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Long-term operation of ultrafiltration membrane in full-scale drinking water treatment plants in China: Characteristics of membrane performance

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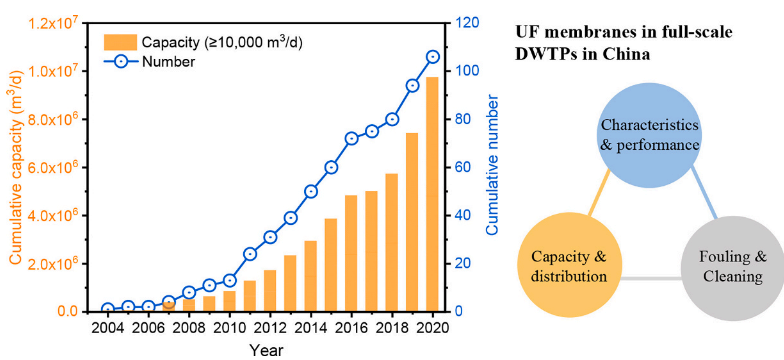
HIGHLIGHTS

- UF membrane in 106 full-scale DWTPs in China reached ~10 million m^3/d by 2020.
- DWTPs with UF displayed spatial distribution, with most in Eastern China.
- Combination of UF with traditional drinking water treatment units employed widely.
- PVDF occupied 73.7 % of membrane material and 60 % of UF operated at submerged mode.
- Over half of the UF membranes in full-scale DWTPs operated for over 5 years.

GRAPHICAL ABSTRACT



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ABSTRACT

The installed capacity of ultrafiltration (UF) membranes in drinking water treatment plants (DWTPs) has increased significantly, it is necessary to summarize the long-term characteristics of UF membranes in full-scale DWTPs in China. Firstly, the characteristics of UF membranes applied in large-scale DWTPs in China were systematically evaluated based on capacity, location, membrane materials and filtration mode. By 2020, the total capacity of large-scale DWTPs using UF membranes has reached ~10 million m^3/d. Polyvinylidene fluoride was the dominant UF membrane material with a proportion of 73.7 %, followed by polyvinyl chloride. Secondly, the performance of UF membranes in various typical DWTPs during long-term operation was analyzed. UF membrane in five typical treatment processes displayed good contaminant removal performance, while the combination of UF membrane with coagulation, sedimentation and filter was widely used. The fouling evolution of UF membranes during long-term operation was systematically analyzed. Then, the effect of pretreatment methods and cleaning approaches on membrane in full-scale DWTPs were assessed. The use of hydraulic cleaning, maintenance cleaning and recovery cleaning could ensure the stable operation of UF membranes, while these

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cleaning methods may adversely influence membrane lifetime. This study could provide useful guidance for long-term operation of UF membrane in full-scale DWTPs.

1. Introduction

Water crisis is a critical issue for human beings, with more than 650 million people in the word lack of clean and safe water, and it is one of the greatest challenges in 21st century to provide water universally in a safe, reliable and affordable manner [1]. Conventional treatment processes which are comprised of coagulation, sedimentation, sand filter and disinfection may be difficult in producing drinking water effectively due to microbiological contamination. The outbreak of cryptosporidiosis in Milwaukee (USA) in 1993 promoted the rapid development of emerging technologies in drinking water treatment plants (DWTPs) [2]. Increasing attention has been paid on membrane technologies in DWTPs to ensure the safety of drinking water. Since the first interest in membrane filtration in DWTP in 1987, ultrafiltration (UF) membrane has been widely employed for drinking water production [3]. UF technology is a promising alternative in DWTPs due to its superiority in ensuring biological safety, and the UF global market is estimated to reach $2.14 billion in 2023 [4]. However, membrane fouling is a key issue for the widespread application of UF, resulting in reduction of water flux and separation efficiency, which in turn reduces membrane lifetime and increases operating costs [5,6].

The performance of UF membrane depends on many influencing factors, such as feed water, membrane characteristics and operation conditions [4,6,7]. For feed water properties, the characteristics of raw water (e.g., hydrophilicity, molecular weight distribution and charge) [8–11] greatly influenced the selection of the whole treatment process, including appropriate pretreatment and/or posttreatment units. With respect to membrane characteristics, a series of inorganic and polymeric UF membranes were reported for drinking water production, with the polymeric ones widely used due to their cost-effective properties [12–15]. Membrane materials with anti-fouling and anti-ageing are ideally required [16], while the economic feasibility and maturity should be considered carefully. As for operation conditions, the filtration parameter, hydraulic cleaning parameter, and chemical cleaning conditions greatly influenced the UF membrane performance [13,17–19]. Generally, there are still problems for the adaptation of pretreatment to pollutant removal to ensure the stable operation of UF membrane in DWTPs, and the formation and regulation of membrane fouling using appropriate operation conditions.

The primary concerns in the UF application include filtration flux, cleaning performance, membrane life and operation cost. Filtration flux is a main parameter for UF design, playing a great role in membrane performance [20,21]. A series of operation conditions involving filtration flux have been proposed to prevent membrane fouling, such as under sub-critical flux operation [22,23], limiting flux [24], threshold flux [25,26] and sustainable flux [20]. Alternatively, gravity-driven membrane (GDM) ultrafiltration has been developed, and it is characterized by low driving pressure (0.4–1.0 m) and stable flux (e.g., 2–10 LMH) [21]. The GDM system is especially suitable for decentralized water supply, while the low flux would obviously increase the capital expenditure in full-scale application [21,27]. Membrane cleaning is an important approach for alleviating fouling to maintain the sustainable operation of UF membranes, and cleaning methods can be divided into physical cleaning, chemical cleaning and non-conventional cleaning approaches (e.g., ultrasonic and electric cleaning) [7]. As a key physical cleaning, backwashing is carried out with a reversed flow pushing from the permeate side to the feed side via a membrane using water or air (i.e., hydraulic backwashing or air backwashing) [13]. Combination of both hydraulic backwashing and air backwashing is usually employed in full-scale operation [28]. As the physical cleaning efficiency decreased in long-term operation [28], it is necessary to perform in-situ maintenance cleaning (e.g., NaClO) and off-line chemical cleaning (e.g., an alkali washing and an acid washing) [7,29]. Together with membrane fouling (or resistance), the frequent use of chemical and physical cleaning would result in membrane ageing [30–33], and eventually the replacement of membranes is required [32]. Another concern in UF operation is operation cost, which primarily contains energy consumption, chemical consumption, labor costs and so on. The energy consumption occupies a large proportion in the total treatment cost, while energy and chemical consumption would increase significantly in long-term operation [28].

