPs and to evaluate the potential of the technologies for producing technical water for non-potable use in public areas, such as public blue-green solutions. Specifically, the study simultaneously assesses the quality of effluent wastewater against chemical surface water status, prioritised OMPs, bathing water quality, and irrigation standards in areas with increased risk for aerosolization. The study provides groundbreaking insights into advanced treatment technologies for removing OMPs while enabling water reuse for blue-green solutions. Results support sustainable, full-scale implementation not only at Getteröverket WWTP but also at other Swedish and European wastewater utilities seeking effective and sustainable quaternary treatment solutions.

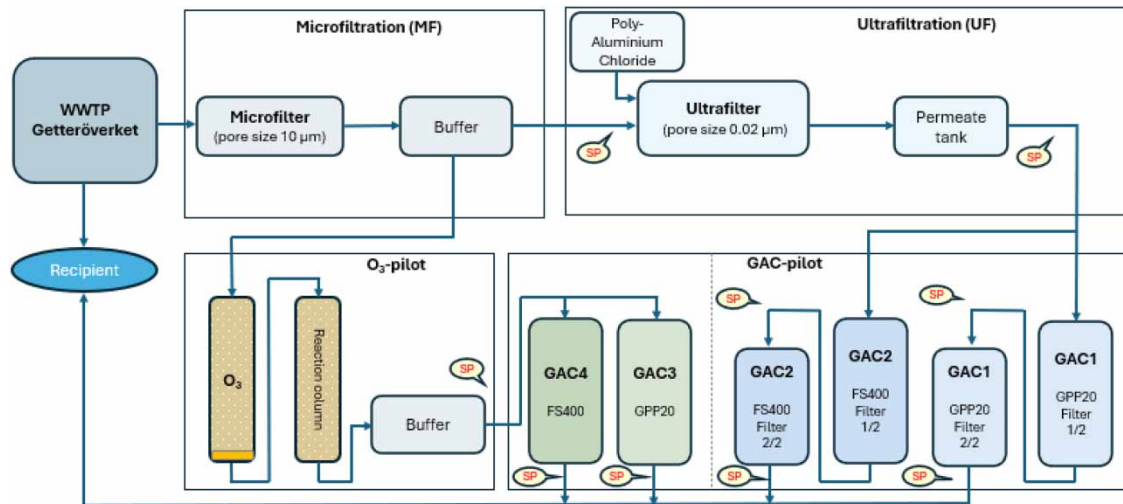
## 2. MATERIAL AND METHODS

### 2.1. Pilot trials

The pilot trials were performed at Getteröverket, a conventional WWTP located in Varberg, Sweden. Incoming wastewater to the WWTP is treated by pre-sedimentation, biological nitrogen removal, and phosphorous removal by chemical precipitation with aluminium. Effluent wastewater from the WWTP was continuously filtrated through a drum filter with a pore size of 10 µm prior to continuous treatment in two pilot plants that were installed on site, directly next to the WWTP effluent discharge. Figure 1 shows a simplified process scheme of the pilot main units, including the microfiltration step and the technique combinations used in this study, i.e., UF:GAC and O<sub>3</sub>:GAC combinations.

#### 2.1.1. UF:GAC-combination (UF:GAC-line)

Long-term continuous trials with UF:GAC combination were performed from February 2021 until the end of August 2023. Micro-filtrated (MF) wastewater was treated in a hollow fibre polymeric polyethersulfone (PES)/polyvinylpyrrolidone (PVP) 'dead end' UF module, type X-FLOW AQUAFLEX 64 with a filter area of 64 m<sup>2</sup>. The UF membrane had a nominal pore size of 0.02 µm, which implies that particles larger than 0.02 µm are efficiently removed from the wastewater. The inlet wastewater was pre-coagulated with 2.0 mg Al/m<sup>3</sup> water to enhance the removal of total organic carbon (TOC) and to reduce membrane fouling during filtration (Delgado



**Figure 1** | A simplified process scheme of the pilot plants and the sampling points.

Diaz *et al.* 2012). The filtration was performed by 40-min filter cycles with a relatively low flux (productivity) of 40 L per square metre per hour. Low productivity was chosen to avoid membrane fouling during the long-term trials and to ensure a constant supply of permeate to the subsequent GACs. Hydraulic cleaning with permeate was performed prior to each filter cycle, and chemically enhanced backwashing (CEB) with sulphuric acid, sodium hypochlorite and sodium hydroxide was executed every 25 filter cycles. During the trials, both permeability and transmembrane pressure were continuously monitored to ensure membrane recovery after CEB-procedures and accurate membrane performances. Turbidity, UVabs, colour and TOC in both inlet and permeate were monitored on-line with optical i::scan sensors.

Permeate water was treated further in two separate two-stage GAC lines in lead/lag configuration. The first GAC-line (UF:GPP-line) was filled with GPP20, a reactivated carbon suitable for non-food industrial liquid phase applications only. The second line (UF:FS-line) was filled with Filtrasorb 400 (FS400), which is a virgin carbon produced by steam activation of bituminous coal that is pulverized and then agglomerated.

