Reuse of municipal wastewater for different purposes based on a modular treatment concept

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ABSTRACT

Due to water scarcity and water pollution, the importance of water reuse is increasing more and more. As part of a German research programme on water reuse, the effluent of a wastewater treatment plant in the coastal region of northern Germany was used to investigate within the project MULTI-ReUse the direct treatment of tertiary effluent for usage in different applications in industry or agriculture. A modular constructed pilot system has been operated to optimize different treatment chains producing different water qualities simultaneously. The technological focus was put on membrane technologies, namely ultrafiltration (UF) and reverse osmosis (RO), and also biofiltration, adsorption and disinfection were part of the piloting. Beside the development of monitoring strategies for ensuring biological and chemical safe water qualities, the operational stability and the safe transport of water to the consumers were examined. The direct treatment of wastewater is a demanding task due to the lack of dilution and hydraulic retention time in the receiving water (environmental buffer). However, the multiple barrier approach guaranteed constant secure water. Fine adjustments of individual processes were particularly important. A stable operation of the UF could be realized in particular by using more or less intermittent inline coagulation as coating. The RO performance could be improved significantly by using monochloramine as disinfectant to minimize biofouling.

Key words | adsorption, organic micropollutants, pathogen removal, reverse osmosis, ultrafiltration, water reuse

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HIGHLIGHTS

- The first project in Germany on direct reuse of municipal wastewater for industrial purposes.
- Optimization of UF operation by initial hydroxide coating followed by low levels of coagulant dosing.
- Implementation of an ultra-low-pressure reverse osmosis membrane (ULPRO).

INTRODUCTION

Water reuse can be of interest for various reasons. On the one hand, more frequent and severe drought conditions due to climate change (Solomon *et al.* 2007), excessive water consumption and competing environmental, industrial and agricultural needs for water can stress the availability of conventional freshwater resources (Drewes & Horstmeyer 2015). On the other hand, the low quality of regionally available fresh water resources gives reason to

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focus on the use of alternative resources. In this context, collected wastewater has been explored as an additional water resource since decades (Asano *et al.* 2007). Up to date, numerous wastewater reclamation facilities have been successfully implemented for different water reuse purposes (Curl *et al.* 2019). Even the direct potable reuse is becoming more and more accepted as state-of-the-art technologies have proved to provide the desired water quality with high reliability.

Despite the successes that water reclamation projects have achieved internationally, in Germany this topic is still being discussed very critically and defensively by many public actors (UBA 2017). However, according to climate change predictions, more warm and dry summers are to be expected in central Europe. The effect on the groundwater level and surface water reservoirs could be noticed in several areas in Germany during the years 2018 and 2019 (Hellwig *et al.* 2020). The expected simultaneous rise in sea levels also leads to a disturbance in the balance between groundwater and the seawater interface, which results in increasing salinization of the groundwater body. This makes the North Sea coast of Europe a good example for the problem of saltwater intrusion (Auken *et al.* 2012).

Against this background, a regional water board in Lower Saxony, Germany, decided to explore the possibilities of municipal wastewater reuse for industrial purposes in a local case study. Nordenham is a mid-sized German town with water-intensive industries and trades. It is located in the Wesermarsch, a coastal region in the northwest of Germany, which is affected by saltwater intrusion. The entire district does not have its own drinking water supply. In the surrounding districts, drinking water obtained from groundwater resources is treated and pumped over long distances to the industrialized area. Here, the water demand for industrial processes with very low water quality requirements is covered as much as possible with water from the high salinated river Weser. However, for the majority of applications, drinking water is used (Kroemer *et al.* 2019). To lower or to completely avoid the demand for drinking water in cooperating industrial companies could lead to a significant relief of the used drinking water aquifers. This case study was a main part of the MULTI-ReUse project of a German multicentre research consortium. Within the project, a treatment system for water reuse has been tested at the site of the municipal wastewater treatment plant (WWTP) of Nordenham with the aim of producing process water in three different qualities.

The technological studies focused on the membrane processes ultrafiltration (UF) and reverse osmosis (RO) supplemented by further treatment steps. Depending on the intended use, the processes can be combined into different treatment chains (Fit-for-Purpose) (EPA/600/R-12/618 2012). By choosing these state-of-the-art technologies, focus was on the demonstration of technologies under site-specific conditions. Additionally, the development of new RO membranes and microbial monitoring methods was supported. This article describes the set-up and the main results of the pilot plant operation, beside the aspects of waste disposal and water transport are discussed for the specific case study.

MATERIALS AND METHODS

Pilot plant

The MULTI-ReUse pilot plant consists of modular units for three treatment lines (Figure 1):

1. *Line 1 for ReUse Water 1*: pre-filtration (sieve), optional adsorption with powdered activated carbon (PAC), flocculation, UF and UV disinfection; water for rinsing and cooling purposes.
2. *Line 2 for ReUse Water 2*: pre-filtration, flocculation, UF, UV disinfection, sand filtration and granulated activated carbon (GAC) filtration; water for irrigation purposes.
3. *Line 3 for ReUse Water 3*: pre-filtration, optional PAC and flocculation, UF, chemical disinfection, RO and UV; water for industrial purposes.

Two UF lines were operated in parallel. The treatment lines were protected by a sieve filter (AMIAD TAF-750) with a mesh size of 200 μm. The sieve filter was rinsed automatically. Coagulation was used prior to UF to reduce fouling by decreasing the cake resistance, limiting pore blockage and increasing the backwash efficiency (Barbot *et al.* 2008). Coagulation also increases the removal of organic matter, phosphate and disinfection by-product (DBP) precursor (Guigui *et al.* 2002). Aluminium

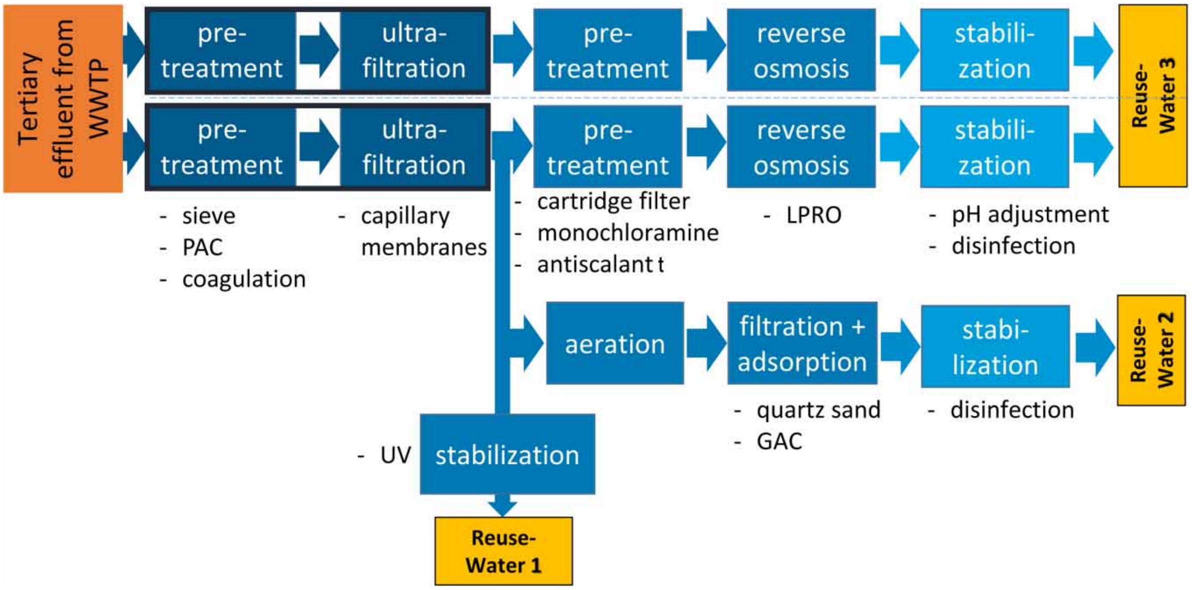

Figure 1 | Flow chart of the pilot plant.