Up to now, only a few studies involved the long-term operation of UF membranes in full-scale DWTPs [28,31,33–36]. Seven-, eight- and one-year operation of UF membranes were analyzed by Yu et al. [28], Touffet et al. [31] and Xiao et al. [36], respectively, who concluded that membrane foulants included inorganic matter, organic matter and microorganisms, among which protein and protein-like substances had an important impact on irreversible fouling. Yu et al. [35] pointed out that the key fouling factors varied with operation time, while Robinson et al. [33] demonstrated that older membrane was more sensitive to fouling than the new one, and higher doses of NaClO resulted in faster changes in membrane metrics to expected. With the rapid development of UF membrane in DWTPs, it is necessary to systematically summarize the experience and problems in long-term operation. The existing researches focused on only a specific case which was not enough for widespread application of UF, and it is necessary to summarize the overall application of UF in full-scale DWTPs in China.

Therefore, in this work, we have analyzed 106 full-scale DWTPs using UF in China by 2020 and give a conclusion of the current status of applications and look forward to future development. Firstly, we will give an overview of the full-scale UF membrane application in DWTPs in China. Secondly, the performance of UF membranes in various typical DWTPs during long-term operation is to be assessed, focusing on water purification and membrane fouling. Then, membrane filtration performance and the approaches for membrane fouling control in full-scale DWTPs would be summarized. Finally, the challenges and perspectives of UF-based membranes on the established DWTPs with UF from biological safety, membrane fouling, membrane cleaning and membrane ageing will be discussed. This study aims at providing a guide for the sustainable application of UF technology in full-scale DWTPs.

2. Overview of full-scale UF membrane applications

2.1. Current status of water quality of source water in China

Fig. 1 presents the water quality of the major surface water used for drinking water supply in China [6]. According to the water quality, surface water and groundwater can be classified into 5 grades, including Grade I, II, III, IV and V (Chinese standard GB3838–2002 and GB/T 14848–2017) [48,49]. Note that there are obvious differences in standard for surface water and groundwater quality, as listed in Table S1 (Supporting Information). Only the surface water of Grades I-III is able to be used as drinking water resource of centralized water supplies, while groundwater of Grades I-III and Grade IV after appropriate treatment can be used to produce drinking water [48,49]. Although the overall change in water quality for fresh water is becoming better in recent years, water pollution is still a severe issue in local regions. For surface water, in 2021, the percent of lakes reaching Grade I ~ III is 72.9 % and that for rivers is 87 %. Of the 10 water resource zones of Level I in mainland China, in the Haihe River and Songhua River, the proportion of Grade IV, V and inferior V are 39 % and 31.6 %, showing relatively poor water quality (Fig. 1c-1d). As important drinking water sources in

China, lakes and reservoirs are also facing the challenges of water pollution and subsequent degradation. The discharge of urban sewage and industrial wastewater is the main source of contamination in all of these rivers, and the primary pollutants include ammonia nitrogen, biochemical oxygen demand (BOD_5), chemical oxygen demand (COD) and petroleum compounds [6]. Eutrophication remains the most critical problem of lakes in China today, with Taihu Lake, Dianchi Lake and Chaohu Lake offering the most grievous examples of this phenomenon [50,51]. For example, the grade of Taihu Lake's water quality has dropped one class every 10 years over the past 30 years [52]. On the other hand, the standard for drinking water is becoming more and more strict [53,54]. With respect to groundwater, the quality is quite stable (Fig. S1, Supporting Information), with the percentages of Grade I-IV (or excellent, better, good and poor) of 79.4–85.3 %. However, the water resource with Grade IV which needs proper treatment as domestic water resource occupied the largest part in recent years (66.9–70.9 % in 2018–2020). To fill the gap between the source water quality and drinking water standard, UF membrane processes show alternative applications in DWTPs for sustainable drinking water supply.

2.2. Distribution of DWTPs using UF membranes

Fig. 2 shows the cumulative capacity and number of full-scale DWTPs in China, and a capacity of more than 10,000 m^3/d was summarized. The detailed information about the full-scale DWTPs using UF membranes is listed in Table S2 (Supporting Information). As presented in Fig. 2a, four phases could be divided for the application of UF membranes in DWTPs from 2003 to 2020. To be specific, there were several full-scale DWTPs in China using UF membranes since 2003 and the total capacity reached 380,000 m^3/d. The first full-scale UF membrane plant was Cixi Hangfeng Water Supply Plant where UF was as pretreatment process for reverse osmosis (RO) in 2005, while UF membrane was firstly used in Ducun Water Supply Plant as the key treatment unit. Then, several large-scale UF process (e.g., larger than 100,000 m^3/d) were built, and the capacity of UF membranes in full-scale DWTPs increased by 470,000 m^3/d from 2007 to 2010. In the following 7 years (i.e., 2011–2017), both the application of DWTPs using UF membranes increased dramatically, with an increase of 63 and 4,170,000 m^3/d for the number and capacity, respectively. From 2018 to 2020, the application of UF membranes in full-scale DWTPs was much faster, and the capacity increased by 4,747,000 m^3/d in just 3 years. Up to 2020, the number of DWTPs employing UF in China was 106, with a total capacity of about 10 million m^3/d. This capacity is 5.8 % of the total urban water supply production in China [55]. Thus, the application of UF in DWTPs is expected to continue to expand in following years according to the development trend.