Each GAC column had a diameter of 150 mm and was filled with one metre of filter material before starting, resulting in an empty bed volume (EBV) of 17.7 L. Permeate was filtered by gravity from the top of each column to continue targeting an empty bed contact time (EBCT) of 9.6 min in each column and 19.2 min EBCT for each line. EBCT is a key factor in the design of GAC filters for removing MPs from wastewater. An extensive study performed by Fundneider *et al.* 2021 on the effect of EBCT on MP removal demonstrated that further increase of EBCT to above 20–30 min does not enhance filter performance (Fundneider *et al.* 2021). Each column was equipped with a pressure sensor, and the columns were backwashed with drinking water when the pressure levels increased.

### 2.1.2. Ozone:GAC- combination (O<sub>3</sub>:GAC-line)

Continuous trials with the O<sub>3</sub>:GAC combination were performed from the beginning of November 2022 until the end of August 2023. The ozonation was carried out in an ozone pilot comprised of ozone generator type Modular 8HC from Xylem, with a nominal ozone production of 8 g/h, and two polyvinylidene difluoride (PVD) bubble columns (a mixing column and a reaction column). Each column was 4 m high with a total volume of 110 L. The ozone gas was produced by Corona discharge and was injected into the bottom of the mixing column via a diffuser, while the MF water was pumped to the top of the column. The ozone/water mix from the mixing column was pumped to the reaction column to ensure a total retention time of 20 min. An ozone dosage of 4.0 g O<sub>3</sub>/m<sup>3</sup> was chosen for the long-term trials after performing dose-response tests with increasing ozone dosages between 2 and 12 g O<sub>3</sub>/m<sup>3</sup>. During the dose-response trials, a dosage of 4.0 g O<sub>3</sub>/m<sup>3</sup> (corresponding to 0.4 g O<sub>3</sub>/g DOC) implied a 95% reduction of the tested pharmaceuticals.

After ozonation, the ozonated water was pumped to the top of the GAC columns and was filtered by gravity. Identical columns and filter materials (i.e., GPP20 and FS400) were used for the O<sub>3</sub>:GAC-line as well as for the UF:GAC-line. In contrast to the UF:GAC line, the ozonated water was treated in a single-step GAC filter.

A single-step GAC solution was chosen for the O<sub>3</sub>:GAC-line since the majority of the incoming MPs are removed in the ozonation step (Hey *et al.* 2014; Kharel *et al.* 2020). The same EBCT were applied on the individual columns in both technology combinations.

## 2.2. Evaluation of water reuse potential in blue-green solutions

The potential to reuse the effluent water as surface water in blue-green solutions was evaluated according to the limits set forth in the Swedish Water Agency's regulations on classification and environmental quality standards for surface water (HVMFS 2019:25 2019). The effluent chemical concentrations were assessed in relation to the prevailing limits for chemical surface water status in annex number 6. This annex includes all substances and the limit values for determining the chemical status of surface water (HVMFS 2019:25 2019). Other prioritised organic substances, i.e., phenols, hormones, and pharmaceuticals, were evaluated against the assessment criteria for specific pollutants in surface water in the same directive. The regulations in HVMFS 2019:25 are based on the framework for community action in the field of water policy in the European Parliament and Council Directive 2000/60/EC (EU 2000/60/EC 2000), and the Swedish water authorities must apply them for classification of surface water bodies.

The effluent quality of bathing water was evaluated against the assessment of individual tests in the Swedish Agency for Marine and Water Managements Guidelines for bathing water (Havs-och Vattenmyndigheten 2013), which is based on the EU bathing water directive 2006/7/EC (EU 2006/7/EC 2006).

A further set of European directives were used. The potential to reuse the water for irrigation purposes in blue-green solutions was also evaluated against Regulation (EU) 2020/741 of the European Parliament and of the Council of 25 May 2020 on minimum requirements for water reuse (EU 2020/741 2020). Additionally, other indicator organisms such as *Legionella spp.*, intestinal senterococci, and total coliforms (TC) were assessed to ensure microbial safety. Since minimum requirements for biological oxygen demand (BOD) and total suspended solids (TSS) in regulation 2020/741 refer to the EU Council directive 91/271/EEC concerning urban wastewater treatment, Annex 1, Table 1 and Section D, these standard parameters were evaluated according to the requirements in this directive (EU 91/271/EEC 1991; EU 2020/741 2020).

## 2.3. Sampling and analyses

Sampling points were positioned before and after each pilot step to allow evaluation of different configurations (Figure 1). An overview of sampling frequency, sampling type, and analytical laboratory can be observed in Table S2. During the pilot trials, sampling of pharmaceuticals, hormones, PFAS 11, total cell concentrations (TCC), and intact cell concentrations (ICC) were regularly performed, as were increased empty bed volumes (EBVs). Sampling of pharmaceuticals, hormones, and PFAS 11 was carried out as weekly composite samples. During each sampling week, 50 mL water samples were collected from each sampling point every 30 min; they were stored at 4 °C throughout the entire sampling period until they were analysed at IVL Swedish Environmental Research Institute laboratory according to the following methods.