hydroxychloride (feralco PLUSPAC FD ACH)- and ferric-based coagulants ($FeClSO_4$ or $FeCl_3$, CG CHEMIKALIEN) were used with a dosage of $[Al^{3+}] = 2–3$ mg/L and $[Fe^{3+}] = 6–7$ mg/L. The average hydraulic retention time for micro-floc formation was optimized to 40 s.

Hard coal-based PAC (Chemviron PULSORB® WP260-90 S) with a mean size of 0.3 μm (number concentration, Beckman Coulter LS 13,320) was used for the removal of organic micropollutants and DBP precursor (Lin *et al.* 1999; Ivancev-Tumbas *et al.* 2008; Stoquart *et al.* 2012).

After pre-treatment, the water was filtrated with two large-scale UF modules (dizzer XL 0.9 MB 80 WT, INGE; polyethersulphone, capillary inner diameter 0.9 mm; pore diameter 0.02 μm and membrane surface area 80 m^2). The flux varied from 60 to 70 L/(m^2·h), and the filtration time was set to 45 min. Chemical enhanced backwash (CEB) with NaOH (pH ~12.2) and H_2SO_4 (pH ~2.1) was done one up to two times per day. Cleaning-in-place (CIP) intervals were dependent on the amount of fouling but generally occurred one up to two times per year with 300 mg Cl_{free}/L (pH ~12.2) by dosing NaClO and 4 g of oxalic acid/L (pH ~1.8). For ReUse Water 1, the filtrate was finally disinfected by UV radiation (sterilAir® AQD64-4 K).

For ReUse Water 2, the water was further treated successively by inline aeration, biologically active deep bed filtration (bed height 2 m in two serial columns; quartz sand, grain diameter 1–2 mm; filtration rate 5.5 m/h, empty bed contact time (EBCT) 20 min) and GAC filtration (bed height 2 m in two serial columns; Hydraffin AR, 8 × 30 mesh, Donau Carbon; filtration rate 5.5 m/h, EBCT 20 min). Deep bed filtration is required for catalytic and biological oxidation of dissolved Mn(II). GAC filtration was operated for removing micropollutants and DOC. Both filtration steps also serve for biodegradation of BDOC (biodegradable dissolved organic carbon), nitrification (microbiological stabilization) and the growth of autochthonous bacteria to lower the regrowth potential of pathogens (seed experiments).

To prevent massive biofouling on RO membranes and their pre-filters, NaClO and NH_4Cl solutions were added consecutively on line 1 directly into the UF filtrate stream in the stoichiometric ratio of 2:1. Both react *in situ* to 1 mg/L of monochloramine:

$$NH_4Cl + NaClO \rightarrow NH_2Cl + H_2O + NaCl \tag{1}$$

The hydraulic retention time of monochloramine was, due to the 1.8 m^3 UF filtrate tank, approximately 23 min. Free chlorine concentrations up to 0.1 mg/L were tolerated according to the RO manufacturer's requirements. Therefore, the dosage of sodium bisulphite as a scavenger for free chlorine was not necessary. Free and bound chlorine concentrations in the RO feed, concentrate and permeate were checked with the photometric DPD (N, N-diethyl-p-phenylenediamine) method. Dosing times of 22 h down to 4 h were tested, and total and intact cell counts (ICCs) were monitored online with flow cytometric analysis (OBA, Metanor AG). For preventing scale formation on the RO membrane, 3.7 mg/L of the scale inhibitor (Free-Flow 4, Dr Naehring) was added.

Non-woven fabric cartridge filters (WFMB melt-blow cartridge, polypropylene, nominal pore size 1 μm) protected the RO membrane against coarse particles. The load with TSS (total suspended solids) and biofilms of the filter was monitored by the differential pressure measurement. Two series of RO pressure vessels were operated in parallel, each fed by its own UF module. In each series, three 4-inch (101.6 mm) RO elements were housed in three RO pressure vessels. With this configuration, LPRO (low-pressure RO) elements from the Lewabrane® product series were tested in parallel (LANXESS Lewabrane® B085 ULP 4040, B085 FR 4040, B085 LE 4040; membrane area 7.9 m^2 per element). Finally, the RO permeate was disinfected with UV-light (sterilAir® AQD 64C-4 K).

Analytical methods and calculations

The chemical analytics of the water samples was done by the accredited laboratories of EUROFINS, IWW and OOWV. The analysis of the microbiological parameters was carried out by the laboratory of the Public Health Department of Aurich. The applied methods are listed in Table 3 (Appendix).

Several parameters have also been analysed onsite, such as the SDI (Silt Density Index, Portable SDI/MFI-Analyzer, Convergence), free chlorine and bound chlorine by the DPD method (chemicals: Huebers GmbH; photometer: ALLDOS 310-055-1000), pH, O_2 and conductivity (WTW Multi 3630 IDS) and turbidity (Nephla, HACH-Lange).

Since water temperatures can have a significant influence on the measured membrane permeability, it was normalized to the reference temperature of 20 °C for the purpose of monitoring the membrane resistance, including fouling effects (fouling layer and pore blockage). The following equation cited by Mallevialle *et al.* (1996) is used to normalize the UF permeability $P_{\vartheta,\mathrm{UF}}$ to 20 °C in which $\mathrm{TCF}_{20,\mathrm{UF}}$ is the temperature correction factor and ϑ the actual water temperature in °C.

$$\mathrm{TCF}_{20,\mathrm{UF}} = \frac{P_{\vartheta,\mathrm{UF}}}{P_{20,\mathrm{UF}}} = \exp\left[-0.0239\left(\frac{\vartheta}{[^\circ\mathrm{C}]} - 20\right)\right] \qquad (2)$$

To assess the fouling of the RO membrane, the normalized permeate flow was calculated according to the international standards (ASTM D4516 2010).

Sampling scheme

Throughout the pilot phase, water samples were taken monthly as random samples along the entire process chain. Sampling for the investigation of organic micropollutants was carried out as 24-h mixed samples at selected times in order to characterize the micropollutant retention of the RO and the GAC. The device for taking the mixed samples collected a distinct volume into a sampling vessel automatically every 30 min.

RESULTS AND DISCUSSION

Operation conditions

The pilot study started in August 2017. Up to December 2018, each treatment step was commissioned and optimized individually. In the period from January to December 2019, all treatment steps were operated in combination and further optimized to demonstrate the stability of the processes and the water quality.