As for the geographic distribution (Fig. 2b–d), most of DWTPs using UF membranes are located in Eastern China. Based on the watershed division (Fig. 2b), the biggest capacity (2,861,000 m^3/d) was built in Haihe River Basin, which occupied about 29 % of total capacity, followed by Zhejiang Fujian Region, Yellow River Basin, Perl River Basin, Yangtze River Basin, Huai River Basin, Songhua River Basin, Northwest Rivers and Southwest Rivers, and the last basin is Liaohe River which capacity is 33,000 m^3/d. Based on provinces (Fig. 2c), Shandong Province (1.5 million m^3/d), Zhejiang Province (1.41 million m^3/d), Beijing (1.32 million m^3/d) and Guangdong Province (1.01 million m^3/d) were the top four regions with the largest capacity, while the top provinces (or cities) in number was in the order of Shandong Province (25 plants) ≫ Hebei Province (10) > Jiangsu Province (9) = Heilongjiang Province (9) > Zhejiang Province (8) = Beijing City (8) > Guangdong Province (5) (Fig. 2d).

2.3. Membrane material and membrane module characteristics

With respect to the UF membrane materials used in DWTPs, polymer membranes were widely used with little usage for inorganic membranes

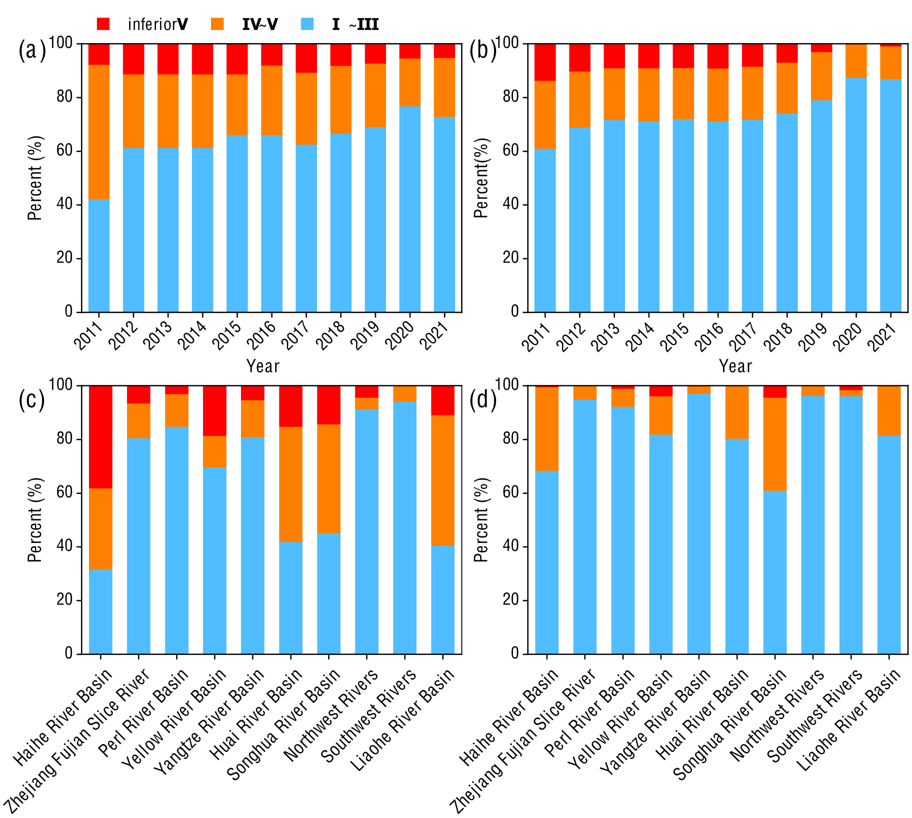

Fig. 1. Water quality based on (a) lakes (reservoirs), (b) rivers, and (c) water resource zones of level I in 2011 and (d) in 2021. Data are collected from references [37–47].