Pharmaceutical and antibiotic residues in wastewater were analysed by LC–mass spectrometry (MS)/MS according to a method described by Gros *et al.* (2006), with two deviations (Gros *et al.* 2006). The first method deviation is the addition of 200 mg of ethylenediaminetetraacetic acid to the water sample to break any interactions between the analytes and metal ions present in the sample. The second deviation entails that the elution of the substances from the extraction columns has been carried out with methanol and acetone. The separation column used is a Kinetix™ biphenyl core–shell column with the dimensions 100 mm × 3 mm with 2.6 μm particle size (Phenomenex), and the elution buffers used in ionization in positive electrospray ionization (ESI) are 0.1% formic acid in water and 0.1% formic acid in methanol. Water samples were extracted using solid phase columns (Oasis HLB, Waters). 13C15N-carbamazepine, 13C6-diclofenac, 13C6-hydrochlorothiazide, d7-atenolol, d7-metoprolol, and d3-ibuprofen were used as internal standards for quantification.

Water samples were extracted using solid-phase columns (ENV+ and primary secondary amine (PSA), Biotage to determine the levels of oestrone (E1), oestradiol (E2), and ethinylestradiol (EE2). Analysis was performed with high-performance liquid chromatography (HPLC) – high-resolution mass spectrometry (HRMS) (Orbitrap). 13C2-E2 and 13C2-EE2 were used as internal standards for quantification.

For analysis of PFAS, the water samples were extracted with SPE columns (Oasis WAX, Waters) and analysed with HPLC-MS/MS. 18O2-PFHxS, 13C4-PFOS, 13C4-PFBA, 13C2-PFHxA, 13C4-PFOA, 13C5-PFNA,

**Table 1** | Microorganisms and standard parameters results, in and out from both technique combinations

	Effluent WWTP In Pilots	Average effluent concentrations				Bathing water quality, according to guidelines for individual tests (Havs- och Vattenmyndigheten 2013)			Limits for qualification for irrigation standards (EU 91/271/EEC 1991)			
		UF: FS	UF: GPP	O <sub>3</sub> FS	O <sub>3</sub> GPP	AQ	AQR	NAQ	Class A	Class B	Class C	Class D
Intestinal enterococci (cfu/100 mL)	1,743 ± 2,042	<10	<10	<10	<10	≤100	>100–300	>300	↵	↵	↵	↵
<i>E. coli</i> (cfu/100 mL)	4,600 ± 954	<10	<10	12.3 ± 12.7	<10	≤100	>100–1,000	>1,000	≤ 10	≤ 100	≤ 1,000	≤ 10 000
TCs (cfu/100 mL)	13,000 ± 5,196	<10	<10	1,926 ± 3,269	622.7 ± 1,020	↵	↵	↵	↵	↵	↵	↵
<i>Legionella spp.</i> (cfu/100 mL)	<1,000	<10	<10	<1,000	<1,000	↵	↵	↵	<1,000	<1,000	<1,000	<1,000
BOD <sub>7</sub> (mg/L)	3.60 ± 2.30	0.41 ± 0.30	0.42 ± 0.31	1.50 ± 0.71	1.65 ± 0.92	↵	↵	↵	≤10	25	25	25
COD (mg/L)	31.3 ± 3.1	14.0 ± 0.71	14.2 ± 2.55	16.6 ± 1.27	17.6 ± 1.56	↵	↵	↵	≤10	125	125	125
TSS (mg/L)	5.58 ± 2.64	0.25 ± 0.07	0.15 ± 0.07	0.40 ± 0.14	0.60 ± 0.42	↵	↵	↵	≤10	35	35	35
Turbidity (NTU)	0.72 ± 0.11	0.16 ± 0,02	0.17 ± 0.03	0.36 ± 0.04	0.36 ± 0.04	↵	↵	↵	≤5	↵	↵	↵

Note. Guidelines for bathing water quality and qualification of irrigation standards.

AQ, approvable quality; AQR, approvable quality with remarks; NAQ, non-approvable quality.

13C2-PFDA, 13C2-PFUnDA, 13C2-PFDoDA were used as internal standards for quantification. The amount of PFOS was given as the sum of linear- and branched PFOS.

Samples for flow cytometry (FCM) were collected in sterile 15 mL Falcon tubes, stored at 4 °C and analysed the same day they were collected. FCM analyses were performed according to the protocol described in *Prest et al. (2013)* on a BD Accuri C6 Plus (BD Biosciences, Belgium) (*Prest et al. 2013*). This flow cytometer is equipped with a 50 mW laser, emitting at a wavelength of 488 nm. Green and red fluorescence was read at FL1 (533 ± 30 nm) and FL3 (>670 nm), respectively. TCC were determined by staining samples with 5 µL of SYBR Green I at 100× diluted with dimethyl sulphoxide (final volume: 500 µL; final SYBR Green I concentration: 1×). After staining, samples were incubated for 15 min in the dark at 37 °C. For measurements of ICC, propidium iodide was included in the staining protocol (0.3 mM final concentration, 500 µL final volume).

*Escherichia coli*, TC, *Legionella spp.*, and intestinal enterococci sampling was carried out during summertime (July and August 2023) since assessment with FCM displayed an increase in viable cell counts during warmer periods, and due to increased water temperature, which facilitates bacteria growth. The samples were analysed at an Société Générale de Surveillance (SGS)-accredited laboratory according to the following methods: *Legionella spp.* (ISO 11731, 11731-2), *E. coli* (SS028167-2 MF), intestinal enterococci (SS-EN ISO 7899-2), and TC (SS028167-2 MF). Standard parameters were analysed as follows: BOD (SS-EN ISO 5815-1:2019), TSS (SS-EN 872:2005), and chemical oxygen demand (COD) (LCK385).