In the WWTP of Nordenham, physical, chemical and biological processes are combined for the treatment of the municipal wastewater, which is collected from households, small and medium enterprises, and a hospital. The last treatment step of the WWTP is a clarifier. The sewer system consists of both separate and combined rainwater and sanitary sewers. In dry weather periods, the effluent volume flow is ~4.000 m^3/d. During rain events, it increases by the factor of three. The effluent shows the seasonal variations of high ammonium and nitrite peaks during periods with low temperature and can be characterized as a typical municipal WWTP effluent unaffected by big industry. One special characteristic of the raw and treated wastewater is the high content of dissolved manganese, which enters the sewer system with the groundwater. The results of the major physical–chemical parameters that have been monitored in a monthly routine are listed in Table 2.

As hygienic safety of customers and operators is a prevalent goal in water reclamation, a regular monitoring of traditional microbial hygiene indicator bacteria was performed. Figure 2 shows the results of this monitoring for the WWTP effluent.

Although the purpose for water reuse in the case study at Nordenham was not irrigation but industrial use, the elimination of micropollutants by RO and GAC has been investigated at the pilot plant, because water reuse for agricultural or landscape purposes is of interest in some areas in Germany. Table 1 shows a selection of micropollutants that were found in the wastewater effluent of the WWTP of Nordenham.

Produced water qualities: hygienic aspects

During the entire pilot phase, the UF proved to be an efficient microbiological barrier: the filtrates were – apart from the commissioning phase – free of *Escherichia coli* and coliform bacteria (<1 MPN/100 mL) as well as intestinal enterococci, *Clostridium perfringens* and *Legionella* sp. (<1 CFU/100 mL). If the maximum values in the feed (usually coliform bacteria $>1\times10^5$ CFU/100 mL and enterococci up to 2.2×10^6 CFU/100 mL, see Figure 2) are taken into account, permanent bacterial retention of more than 5–6 log units was achieved by the UF. Beside the traditional microbiological hygiene indicator bacteria, the total cell counts have been monitored by means of flow cytometry. The results of online flow cytometric measurements (sampling in cycles of 15 min) presented 10^6–10^7 intact cells/mL in the feed and 10^2–10^3 intact cells/mL in the filtrate of the UF. Thus, this real-time monitoring method indicated only 3–4 log units of bacterial retention. The different retention results can be explained by the differences in the investigation methods. As with the flow cytometry all floating cells are detected, also autochthonous microbial flora from the non-sterile filtrate containing parts (e.g. cells which are released from biofilms inside the inner wall of the filtrate pipes) are counted, which actually should not be taken into account for the evaluation of the UF retention. This means, by looking at the total cell counts in the non-sterile system, that the UF retention is likely to be underestimated. As

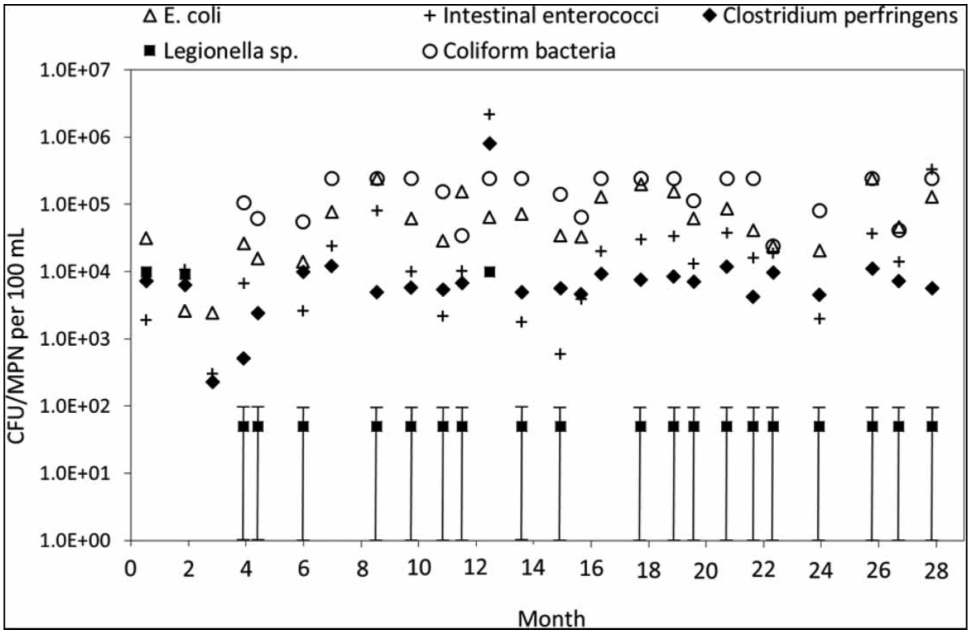

Figure 2 | Microbiological findings of hygiene indicator bacteria in monthly random samples of the WWTP effluent after pre-filtration (sieve, 200 μm) during the entire period of the pilot plant operation.

Table 1 | Selection of detected organic micropollutants in the effluent of the WWTP (24-h mixed samples)

		Minimum	**Average**	**Median**	**Maximum**	***n***
1H-Benzotriazole	µg/L	4.3	6.0	6.4	7.7	8
4-Methyl-1H-benzotriazole	µg/L	6.0	10.2	10.5	15.4	8
5-Methyl-1H-benzotriazole	µg/L	0.3	0.6	0.6	0.9	8
Acesulfame	µg/L	1.0	2.0	1.3	3.4	7
AMPA	µg/L	1.4	2.1	1.8	3.1	8
Carbamazepine	µg/L	0.3	0.7	0.7	1.0	9
Clarithromycin	µg/L	0.1	0.2	0.2	0.4	9
Diclofenac	µg/L	1.4	2.6	2.5	3.6	9
EDTA	µg/L	0.1	12.6	15.1	27.8	7
Glyphosate	µg/L	0.3	0.4	0.4	0.7	8
Iomeprol	µg/L	0.1	1.3	1.2	2.5	8
Iopromide	µg/L	2.5	8.8	8.5	16.4	8
Metoprolol	µg/L	0.7	1.7	1.4	3.5	9
Sulfamethoxazol	µg/L	0.0	0.0	0.2	0.3	9
TMDD	µg/L	0.2	1.2	1.0	3.5	8

microbiological monitoring was a main focus within the MULTI-ReUse project, a separate publication on this topic has been submitted by Nocker *et al.* (2020). For further detailed information on methods and results, this publication is recommended. Virus retention was not examined during the MULTI-ReUse project. However, for the chosen UF membrane type, a retention of >4 log units was detected in a preliminary project in the laboratory experiments using MS2 bacteriophages (Lipp *et al.* 2017) with a diameter of approximately 24–26 nm (Wick & McCubbin 1999).

The above-mentioned hygiene indicators could not be detected in the filtrates of the sand and GAC filters and in the RO permeates as well. These results are certainly supported by the bacteria retention of the UF, which protects the whole subsequent treatment steps from microbial contaminations of the WWTP effluent. The total cell counts in the filtrates of the sand and GAC filters were significantly higher (10^5–10^7 cells/mL) than in the UF filtrate. This result was expected as the filters were operated to enable biological activities inside the filter bed. The RO permeates showed total cell counts on the same low level as the UF filtrates. However, a bacterial regrowth in the UF filtrate and the RO permeate was detected after storing the samples for 7 days at 22 °C, if no chemical disinfectants were applied. This proves that the water samples still contained assimilable nutrients and an autochthonous bacterial flora which is able to multiply under these conditions. For further information, refer Nocker *et al.* (2020).