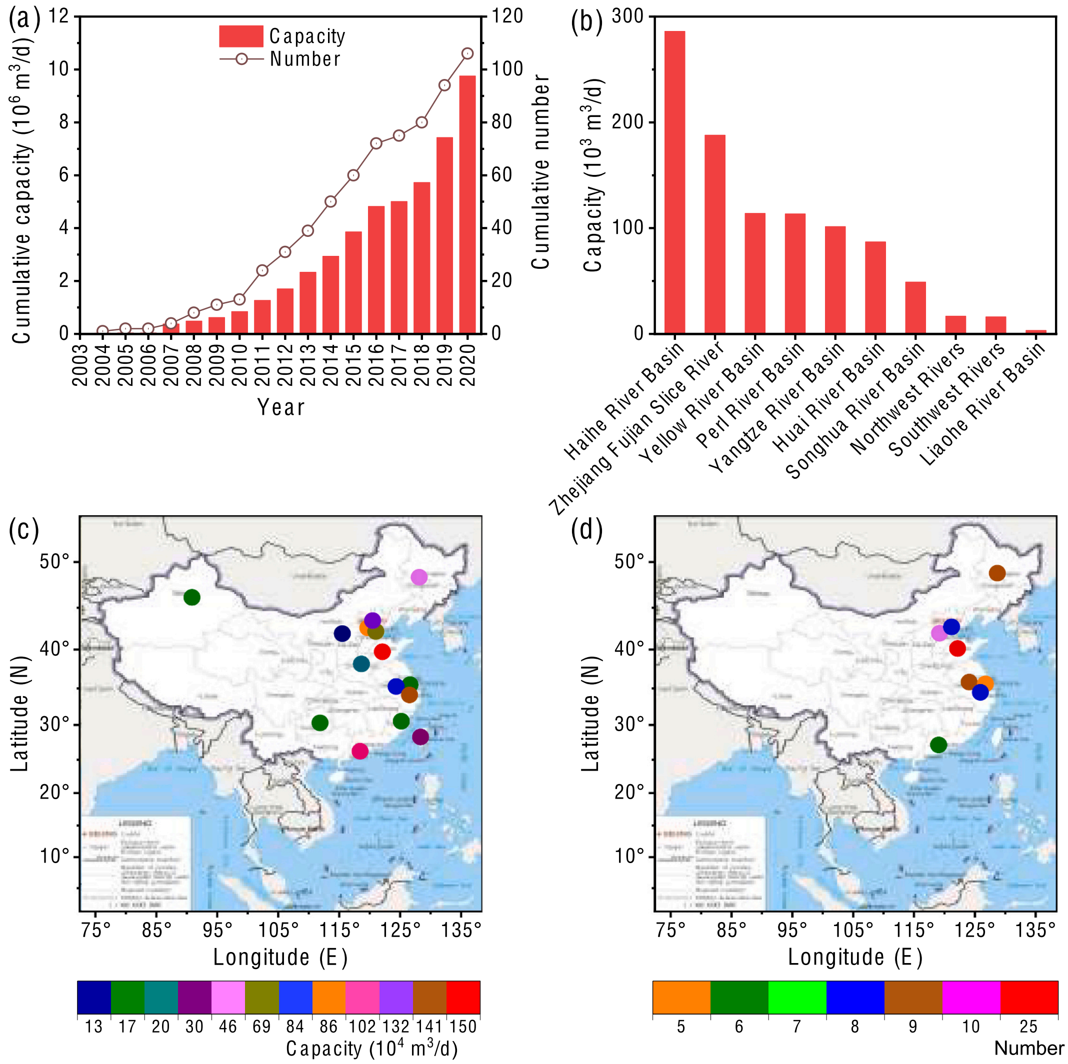

Fig. 2. The capacity and number of DWTPs using UF membranes in China: (a) Cumulative capacity and number, (b) distribution of the capacity based on main river basins, (c) distribution of the capacity based on provinces, and (d) distribution of the number based on provinces. UF membrane system with a capacity of more than 10,000 m^3/d was summarized. The data are summarized in detail in Table S2 (Supporting Information).

(Table S2, Supporting Information). The common organic materials are polyvinyl chloride (PVC), polyvinylidene fluoride (PVDF), polyethersulfone (PES) and polysulfone (PS) [57,58]. Figs. 3a–b illustrate the main UF membrane materials based on the analysis of 106 full-scale DWTPs mentioned in this article. From 2003 to 2007, PVC was the main UF material which occupied 81.58 %. In 2008–2010, both PVC and PVDF were almost equally used, with for the percentage of 46.24 % and 49.46 %, respectively. In the following 7 years, the application of both PVC and PVDF membranes increased significantly, with the capacity of 1.251 and 2.491 million m^3/d, respectively. Finally, in the past 3 years (2018–2020), PVDF became the dominant membrane material with the percentage of 93.26 %, due to its excellent physical and chemical properties [56]. In general, up to 2020, PVDF (73.7 %) and PVC (18.2 %) membranes were the primary UF membranes in full-scale DWTPs in China, followed by PES (4.3 %) and PS (2.1 %).

According to the types of filtration modes, UF membrane can be operated under pressure mode or under submerged mode. Compared with pressure mode, the membranes under submerged modes were built later (Fig. 3c). The first DWTP in China using UF membranes under pressure mode was established in 2004 (Cixi Hangfeng Water Supply Plant), while the submerged one was firstly used in 2008. Up to 2020, the capacity of DWTPs under pressure mode and under submerged mode was 3.85 and 5.90 million m^3/d, respectively (Fig. 3d). Considering the difference in number of DWTPs using UF membranes under pressure mode (n = 60) and under submerged mode (n = 46), the average treatment capacity of both modes was 64,200 and 128,300 m^3/d, respectively. Thus, the submerged UF membrane modules are suitable for large-scale DWTPs, while the pressure mode is more widely employed for medium-scale DWTPs. In the past 3 years (2018–2020), the DWTPs with submerged UF membranes increased faster than the pressure mode, both in treatment capacity and number. To be specific, there were 19 DWTPs using UF membranes under submerged mode (3,267,000 m^3/d) and 12 under pressure mode (1,480,000 m^3/d).

Overall, the capacity of DWTPs using UF membranes reached nearly 10 million m^3/d, about 5.8 % of the total drinking water supply in China. Most of UF membranes were used in DWTPs in Eastern China, and PVDF membrane was the primary UF membrane material. The DWTPs using UF membranes under submerged mode (~60 % in capacity) were more than those under pressure mode.

2.4. *Typical treatment processes in DWTPs*

There are five different typical treatment processes in DWTP using UF membrane as illustrated Fig. 4. To maintain the sustainable operation of UF membranes in full-scale DWTPs, it is necessary to address the problem of membrane fouling [59]. Natural organic matter (NOM) has a