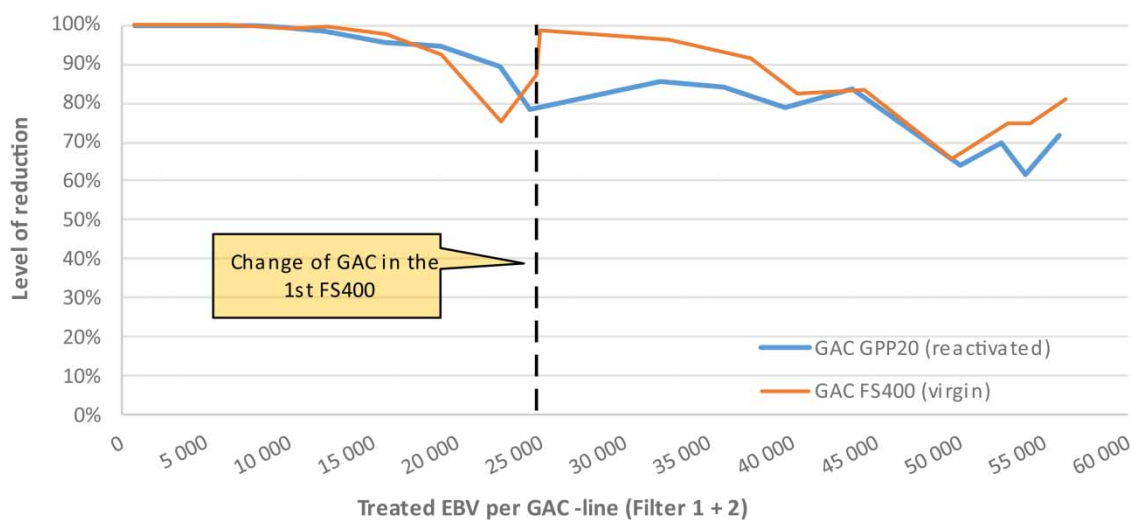
Sampling of substances was carried out for evaluation of the effluent water chemical status simultaneously with the microbial sampling. The samples were analysed at an SGS laboratory according to the methods described in Table S1 in the supplementary information.

### 3. RESULTS AND DISCUSSION

#### 3.1. Pilot trials

##### 3.1.1. Technique combination UF and 2-stage GAC (UF:GAC)

Figure 2 shows the removal efficiency for the sum of all analysed pharmaceuticals over the two GAC lines. The sum of all analysed substances should only be used as an indication, however, and it refers mainly to the 16 most persistent substances that are not removed during the main treatment process of the WWTP. As expected, the UF did not impact the observed concentrations, and results from the UF step are therefore excluded from the figure. The figure shows that both GPP20 and FS400 had approximately the same removal efficiency and a remaining capacity of about >70% removal at >55,000 EBV for the entire filter line. The performance of the two GAC-types was similar for some substances (e.g. citalopram and furosemide) but differed for other substances, such as, e.g. diclofenac, where GPP20 had a much higher overall removal efficiency than FS400, and vice versa regarding



**Figure 2** | Removal efficiency for the sum of all analysed pharmaceuticals removal over the two GAC lines. The bed volumes on the x-axis indicate the total bed volumes in each line, i.e. filter 1 plus filter 2. The orange line represents the reduction over FS400, and the blue line represents the reduction over GPP20. The dashed vertical line illustrates the point when the FS400 was changed in the first filter.

oxazepam (individual substances are shown in the supplementary information, Figure S3). These findings are in line with other studies such as by Benstoem *et al.* (2017) that showed no difference in adsorption performance between reactivated and virgin GAC. However, an earlier study which focused on the changes in pore structures of virgin and reactivated GAC have demonstrated that the pore size distribution of the two GACs differed significantly (Moore *et al.* 2001). These differences might explain the differences in the performance of the GACs for some substances such as diclofenac in this study.

The gradual decline in OMP removal efficiency in the GAC filter is attributed to the decreasing adsorption capacity over time, as more adsorption sites become occupied (Edefell *et al.* 2022). To maintain an average removal efficiency of 80%, the adsorbent FS400 was replaced in the first column after 25,000 EBV, and the filter-operation was adapted so that the fresh filter was placed second in the filter line. Replacing the adsorbent resulted in a clear increase of removal efficiency of that filter line (Figure 2). However, the higher efficiency declined faster than for the pilot line with GPP20 and already reached the same level after approximately an additional 15,000 treated EBV for the entire filter line. These results show that although the removal efficiency in both lines was below the targeted 80% reduction, the average removal efficiency over the entire operating period was significantly higher than 80% for both GPP20 and FS400 at the end of the trials. The average reduction is more relevant for a full-scale implementation, as multiple filter lines will be operated, and they will be at different stages of their lifetime. This means that one filter line with a removal efficiency below 80% will be compensated by another filter line with an efficiency >80%.

The project also investigated the removal efficiency for the indicator substances in the revised sewage directive (EU 2022) and it could be observed that the average removal efficiency for all 12 substances at the end of the trials was 60% for GPP20 and 70% for FS400, respectively. However, considering four substances from category 1 and two from category 2 as required by the directive, the observed removal efficiencies at the end of the trials were 76 and 85% for GPP20 and FS400, respectively.