Produced water qualities: physical–chemical parameters

Results on the chemical composition of the water generated and the WWTP effluent are shown in Table 2.

The ReUse Water 1 is free of particles and pathogens. Flocculation before the UF reduced the DOC concentration on average to 10 mg/L (−19%) and the concentration of P_{total} to 0.061 mg/L. The concentrations of the metals such as aluminium and iron contained in the wastewater effluent were also significantly reduced by 61 and 93%, whereas dissolved salts pass through the UF membrane.

Further treatment of ReUse Water 1 by filtering it through a biological active quartz sand filter and a GAC filter leads to ReUse Water 2 quality. ReUse Water 2 is additionally low in manganese (−68%), ammonium (−70%) and organic micropollutants after the GAC passage. Figure 3 shows the removal efficiency of the GAC filter for several micropollutants at increasing operating times,

Table 2 | Average values and standard deviation of the analysis results of the WWTP effluent and three produced ReUse Water types

		WWTP effluent		**ReUse Water 1**		**ReUse Water 2**		**ReUse Water 3**	
Parameter	**Unit**	**Average St. dev.**	***n***	**Average St. dev.**	***n***	**Average St. dev.**	***n***	**Average St. dev.**	***n***
pH	–	6.9 ± 0.1	25	6.8 ± 0.1	24	6.8 ± 0.1	18	5.3 ± 0.2	23
Conductivity (25 °C)	μS/cm	1.314 ± 375	26	1.329 ± 376	25	1.401 ± 394	18	26.6 ± 12.0	24
Turbidity	NTU	2.26 ± 1.9	24	0.14 ± 0.11	24	0.24 ± 0.18	18	0.19 ± 0.22	21
N total	mg/L	8.3 ± 2.9	25	7.6 ± 2.7	26	8.4 ± 2.4	16	0.39 ± 0.19	23
P total	mg/L	0.37 ± 0.14	26	0.06 ± 0.06	26	0.09 ± 0.08	17	0.05 ± 0.06	23
COD	mg/L	39 ± 9	27	27 ± 11	27	21 ± 5	15	<5	24
TOC	mg/L	12.4 ± 2.5	30	10.0 ± 3.4	27	8.7 ± 2.5	17	0.39 ± 0.43	24
SAC_{254}	1/m	31.1 ± 17.4	21	23.9 ± 8.5	24	22.6 ± 12.5	17	0.21 ± 0.17	22
SAC_{436}	1/m	2.97 ± 1.69	6	1.12 ± 0.4	24	0.9 ± 0.3	18	0.05 ± 0.06	24
AOX	mg/L	0.05 ± 0.07	22	0.03 ± 0.02	21	0.02 ± 0.01	18	<0.01	18
Al total	mg/L	0.07 ± 0.03	33	0.03 ± 0.03	32	0.02 ± 0.01	18	0.009 ± 0.009	25
Fe total	mg/L	0.60 ± 0.19	35	0.04 ± 0.03	31	0.02 ± 0.02	18	0.007 ± 0.007	25
Mn total	mg/L	0.41 ± 0.12	36	0.41 ± 0.10	33	0.13 ± 0.21	16	0.003 ± 0.002	25
Cl^-	mg/L	215 ± 102	24	215 ± 102	28	245 ± 113	18	2.9 ± 2.0	24
SO^{4-}	mg/L	95.7 ± 25	23	91.8 ± 26	28	101 ± 25	18	0.6 ± 0.4	24
NH^{4+}	mg/L	1.75 ± 2.47	27	1.86 ± 2.52	26	0.39 ± 0.66	18	0.10 ± 0.09	22
NO_2^-	mg/L	1.10 ± 1.94	27	1.12 ± 1.94	27	0.24 ± 0.41	18	0.06 ± 0.05	24
O_2 (aq)	mg/L	3.53 ± 1.37	25	6.31 ± 0.69	25	2.17 ± 1.82	18	5.73 ± 1.76	24
Ca^{2+}	mg/L	78.8 ± 3.2	29	78.8 ± 12.0	29	77.5 ± 13.8	17	0.1 ± 0.1	25
Silicate total	mg/L	27.9 ± 5.0	20	27.6 ± 4.9	26	27.2 ± 5.0	18	0.25 ± 0.1	25
Base-neutralizing capacity	mmol/L	0.9 ± 0.4	27	1.0 ± 0.4	27	0.8 ± 0.3	16	0.7 ± 0.4	22
Alkalinity	mmol/L	4.5 ± 0.8	28	4.4 ± 1.3	27	3.9 ± 0.8	18	0.2 ± 0.2	22

characterized by the treated ‘bed volume’ (m³ filtrate/m³ GAC filter bed). From Figure 3, the progress of the breakthrough of some micropollutants can be observed. As one extreme, the benzotriazoles can be mentioned, which were removed at nearly 100% over the complete operation time. On the other side, at the end of the pilot phase, a certain mass of already adsorbed sulfamethoxazole had been replaced by competing organics with a desorption effect: the concentration sulfamethoxazole was higher in the filtrate than in the raw water (negative removal). Due to the biological degradation of the DOC in both filters and adsorption on the GAC, the concentration of DOC is reduced by 10% additionally to the removal caused by flocculation (total retention 30%).

ReUse Water 3 is the water with the highest purity. The average concentration of TOC is 0.3 mg/L (−97%). By using the RO membrane filtration, strong attention was paid to the elimination of salts and hardness formers, as these are critical in many industrial process steps. However, it was not the aim to receive a water free of salinity, but to find a good compromise between the low TDS content of the permeate on the one hand and the low energy demand of the pumps on the other hand.

The retention of organic micropollutants was constantly high. The lowest retention was detected for all benzotriazoles (83% on average). For the purpose of groundwater recharge, depending on the retention of the RO membrane and the non-polar micropollutants contained in the RO permeate, a final GAC filtration step could be operated optionally. This depends on the properties of the micropollutants remaining in the permeate and also on the need for a redundant barrier.

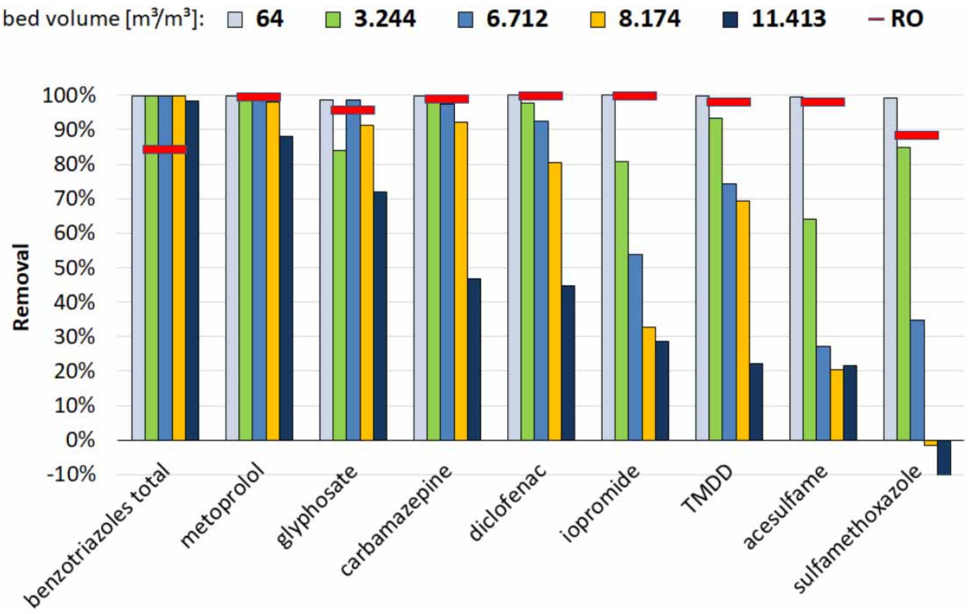

Figure 3 | Retention of micropollutants in the GAC filter according to treated bed volumes (m³ filtrate/m³ GAC filter bed) and the averaged retention by the RO membrane over the entire operation time.