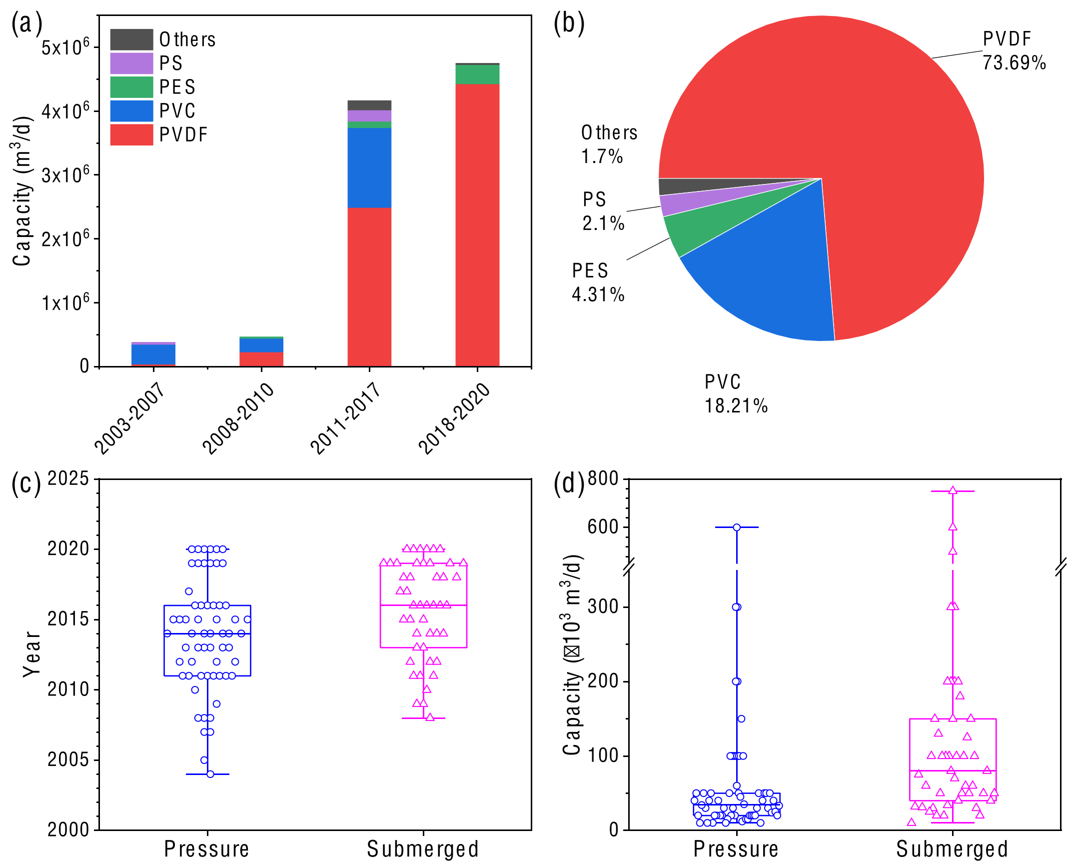

Fig. 3. The characteristics of UF membrane materials and membrane modules in full-scale DWTPs in China: (a) distribution of UF membrane materials as time elapsed, (b) distribution of UF membrane materials in total capacity, (c) temporal distribution of UF membranes under different filtration modes (pressure filtration versus submerged filtration), and (d) the capacity distribution of pressure and submerged membranes. The maximum, the minimum and the median values were displayed with lines in charts c and d. The data are summarized in detail in Table S2 (Supporting Information).

huge influence on the membrane fouling [60]. Coagulation is commonly used as a prior to remove organic substance and small colloids and reduce pore blocking, cake layer resistance, increasing backwash efficiency [61]. Only coagulation was used as pretreatment in the Process 1, and the coagulated water was fed to the UF unit. For example, the coagulation-UF hybrid process (i.e., Process 1) was used in Lujing water supply plant, and the removal of hydrophobic base and hydrophilic matters after coagulation was 27.95 % and 13.33 %, respectively. Process 2 primarily contains coagulation, sedimentation and UF unit, greatly reducing turbidity of the feed for UF unit [62]. Zhaoqing High-tech Zone Water treatment Plant adopts grid flocculation tanks plus inclined pipe sedimentation tank as pretreatment process of UF to treat Beijiang water. Compared to Process 2, a sand filter or biological activated carbon (BAC) filter was added in Process 3. For example, Dongying Nanjiao Water Plants Phase I used the process of folded plate flocculation, sedimentation, sand filter was used as pretreatment of UF unit, the excellent pollutant removal (with the average turbidity below 0.02 NTU and COD_{Mn} below 2 mg/L) ensured the stable operation of UF membrane. The Process 4 consists of conventional coagulation, sand filter or BAC filter, O_3-BAC and UF unit. This process could ensure the turbidity of treated water was less than 0.05 NTU, COD was less than 2 mg/L (Daqing Zhongyin Water Supply Plant). Different with the above four processes, UF membrane could be used as the pretreatment of nanofiltration (NF) or RO, i.e., the Process 5 (UF/NF or UF/RO). The NF or RO system could further remove hardness, total dissolves solids (TDS), dissolved ions, nitrate and some organic pollutants from water. Normally, part of UF permeate goes through NF or RO unit and part goes directly to the clean water tank to mix with the NF or RO permeate to ensure the hardness of the finished water [63], Zhangjiagang 4th DWTP mixes UF permeate water and NF permeate water with the ratio of 1:1 to make the finished water maintain a total hardness of about 116 mg/L. Sometimes, NF/RO permeate mixing with other water with low quality can also achieve this purpose. Changyang No. 3 Water Supply Plant uses this process to treat groundwater with high salt, and the treated water mixed with raw water in the ratio of 1–2 to make the effluent water quality meet the standard.

Fig. 5 shows the distribution of 106 DWTPs using UF based on treatment processes. As presented in Fig. 5, the Process 2, Process 3 and Process 4 were widely used, accounting for 83.13 %. In terms of year distribution, dual membrane system was first to start being applied in 2004 (Cixi Hangfeng Water Supply Plant). The first DWTP using Process 4 was established in 2007. Both Process 1 and Process 2 firstly appeared in Nantong Lujing Water Supply Plant (2009) and Dashuitang Water Supply Plant (2008), respectively. In recent years, the application of Process 1 and Process 5 is on a decreasing trend, and the number of DWTPs using Process 3 and Process 4 has increased more steadily, while Process 2 is showing an expansion trend which indicated a greater adaptability.