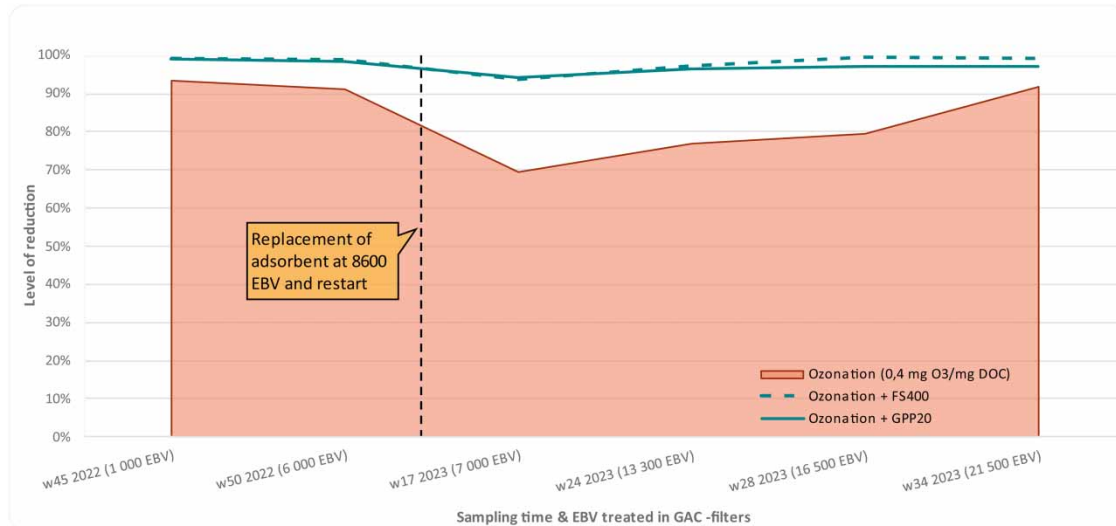
In addition to the removal of pharmaceuticals, the treatment system was also investigated regarding the removal of various PFAS. It was observed that PFOS is removed to a greater extent than PFOA and  $\Sigma$ PFAS11. (The concentrations of PFAS substances are shown in the supplementary information, Figure S4.) Furthermore, a generally better removal effect was achieved for FS400 than for GPP20 – at least up to 20,000 EBV. A negative reduction for various PFAS over the GAC-pilot lines could be observed after 20,000 EBV for FS400 and 40,000 EBV for GPP20, respectively. The negative reduction can be explained by the desorption (release) of the previously adsorbed PFAS substances in the filter material. This effect is important for the future implementation and operation of GAC filters, as the filter material must be replaced much more frequently to achieve high PFAS reduction, depending on, e.g., different reuse applications.

### 3.1.2. Technique combination of ozone and GAC (ozone:GAC)

Prior to the continuous trials, dose-response tests with five different ozone doses in the range of 0.28–1.7 g O<sub>3</sub>/g DOC were performed to determine the appropriate ozone dosage. These initial dose-response tests indicated expected outcomes compared to previous trials in the literature (Baresel *et al.* 2015; Baresel *et al.* 2017). While, e.g., furosemide was already removed efficiently at the lowest ozone dose, a higher ozone dose (0.86 mg O<sub>3</sub>/mg DOC) was required to reduce oxazepam by at least 80%, which aligned with expectations. All investigated pharmaceuticals were removed by >98% at an ozone dose of 1.3 mg O<sub>3</sub>/mg DOC. However, ozone dosages >1.3 mg O<sub>3</sub>/mg DOC also resulted in bromate concentrations after ozonation >3 µg/L, which indicated that there is a clear risk of bromate formation that may exceed recommended levels. Generally, if the bromide concentration in the water to be treated exceeds 0.4 mg/L, ozonation should not be implemented due to the risk of bromide being oxidized to bromate (Kompetenzzentrum *et al.* 2017). Bromate is toxic and has a regulatory limit of 10 µg/L in Swedish drinking water regulations (SLVFS 2001:30). However, this limit applies specifically to drinking water and not to treated wastewater. A discharge limit of 50 µg/L for treated effluent has been proposed based on observed negative effects of bromate (Soltermann *et al.* 2016).

The long-term trial with the two O<sub>3</sub>:GAC-lines – one with GPP20 and one with FS400 as adsorbent – faced operational problems in the preceding treatment steps, and the filter material in both GAC filters thus had to be replaced approximately 12 operating weeks. Despite the forced restart of the GAC-filters, 21 500 EBV could be treated within the following 8 months of the trials due to the low EBCT of 10 min.