Process stability UF

To find optimal operating conditions, coagulants based on both aluminium ($Al_2(OH)_5Cl\cdot 2–3H_2O$) and iron ($FeClSO_4$ and $FeCl_3$) were tested for UF. After commissioning, both UF lines were operated in parallel with a flux of 60 L/(m^2·h) and 3 mg Al^{3+}/L. The contact time of the coagulant was 20 s. The results show that the UF performed well with high stability. Low increases in transmembrane pressure (TMP) with constant flux resulted in high permeability of around 300 L/(m^2·h·bar). The operational behaviour for a period of 2 weeks (week 0–2) is shown in Figure 4.

After this period, UF2 was operated with $FeClSO_4$ at a concentration of 5 mg Fe^{3+}/L, while for UF1 the Al-dose was kept constant. This resulted in an unstable filtration process and permeability dropped steeply in UF2. An increase of the CEB frequency from 1 to 2 CEBs/d could not stabilize the filtration process. Increasing the iron concentration up to 10 mg Fe^{3+}/L improved the performance and verified the suitability of all tested iron- and aluminium-based coagulants for the use in this application.

The different filtration behaviour can be explained by the different characteristics of the coagulants. Al-based coagulants are supposed to build up denser cake layers than Fe-based coagulants even at low dosages, which are more efficient in retaining foulants (Ma *et al.* 2014). Nevertheless, the Fe-based coagulants were chosen for further optimizations because quite high silica contents are present in the raw water and the danger of irreversible aluminosilicate scaling on the RO membranes was supposed to be avoided.

Over the course of the pilot study, the type of dosage of the coagulant was changed several times, particularly when using $FeCl_3$. With continuous coagulant metering, frequently sharp TMP increases at the beginning of filtration intervals were observed (see Figure 5). Presumably, the dosed amount of coagulant (6 mg Fe^{3+}/L) was insufficient to include or adsorb the organic matter into flushable hydroxide flocks. This resulted in pore blockage especially at the beginning of the filtration intervals, when the layer of coagulant on the membrane is still thin. CEBs were able to reduce the TMP, but were not efficient enough to restore the initial level. To counteract an initial increase in TMP, a higher coagulant dosage was used at the start of each filtration process (12 mg Fe^{3+}/L). After a certain time (e.g. a third of the filtration time), the dosage rate was significantly reduced (3 mg Fe^{3+}/L). This method is a combination of the so-called initial hydroxide coating (Buchta *et al.* 2017) on the membrane and continuous dosing. When using this

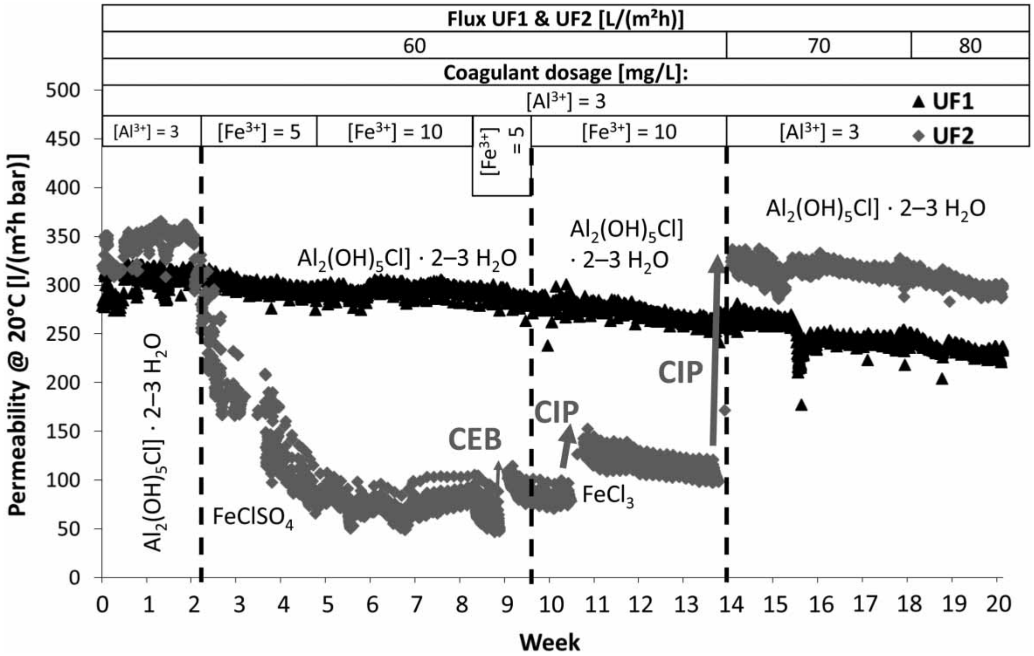

Figure 4 | Performance of UF1 and UF2 by treating the WWTP effluent with several amounts of Fe(III)- and Al(III)-based coagulants.

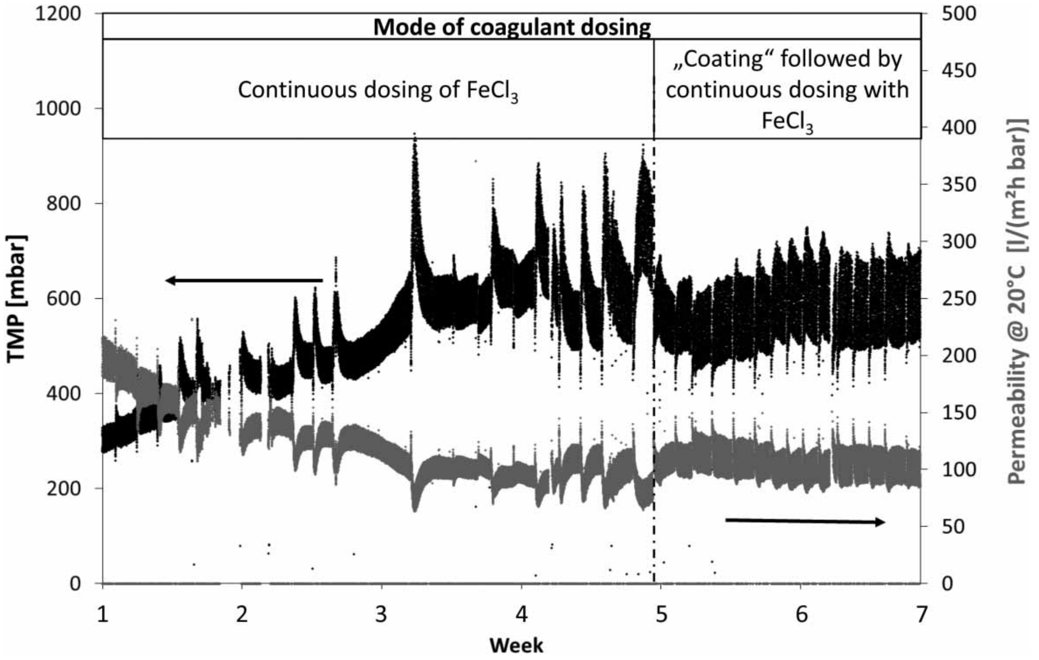

Figure 5 | Improving the performance of the UF by changing the mode of coagulant dosing.