For long-term performance of UF membranes, the essential difference in the processes is the use of different pretreatment methods which could alleviate the membrane fouling rate to some extent. Using coagulation before UF not only increased the permeate flux, but also retarded the permeability decline [64]. The addition of BAC or powdered carbon adsorption enhanced the removal of organic matter, extending the flux and filtration duration [34,65]. What's more, O_3-BAC can treat organic matter and algae that are difficult to remove in conventional treatment processes [66]. UF as a pretreatment of NF/RO removes the suspended solids, colloidal and macromolecular organic matter to prolong the

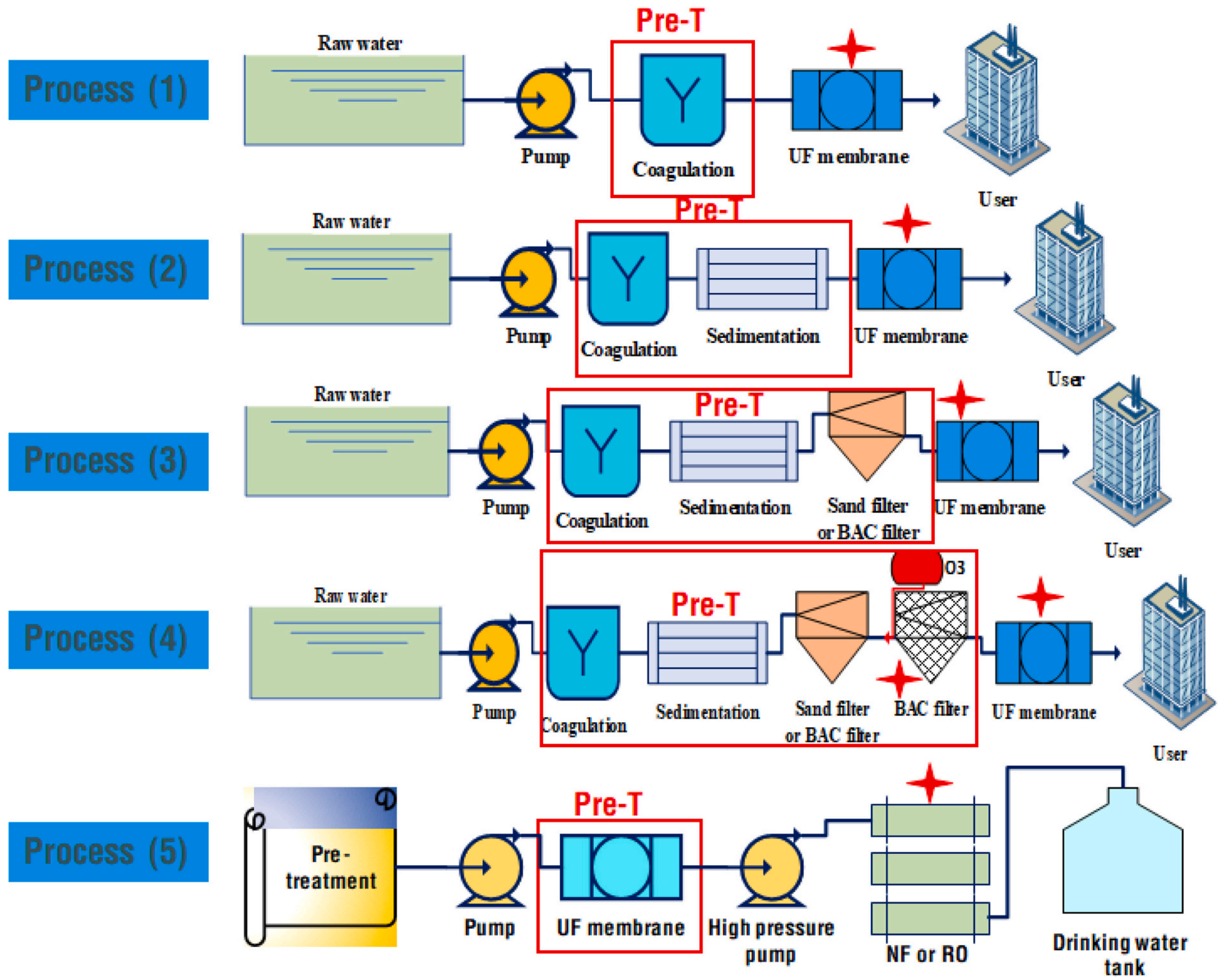

Fig. 4. The typical UF treatment processes in DWTPs in China. BAC, biological activated carbon; NF, nanofiltration; RO, reverse osmosis.

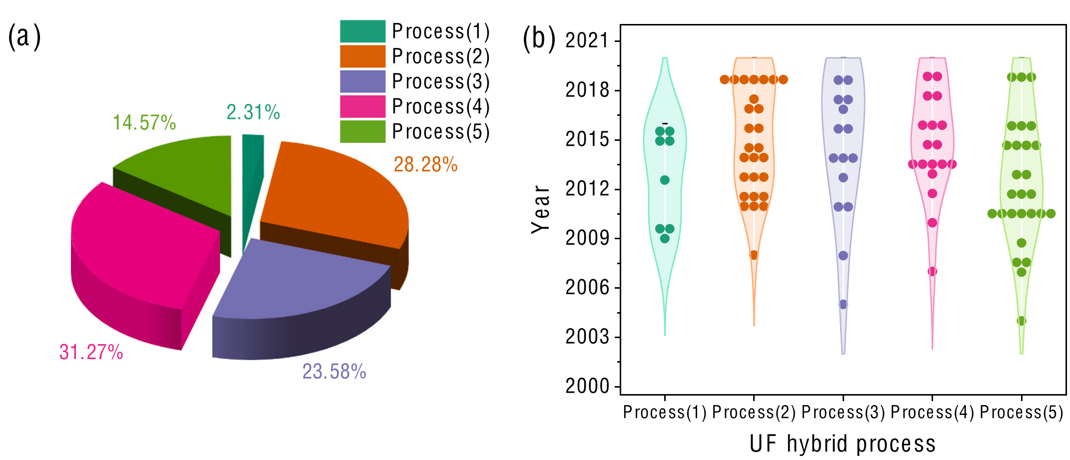

Fig. 5. The comparison of typical UF processes in DWTPs in China: (a) based on treatment capacity, and (b) based on established time. The data are summarized in detail in Table S2 (Supporting Information).