Figure 3 shows the removal during ozonation and the removal during the combined ozonation and GAC filter (GPP20 and FS400) for all analysed pharmaceuticals. As illustrated, not all substances were removed by



**Figure 3** | Removal of the sum of the pharmaceuticals over the ozonation and the combined ozonation and GAC filters (GPP20 and FS400). The orange area represents the reduction of pharmaceuticals during ozonation, while the solid green line and the dashed green line represent the removal of pharmaceuticals after treatment with both ozonation and GAC (ozonation plus GPP20, and ozonation plus FS400, respectively). The vertical dashed line illustrates the time of replacement of the adsorbents.

ozonation at the applied dose of  $4 \text{ g O}_3/\text{m}^3$ , and the removal efficiency varied over the test period. The variations may be explained both by a varying DOC concentration in the treated water, which by an ozone dose control related to a water flow implies DOC-related dose variation and by variations in the incoming concentrations of pharmaceuticals in the inlet water, which affects the removal rates of these substances. However, as shown in Figure 3, the variations after the ozonation were compensated by the subsequent GAC filters, and the pharmaceuticals were efficiently removed. This is especially clear for substances that are less efficiently removed by ozone oxidation, such as oxazepam, as also other studies have shown. Fakioglu *et al.* (2024) indicated, e.g., that only 36% of oxazepam was removed after ozonation with ozone dosage of  $0.28 \text{ g O}_3/\text{g DOC}$ , and (Baresel *et al.* 2016) identified oxazepam as the substance that required highest ozone dose of all investigated pharmaceuticals to achieve similar reduction. The total reduction for the sum of all analysed substances and the specific removal for each substance was over 80%. For most substances, however, it was  $>97\%$ , even after 21,500 EBV. This was also true for investigated indicator substances in the revised sewage directive (EU 2022). The substances were reduced at the end of the pilot trials with  $>95\%$  in both  $\text{O}_3$ :GAC pilot lines (individual substances are detailed in the supplementary information, Figure S1).

Variations in concentrations between the different sampling dates could also be observed for various PFAS. PFAS removal could be observed over the different purification steps, especially GAC, but with decreasing removal efficiency with increasing treated EBVs (the concentration of PFAS substances is shown in the supplementary information, Figure S2). A PFAS reduction in the ozonation step was observed but not expected, as PFAS lacks electron-dense sites in the molecular structure that can be attacked by ozone (Vecitis *et al.* 2009; Sonntag & Gunten 2012; Yang *et al.* 2014). It is possible that something in the treated water catalyses oxidation with ozone. Ozone can react with PFAS in the presence of a catalyst, e.g. iron, and in this heterogeneous oxidation process, attacks occur on the PFAS structure, leading to reduction of the substance (Franke *et al.* 2019). Another possible explanation may be that PFAS precursors are converted into stable and analysed PFAS. This observation was made in another study; in that study, however, the ozone was in the presence of iron oxide (catalyst) and persulfate. No oxidation occurred in the experiments with only ozone (Franke *et al.* 2019).

### 3.1.3. Flow cytometry

**3.1.3.1. Technique combination UF and GAC.** FCM results showed that the UF plant achieved a log reduction of TCC of about  $4 \pm 0.2$  at the beginning of the trial period. However, bacterial growth was noted in the sampling line for the permeate, leading to an increase of TCC from about  $800 \pm 391 \text{ cells/mL}$  in spring 2021 to  $3,400 \pm 920 \text{ cells/mL}$  in spring 2023, resulting in a decrease of log reduction to  $3.3 \pm 0.2$  towards the end of the trial period.

The examination of the GAC process showed similar FCM results for the two different lines with an increase in ICC in the summer months (Figure S5). The increase in viable cells can be attributed to several factors. Elevated water temperatures during the summer enhance bacterial activity within the biofilm of the GAC columns, thereby increasing the ICC. Furthermore, higher concentrations of organic matter and nutrients, often resulting from increased anthropogenic activities, provide favourable conditions for bacterial proliferation, contributing to a higher abundance of ICC. The data showed a moderate correlation between water temperature and TCC ( $R^2 = 0.47$ ,  $p < 0.05$ ). However, there was no apparent correlation between TCC and EBV ( $R^2 = 0.0035$ ,  $p = 0.17$ ) (Figure S6). The correlations between increased water temperature and increase in TCC are in line with results presented earlier by [Takman et al. 2023](#)).

**3.1.3.2. Technique combination of ozone and GAC.** Most bacteria were killed during the ozonation step in the pilot. On average, the proportion of intact cells in the water after ozonation was  $5 \pm 1\%$ . The percentage of ICC increased to around  $47\% \pm 15\%$  after the GAC, which indicates microbiological growth in the GAC columns. Both GACs follow the same trend with an increase in the number of bacteria during the summer.

### 3.2. Evaluation of water reuse potential

The effluent analysis results for microorganisms and standard parameters (BOD, COD, TSS, and turbidity), the requirements for bathing water quality, and the limit concentration for different irrigation standards are summarised in [Table 1](#). As observed in the table, substantial removal of *E. coli*, TC, *Legionella spp.*, and intestinal enterococci was achieved with both advanced water technique combinations. However, more significant bacterial removal was observed with UF:GAC technology than with Ozon:GAC technology. Regardless of filter material, the effluent concentrations of intestinal enterococci and *E. coli* from both technique combinations were below the limit of detection (LOD) ( $<10$  cfu/100 mL), except that effluent concentrations of *E. coli* from O<sub>3</sub>:FS. *E. coli* from O<sub>3</sub>:FS had an average of 12.3 cfu/100 mL with a maximum concentration of 27.0 cfu/100 mL on one sampling occasion (not shown in the table). Thus, the effluent concentrations from all the tested GACs (both technologies) were below the approvable thresholds for bathing water individual tests ([Havs-och Vattenmyndigheten 2013](#)). The concentrations are also at least tenfold below the limits for excellent water quality based on the EU bathing water directive 2006/7EC ([EU 2006/7/EC 2006](#)). For coastal waters, the limits for excellent water quality are 100 cfu/100 mL for intestinal enterococci and 250 cfu/100 mL for *E. coli*. For inland waters, the limits are 200 cfu/100 mL for intestinal enterococci and 500 cfu/100 mL for *E. coli*. It should be observed, however, that the EU limits are based on a 95-percentile evaluation of the recent four bathing seasons, whereas the tests in this study only involved one season ([EU 2006/7/EC 2006](#)).