method, a filter cake is built up on the membrane surface rapidly during the high-dosage phase directly at the beginning of a filtration cycle. This represents an additional barrier to dissolved and colloidal matter which protects the membrane from foulants and can be removed easily by the hydraulic backwash (Ma *et al.* 2014). The different

TMP courses are shown in Figure 5. It is evident that the stable continuous dosing led to stronger TMP peaks compared with the combined dosing strategy. The operation became much more stable despite adding the same amount of 6 mg Fe^{3+}/L coagulant on average for the overall filtration cycle.

Figure 6 shows that the UF capillary membrane is very sensitive to malfunctions such as the failure of a metering pump (e. g. caused by air bubbles) for the addition of H_2SO_4 during a CEB. In this case, the operation stabilized automatically after the problem was resolved. In the event of a failure of the coagulant metering pump, however, it can be assumed that the membrane will not recover on its own, but will be needed to be subjected to intensive cleaning. Operating problems of the WWTP, which were confirmed after consultation with the operator, also showed high stress levels for the UF process. A deterioration in the quality of the WWTP effluent can disturb immensely. In some cases, the increase of the coagulant dosage rate and also increase of the CEB frequency (2 CEBs/d) were able to stop the increase of the TMP. It was finally necessary to switch off the pilot plant and to clean the UF with NaOH and NaClO (pH 12.2 and 300 mg Cl_{free}/L).

Process stability RO

During the first year of the pilot phase, the focus for the RO process was lying on the membrane development and the design of the spiral-wound elements. A new membrane type for the filtration of low to moderate saline waters has been successfully established. The aim of keeping a high retention of ~98% for TDS, 97% for TOC and >90% for the majority of organic micropollutants, while reducing the operational pressure by about 30% compared with a standard product, could be reached. Further details of these experiments are described elsewhere (Ogier & Lipnizki 2019)

Biofouling and scale formation can reduce the performance of the RO. To ensure operational stability for the production of ReUse water 3, monochloramine (NH_2Cl) was used in addition to antiscalant for RO1 to avoid biofouling. RO2 served as a reference line without monochloramine disinfection. The dosage of monochloramine was noticeable, both in the differential pressure of the cartridge filter and also in the permeability of the filtration process. Especially in the summer months due to high water temperatures of above 20 °C and the highly

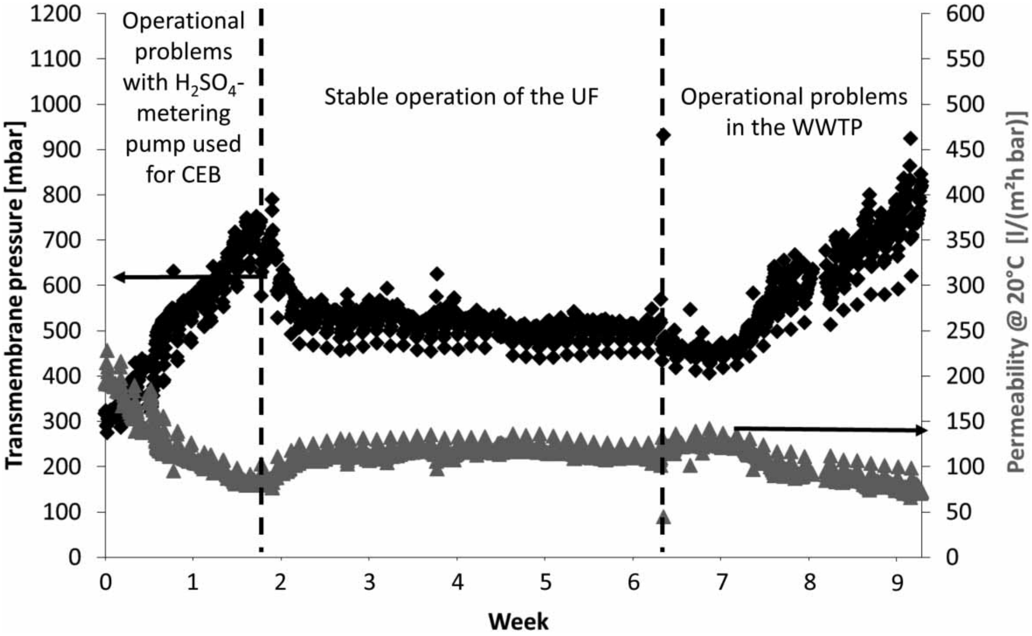

Figure 6 | Stable operation of the UF and effects in case of operational disturbances both at the treatment plant and at the pilot plant.

concentrated inlet water (lack of rain water in the mixed water sewer system), the biological activities were clearly visible on the cartridge filters. At times with peak water temperatures, the cartridge filters had to be replaced after 2 weeks at the latest. By using monochloramine, this period could be extended up to 2 months. Furthermore, the cleaning of the UF filtrate tank (storage for backwash water) in 2-week intervals was no longer necessary. Figure 7 shows the effect of 1 mg NH_2Cl/L for a dosing time of 22 h/d on the cartridge filter upstream to RO1 in comparison to that of RO2 over a period of 4 weeks (see Figure 8) in the hot summer of 2019. The deposition on these cartridge filters was examined by preparing a distinct piece of each cartridge with aqua regia solution and analysing the extract by ICP-OES. The summarized mass of all analytes in relation to the initial sample weight was significantly lower at RO1 (9.7 g/kg) compared with RO2 (23 g/kg). The chemical elements with the highest share among all elements identified by this method were iron (RO1: 2.8 g/kg; RO2: 11 g/kg) and calcium (RO1: 3.5 g/kg; RO2: 6.2 g/kg). The fact that the Fe deposition on the RO2 cartridge was about four times higher than on the RO1 cartrige and the amount of phosphorous was also significantly higher (RO1: 0.16 g/kg; RO2: 1.0 g/kg) confirms the assumption that the biological activity was effectively controlled by the disinfectant dosage.

The concentration of chloramine (i.e. bound chlorine) was regularly checked, using the DPD method, before the water entered the RO pressure vessel to ensure that the membrane would be in contact with the disinfectant. The average concentration of bound chlorine was 0.57 ± 0.21 mg/L depending on the quality of the feed water.