membrane life, increasing the permeate flux and cleaning interval of NF/RO membrane [67]. However, a complex process does not certainly result in decrease in membrane fouling. For example, by comparing coagulation and coagulation + sand filter as UF pretreatment, Xia et al. [68] found that the short process was better for fouling control because the fraction of large-sized floc was rejected by sand filter and small-sized floc embedded in membrane pores increased membrane fouling. Thus, each process has its merits and applicable condition, and the choice of drinking water treatment process depends on many factors. According to Standard for drinking water quality (GB5749-2006) [53], turbidity, COD_{Mn} and ammonia (calculated by N) was less than 1 NTU, 3 mg/L and 0.5 mg/L, respectively. Typical DWTPs in China were selected to analyze the characteristics of UF permeate, and Fig. 6 presents the water purification in typical UF treatment processes in full-scale DWTPs in China.

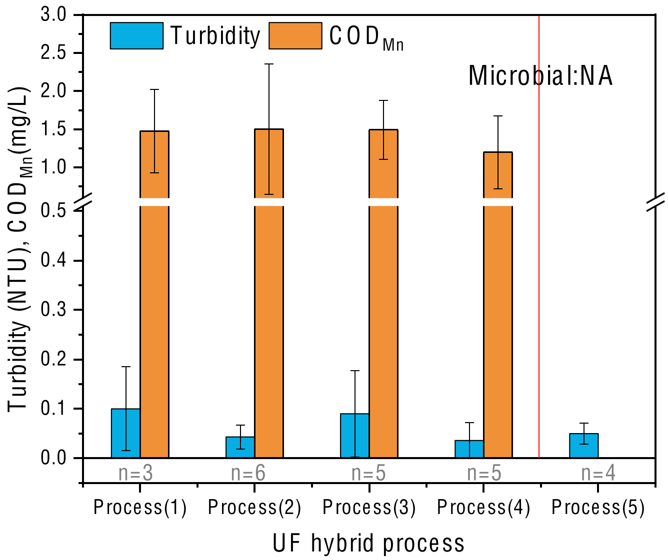

Fig. 6. Characteristics of UF permeate quality in typical UF processes in full-scale DWTPs in China. The data are summarized in detail in Table S4 (Supporting Information).

The typical processes (Process 1–Process 4) did not differ significantly in the removal of pollutants, with the average turbidity usually less than 0.1 NTU and COD_{Mn} less than 2.0 mg/L. Moreover, ammonia concentration in the finished water was usually less than 0.3 mg/L, and no microbial indexes were detected. Thus, excellent water quality of UF permeate of all processes in full-scale DWTPs was observed.

Overall, there were five typical drinking water treatment processes involving UF membrane. The combination of UF membrane with coagulation, sedimentation (and sand/BAC filter) (i.e., Process 4, Process 2 and Process 3) were the commonly used treatment process, and Process 2 may have a wider application in the future as it shared the largest percentage in the newly installed DWTPs (in 2019 and 2020). Anyway, all these processes show good contaminant removal performance, fully satisfying Standard for drinking water quality (GB 5749-2006) [53], and the selection of the UF hybrid process should be designed according to the characteristics of the raw water quality.

3. UF filtration and cleaning performance in full-scale DWTPs in China

UF operation conditions including membrane filtration and membrane cleaning parameters have influences on the performance of UF membrane in full-scale DWTPs. Filtration flux, filtration length, backwashing flux, backwashing length and cleaning method were key parameters in UF operation [13].

3.1. Membrane filtration performance

Fig. 7 summarizes the key operation parameters of UF membranes in full-scale DWPTs in China, focusing on filtration modes. As for filtration flux, the UF fluxes under pressure mode were in the range of 37.5 to 115 L/(m^2·h) (LMH) with the median value of 70 LMH, which was 2.33 times of the median flux of UF membranes under submerged mode (Fig. 7a).