The effluent *E. coli* concentrations met the criteria for irrigation as class A quality water for technique combination UF:GAC with both tested GACs (i.e., UF:GPP and UF:FS), while the effluent *E. coli* concentrations from O<sub>3</sub>:GAC combination were compatible with class A with O<sub>3</sub>:GPP and class B with O<sub>3</sub>:FS. *Legionella spp.* concentrations from all the tested combinations met the requirements for irrigation in areas where there is a risk of aerosolization according to the EU reclaimed water quality requirements for all irrigation classes, albeit only reaching irrigation class B criteria due to high COD concentrations ([Table 1](#)) ([EU 2020/741 2020](#)).

[Table 1](#) also includes data for TC, which is used worldwide as an indicator organism to ensure microbial safety. Its abundance in wastewater is well known and has been reported in several studies ([Guo et al. 2009](#); [Shi et al. 2022](#); [Chen et al. 2023](#)). As shown in [Table 1](#), high TC was observed in the effluent water from O<sub>3</sub>:GPP ( $622.7 \pm 1,020$  cfu/100 mL) and O<sub>3</sub>:FS ( $1,926 \pm 3,269$  cfu/100 mL), while the effluent water from the UF:GAC technique combination indicated concentrations below LOD ( $<10$  cfu/100 mL) for both filter materials. An explanation for this difference may be that UF with direct coagulation has a high microbiologic barrier effect, assuring extremely high bacterial log reduction, as observed in the FCM results. The FCM results also indicate an increase in TCC over the GACs during warmer periods (Figure S6). The increase of TCC did not correlate with an increase in TC since its current was below LOD on all sampling occasions. Similarly, an increase of TCC during warmer periods did not correlate with an increase of *E. coli*, intestinal enterococci, or *Legionella spp.*

The limited TC reduction in the O<sub>3</sub>-GAC pilot could possibly be explained by the specific experimental setup. The ozone dosage in the experiment was mainly optimized for pharmaceutical removal. Higher ozonation dosages are necessary to increase TC reduction. Studies performed earlier by [Wu et al. \(2018\)](#) illustrated that low O<sub>3</sub> doses (O<sub>3</sub>/DOC  $< 0.25$  g/g) damage the deoxyribonucleic acid (DNA) of *E. coli*. DNA damage rate significantly decreases with increased bacteria concentrations, however, and it is affected by other factors such as

DOC and SSs (Wu *et al.* 2018; Shi *et al.* 2022). Therefore, further site-specific studies should be performed to evaluate the optimal ozone dosage effects on TC removal. Another factor that may increase TC reduction is an increase of EBCT. Previous studies have shown that an increase of EBCT up to 20 min may increase the efficiency of GAC (Fundneider *et al.* 2021). Nonetheless, specific studies are required to evaluate the impact of EBCT on TC concentrations, since an increased EBCT might also promote TC growth due to an increase in oxygen concentrations.

A third factor that would potentially impact TC reduction is ultraviolet light (UV), included as an additional step in the technology combination. The dosage should be carefully determined however in order to balance the efficiency of disinfection with photoreactivation risks and economy due to increased energy requirements (Guo *et al.* 2009). A study performed by Guo *et al.* (2009) on three types of wastewaters demonstrated that an UV-dose of 40 mJ/cm<sup>2</sup> reduced the TC concentrations to below detection limits of 0.1 CFU/mL (Guo *et al.* 2009).

Chemical surface water status was also assessed. The overall results indicate that good chemical surface water status can be reached. The effluent concentrations of 43 of the 49 tested substances were below the limit values for evaluation of chemical surface water status in Sweden according to HVMFS 2019:25 (both inland water and other surface bodies) (Table S1). Unfortunately, the LOD of the remaining six tested substances was above the limit values and could therefore not be fully evaluated.

Table 2 displays the average and maximum concentrations of prioritised substances and their limit concentrations according to the requirements in HVMFS 2019:25 for inland- and coastal water. It can be concluded that the pilots played a crucial role in meeting the limits of hormones. The average concentrations of hormones (E2 and EE2) in the effluent of the WWTP were above the required average concentrations for coastal water, while the average concentrations of E2 met the limits and the average concentration of EE2 exceeded the average limit for inland water. The hormones were completely removed with both technique combinations, regardless of filter material. This correlates well with previous studies, which have shown that hormones are removed easily by GAC and easily oxidized with low ozone doses (Sonntag & Gunten 2012).