To reduce the dosing amount of NH_4Cl and NaClO to the bare minimum, tests were carried out with different dosing times between 22 and 4 h/d. Flow cytometry was used to assess the effectiveness of the disinfection. An optimal dosing time of 13 h/d could be determined without increasing the ICCs. Detailed results from these studies are presented by Nocker *et al.* (2020). The calculated salt retention based on the conductivity was similar in both RO lines. This observation and the results of a membrane autopsy after this operating phase of 6 months indicated that the membranes have not been deteriorated by the disinfection. Due to the membrane passage of chloramine, bound chlorine could be detected with concentrations of 0.39 ± 0.24 mg Cl_{bond}/L in the permeate.

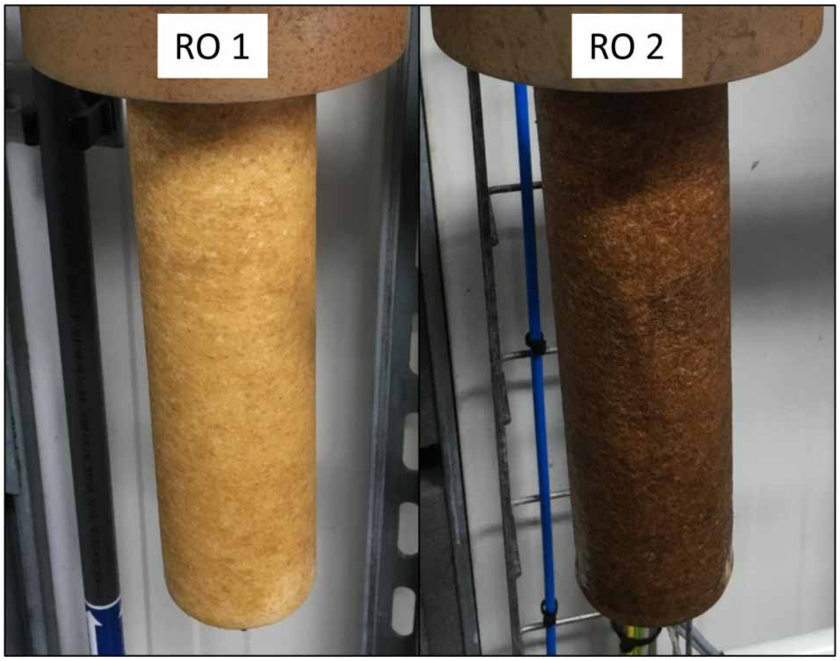

Figure 7 | Cartridge filters of RO1 (with NH_2Cl) and RO2 (without NH_2Cl) in comparison after 4 weeks of operation (26 June 2019–23 July 2019).

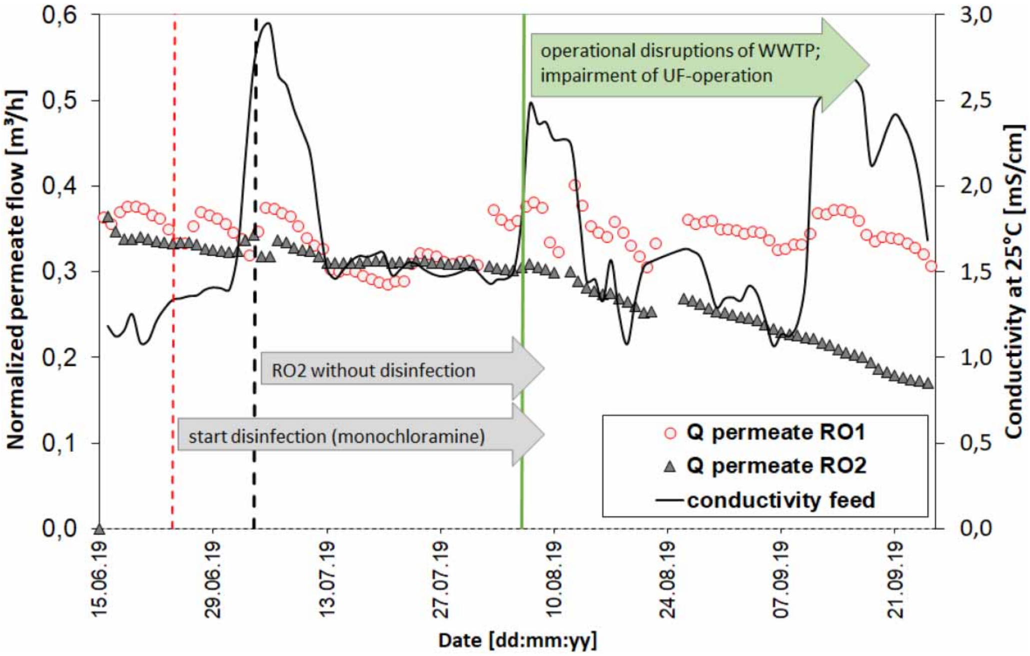

Figure 8 | Permeabilities of RO1 (with NH_2Cl) and RO2 (without NH_2Cl) and conductivity in the feed.

The efficiency of the filtration operation via the RO was monitored via the process data measured online. Figure 8 shows the normalized permeate flow for RO1 (with NH_2Cl dosage) and RO2 (without NH_2Cl dosage) during the summer of 2019. During this operational period, conductivity peaks of up to 3,000 μS/cm in the RO feed were detected. The cause could not be determined precisely. Despite these conditions, the RO lines showed a stable operation over several months. A malfunction of the WWTP operation at the beginning of August 2019 led to an impaired operation of the upstream UF lines (Figure 6, week 6). Consecutively, the high stress load on the RO membranes caused a continuous decline of the normalized permeate flow in RO2. By using monochloramine, however, the operation of the RO1 was stabilized and the operating time until the next CIP cleaning could be extended even under feedwater conditions.

Waste residues

Membrane processes offer many advantages. But one important disadvantage is the volume of concentrates and residues produced during the treatment process and their correct post-treatment or disposal. When operating UF and RO membranes, chemically contaminated water is generated, which must be disposed of properly. This includes the sludge-containing backwash water (9.1%, i.e. recovery 99.1%) from UF processes and chemical cleanings of UF (CEB and CIP) and RO (CIP), as well as the concentrate of RO with scale inhibitors. In the MULTI-ReUse project, the wastewater from the pilot plant was returned to the WWTP inlet. In the case of a large-scale reclamation plant, the relation between raw wastewater and residues from the reclamation plant could approximately reach a ratio of 4:1. The high salinity of the RO concentrate in the complete return of all waste residues could result in reduced performances of the biological treatment in the WWTP, as well as for the reclamation plant itself. Disposing of the RO concentrate therefore requires a separate discharge or further treatment.

One option, which was still being examined at the time this publication was written, is a discharge of RO concentrate into the River Weser. This seems to be the easiest way of disposal; however, a discharge permit is required. The Weser River is a navigable waterway and the river section at the case study site belongs to the estuary (brackish water). The costs for the discharge permit, which is dependent on the quality and volume, should be considered.

Backwash water of UF is largely unaffected by dissolved water additives. The hydroxides of the coagulation agent and PAC, if use is required, are the only water treatment substances added into the treatment process prior to the UF. The particulate matter, including metal-containing flocs (Fe^{3+} or Al^{3+}) and PAC, will be eliminated during the conventional WWT process. In addition, PAC stabilizes the biological treatment of the WWTP and its sludge treatment (dewatering) (Menzel 1997). Its loading capacity can be widely exploited because on the one hand the adsorption in the PAC-UF process does not reach equilibrium conditions and on the other hand the PAC will be applied (counter-current) in two treatment steps with a high concentration level during the biological treatment and a lowered level during the UF step (DWA 2019).