**Table 2** | Prioritised organic substances (hormones, pharmaceuticals, and phenols) and their limit values according to requirements in HVMFS 2019:25 for inland- and coastal waste

	Effluent concentrations (max) (ng/L)					Surface water limits prioritised substances (ng/L)			
	Effluent WWTP	UF FS	UF GPP	O <sub>3</sub> FS	O <sub>3</sub> GPP	Inland water		Coastal water	
	Average concentrations (max) (ng/L).					Maximum conc.	Average conc.	Maximum conc.	Average conc.
Oestradiol (17beta-) E2	0.4 (1.0)	nd	nd	nd	nd	–	0.4	–	0.08
17 $\alpha$ -ethinyl oestradiol (EE2)	0.4 (1.0)	nd	nd	nd	nd	–	0.035	–	0.007
Ciprofloxacin	nd	nd	nd	nd	nd	100	–	100	–
Diclofenac	950 (1,700)	18.0 (270)	15.0 (220)	nd	nd	–	100	–	100
Bisphenol A	84.8 (170)	56.7 (140)	nd	nd	nd	1,600	2,700	–	110
Nonylphenol	48 (140)	46.7 (140)	16.0 (48.0)	nd	nd	300	–	–	300

Note. Maximum conc. refers to the maximum acceptable concentrations, measured on a single measurement occasion. Average conc. requirements are expressed as an annual average concentration.

In contrast to hormones, for which there are no explicit average concentration limits, the maximum concentrations of the analysed phenols in the effluent WWTP were far below the target limits for inland and coastal waters in HVMFS 2019:25, which means that the limited reduction observed in the GACs (except for the reduction in UF:FS) is not critical for meeting the targets. Nevertheless, minimising phenol concentrations is of great importance due to their negative toxicological effects. This is especially relevant due to the global increase in phenol production (Shin *et al.* 2023; Zaborowska *et al.* 2023). Zaborowska *et al.* (2023) predicts a 6% annual increase in Bisphenol A production between 2022 and 2027.

As expected, the concentrations of the pharmaceuticals (ciprofloxacin and diclofenac) were below the limits for inland- and coastal surface water due to high performance in pharmaceutical reduction with both technique combinations (Table 2). It should be noted that for Ciprofloxacin, only maximum concentrations are required at HVMFS 2019:25. Nonetheless, ciprofloxacin was already removed in the WWTP and was not detected in the

incoming water to the pilots (effluent WWTP), and the pilots thus made no contribution to removing this substance. In contrast to Ciprofloxacin, the pilots were crucial for the removal of diclofenac.

In this study, the reduction of diclofenac during ozonation was above 96% during all the trials; this is consistent with the results obtained in the literature (Baresel *et al.* 2015). The average concentrations of diclofenac in the WWTP effluent during the trial period were almost tenfold higher than the average limit in HVMFS 2019:25, while the concentrations of diclofenac after treatment with the pilots were far below the required concentrations. Nevertheless, the maximum concentrations in the UF:GAC-line were more than double the average required concentrations in HVMFS 2019:25. This may be explained by the saturation of the GAC due to the increase of EBVs. The results from this study thus indicate that the filter material should be changed after no more than 50,000 EBVs to ensure sufficient reduction of diclofenac and to avoid exceeding targets in HVMFS 2019:25. This holds particularly true for Getteröverket WWTP since there is no anticipated dilution from other water bodies. If dilution is considered, however, such frequent replacement of GAC may not be necessary. Unlike the UF:GAC-line, where the main reduction of diclofenac occurs in the GAC, the main reduction of pharmaceuticals in the O<sub>3</sub>:GAC-line occurs during ozonation, and high and stable reduction is thus achieved continuously.

#### 4. CONCLUSIONS

This study evaluated two different technique combinations, i.e. UF:GAC and ozone:GAC, for the removal of micropollutants, and the effluent wastewater was assessed to indicate its potential for non-potable use in public blue-green solutions. Based on the results presented, the following specific conclusions can be drawn:

- The effluent water quality from both technologies was within the limit values for good chemical surface water status.
- Approved bathing water quality was achieved with both of the tested technique combinations according to the prevailing limits for individual tests. However, complementary studies are required to ensure compliance over four consecutive seasons, as stipulated by the EU bathing water directive 2006/7EC.
- Class B irrigation quality water was achieved with both technique combinations due to high COD concentrations. Nevertheless, both technique combinations fulfilled the *Legionella spp.* requirements for irrigation in areas with a high risk of aerosolization. Therefore, the water can replace potable water for irrigation in green areas.

Overall it can be concluded that both technique combinations are applicable for wastewater reuse in public blue-green solutions as intended at Getteröverket WWTP. While higher efficiency in microbial removal was observed with the UF:GAC combination, an extension of the O<sub>3</sub>:GAC combination with UV to reduce the microorganisms to similar levels is necessary. Further extension of the UF:GAC combination with UV as an additional microbiological barrier to UF, should also be considered, depending on the reuse application, to increase safety in case of unexpected trouble shootings in the UF-step and seasonal variations. Both combinations were easily managed and adaptable and will meet the requirements for pharmaceutical reduction in the revised sewage directive. Increased pharmaceutical reduction can be achieved either by increasing the ozone dosage or by more frequent replacement of the GAC at lower EBVs.

The study is an important step towards increased water reclamation. However, future studies are required to evaluate other chemical substances that were not evaluated in this study, and other possible applications of effluent water should be investigated. Further discussions and permits are required by the relevant authorities before reuse in such solutions. Public acceptance, given the water's origin, is another key consideration, as this aspect was not covered by this study and needs to be explored further.

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#### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

## CONFLICT OF INTEREST

The authors declare there is no conflict.

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