The chemically contaminated backwash water of the CEB (NaOH: pH ~12; H_2SO_4: pH ~2) and cleaning solutions from CIP cleanings for UF and RO, containing NaOH (pH ~12); oxalic acid/HCl (pH ~1.8) and small amounts of EDTA (only for RO cleaning), should first be neutralized in storage tanks. If no other potentially harmful cleaning chemicals (such as hypochlorite) are used, this waste stream can then be fed into the WWTP inlet.

Sludge-containing backwash water from the deep bed or GAC filtration as they were used in the treatment chain of ReUse Water 2 are free of additional chemicals. The contained particulate matter can easily be eliminated by the conventional WWT process; therefore, a disposal into the WWTP inlet should be suitable (DWA 2019).

Selection of material for the transport piping system

For transporting the ReUse waters to the consumers (point of use), an additional distribution system has to be implemented. The following materials for the piping should be considered:

- metallic materials (metal): cast iron, unalloyed or low alloyed steels,
- metal with internal cement mortar lining (metal/CM) and
- polymeric materials (polymer): most likely polyethylene (PE).

Metallic pipes without CM only have to be considered if an old, existing drinking water distribution system is intended to be used for the distribution of the ReUse waters. The chemical properties of the ReUse waters (according to the values in Table 2) and the corrosion behaviour of the respective materials have to be correlated.

ReUse Water 1 can be described by a high neutral salt content (chloride, sulphate and nitrate). Additionally, the calcite solubility, which is associated with pH, calcium concentration and other parameters of the carbonate system of the water, is relatively high so that the formation of protective surface layers inside metallic pipes may be hindered. This can also be a threat for cement-based materials as the lime within the cement can be dissolved. Beside the weakening of the structure of the cement, this can also lead to the release of particles into the water. Also manganese in ReUse Water 1 can precipitate. These particles and precipitates can cause blocking of valves, depending on the amount of water to be transported (freight). The content of ammonium can cause problems with copper alloys (brass), which might be important when considering the materials of the operating networks of the consumers (e.g. valves and fittings).

The characteristics of ReUse Water 2 are quite similar to those of ReUse Water 1. The manganese and ammonium content could be lowered significantly by additional aerobic biological filtration.

In ReUse Water 3, the concentrations of many water constituents have been lowered significantly, including neutral salts. On the other hand, the alkalinity is almost completely eliminated. As this parameter acts as a buffer within the corrosion system, the water can be characterized as poorly buffered from the corrosion chemical point of view. The pH is relatively low and the carbonate system of this water is far away from equilibrium, leading to very high calcite solubility.

The most important characteristics and the consequences for the selection of materials for distribution networks are summarized by a traffic light colour scheme in Figure 9.

Polymeric materials (e.g. PE pipes) are appropriate for all three ReUse Waters. In the case of ReUse Water 1, manganese has to be considered as described above. Metallic- or cement-based materials require further stabilization of the ReUse Waters (e.g. pH rising and buffering). Due to the high neutral salt content in ReUse Waters 1 and 2, metallic

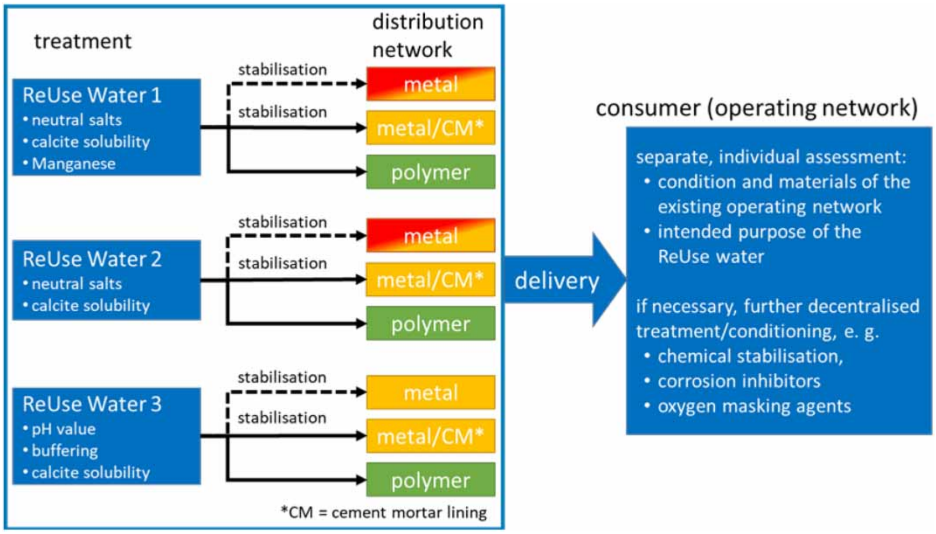

Figure 9 | Decision tree for the selection of materials for distributing the ReUse Waters.

materials can only be used under certain conditions regarding the network structure and the service conditions and might require the addition of corrosion inhibitors. If old, existing drinking water networks made of metallic materials are to be used for the waters 1 or 2, a detailed and individual assessment of the condition of the existing network is necessary in advance.

Regarding the service conditions, it has to be considered that they may differ between drinking water and reuse water. In contrast to a drinking water network, the conditions in a reuse water network – especially for industrial purposes – can be characterized by a constant, plannable consumption.

The materials and conditions of the respective operating networks of the consumers of reuse waters also have to be taken into account, as well as the intended purpose of usage. Due to the great variety between the consumers, this can only be done by individual assessments and will not be further discussed here.

CONCLUSION

In the MULTI-ReUse project, a modular treatment concept for the reuse of WWTP effluent was developed. To ensure consistent stable water quality, reliable, i.e. robust treatment technologies were used on the one hand, and extensive online monitoring in combination with laboratory analyses on the other.

The importance of considering the treatment efficiencies of the WWTP and the reclamation plant as one unit has to be emphasized. There is still a lack of data for the evaluation of the influence of operational problems in the WWTP (e.g. in drought or heavy rain periods) on the membrane processes in the reclamation plant. It is also suspected that certain chemical water constituents occasionally found in the WWTP effluent can disrupt the in/out UF process. The experiments showed that under normal circumstances at least 6 mg Fe^{3+}/L should be used for a stable filtration process when using $FeCl_3$ and at least 2 mg Al^{3+}/L when using $Al_2(OH)_5$-Cl·2–3 H_2O as a coagulant. A combined coagulant coating strategy was successfully used to stabilize the UF process.

For protecting the RO membrane against biofouling and keeping the performance, high *in situ* formation of monochloramine prior to the RO membrane was highly effective. Furthermore, it suppresses the regrowth of bacteria in the RO permeate by passing the membrane and stabilizing the water. Especially in hot summers and drought periods, the potential of biofouling formation is quite high. Consequentially, energy and maintenance costs for intensive cleaning of membrane pipes can be reduced and the treatment be more efficient.

The evaluation of possible materials for a distribution network has shown that in most cases individual assessments are necessary regarding the operating conditions and the purpose of usage.

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DATA AVAILABILITY STATEMENT

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

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