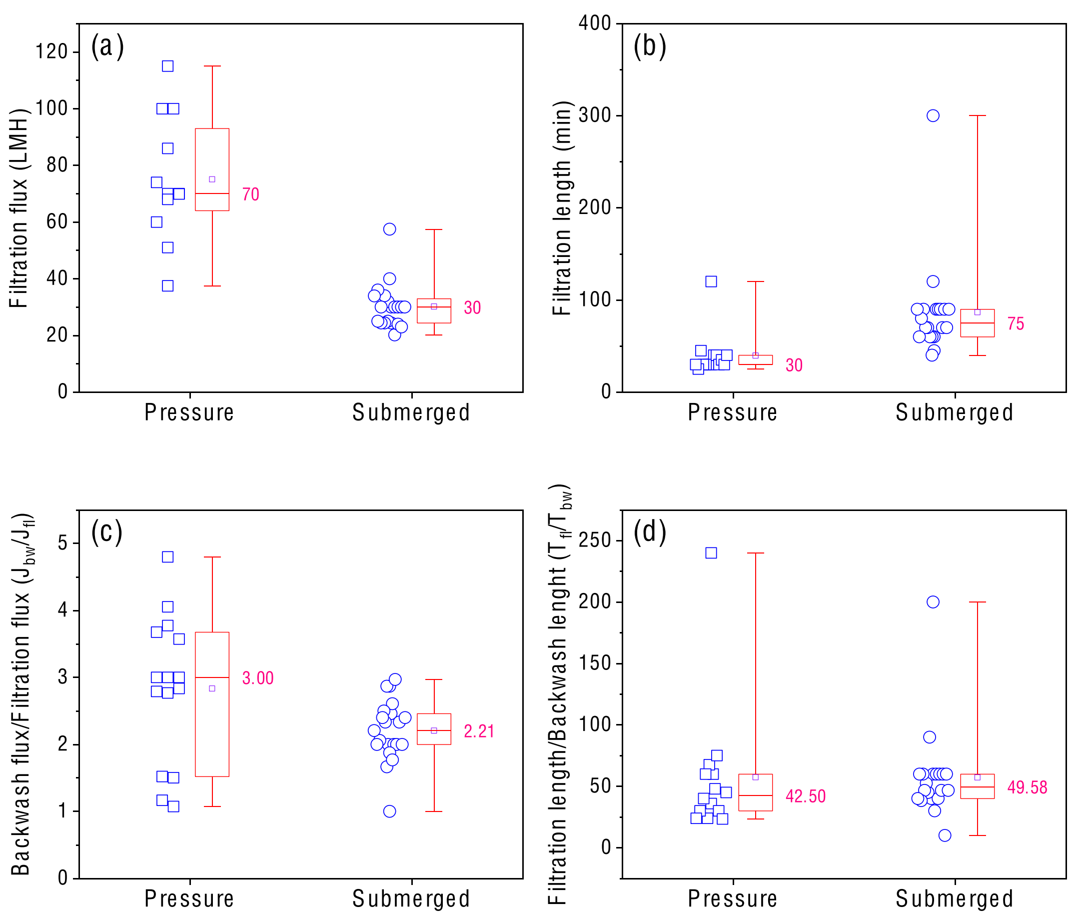

Fig. 7. Comparison of UF flux and filtration length during filtration and backwash in different operation modes: (a) filtration flux, (b) filtration length, (c) ratio of backwash flux and filtration flux, and (d) ratio of filtration length and backwash length. (Number of DWTPs with UF membranes under pressure mode, $n = 14$; under submerged mode, $n = 30$). The data are summarized in detail in Table S5 (Supporting Information).

As presented in Fig. 7b, the median filtration length of UF membranes under pressure mode (30 min) was just 40 % of that under submerged mode (75 min). With respect to the hydraulic backwashing, the median ratio of backwash flux and filtration flux for UF membrane under pressure mode (1–4.8) was 3 which was slightly larger than the submerged membrane (1.1–3.0, with median value of 2.21) (Fig. 7c). These values in full-scale DWTPs were less changed than the bench- or pilot-scale studies reported in published literature [13]. As shown in Fig. 7d, the ratios of filtration length and backwash length under pressure modes were slightly less than that under submerged mode. This indicated that pressure membrane consumed more energy when the UF membrane was cleaned. Although high filtration flux can reduce the membrane area and save space and investment, it tends to increase the membrane fouling and thus increase the operation costs of membrane cleaning.

3.2. Membrane foulants and membrane cleaning

During the long-term operation of UF membranes in full-scale DWTPs, different types of foulants could be observed on membrane surface or in membrane pores, including organics, inorganic matters and biological substances [64]. To be specific, organic matters from water were consisted of aromatic protein, soluble microbial by-product (SMP) and humic substance [36]. In Lujing water supply plant (Nantong), the organic foulants on the membrane were analyzed through the desorption solutions and it was composed of protein/protein-like substances and SMP-like and humic substances, with the proportions of 79.98 %, 20 % and 3 %, respectively. When groundwater was used as raw water in a DWTP [31], membrane's fouling-free layer after five-year operation and it consisted of natural organic matter (biopolymers such as proteins, polysaccharides, aminosugars and polyhydroxyaromatic structures) coming from the water resources as well as inorganic substances (Al, Fe, Si and Ca), flocculent polymer (AN905) and activated carbon from the pretreatment. In the study of Dongying DWTP, the fouling layer contained proteins (organics) and inorganic substances (Al, Si, Mn, Fe, Ca and Mg) [35]. During the 7-year period of operation from 2009 to 2016, there was a significant change in membrane performance, as shown in Fig. 8. Biological fouling in cake layer was mainly composed of a large number of inorganic materials, algae and their extracellular organic matters (EOMs), as well as microorganisms with Proteobacteria and Firmicutes as the main organisms, inorganic substance elements including Na, Mg, Al, Si, Ca, Mn and Fe, and organics mainly hydrophilic organic matter and dissolved biological byproduct-like organic substances [28].

Common physical cleaning includes hydraulic cleaning, mechanical cleaning and ultrasonic cleaning [69], and it removes the reversible foulants within membrane pores and surface. In comparison, chemical cleaning was used to remove irreversible foulants that has absorbed onto the membrane and cannot be removed hydraulically after repeated backwash cycles. Thus, both physical and chemical cleaning methods are necessary for membrane cleaning. In Cixi Hangfeng water plant, backwash was performed every 0.5 h for 1 min with backwash flux of 300 to 400 LMH, and chemical cleaning was carried out once a month, using a combination of acid and alkali cleaning. NaClO and NaOH were used firstly, followed by citric acid, of which the concentration of NaClO was 50–100 mg/L and the duration of cleaning was 4–6 h. What's more, air-assistance is usually carried out to enhance the efficiency of hydraulic backwashing, especially for the cases of submerged UF membranes, i.e., the so-called air-assisted backwash. Guigui et al. [70] improved the removal of colloidal by 30–130 % by air-assisted backwash when compared with pure hydraulic cleaning, verifying a feasible method to reduce backwash time. Table S3 (Supporting Information) summarizes the application of aeration for UF membranes in full-scale DWTPs in China. When the aeration strength was calculated by UF membrane area, the values were 80–166 L/(m^2·h), and the aeration strength ranged from 50 to 90 m^3/(m^2·h) when it was calculated by membrane tank area. The design of aeration strength calculating by membrane tank area referred to that of a sand filter for which a aeration strength of 60 m^3/(m^2·h) (calculated by membrane tank area) was commonly used. As to aeration duration, a series of values for 30–150 s

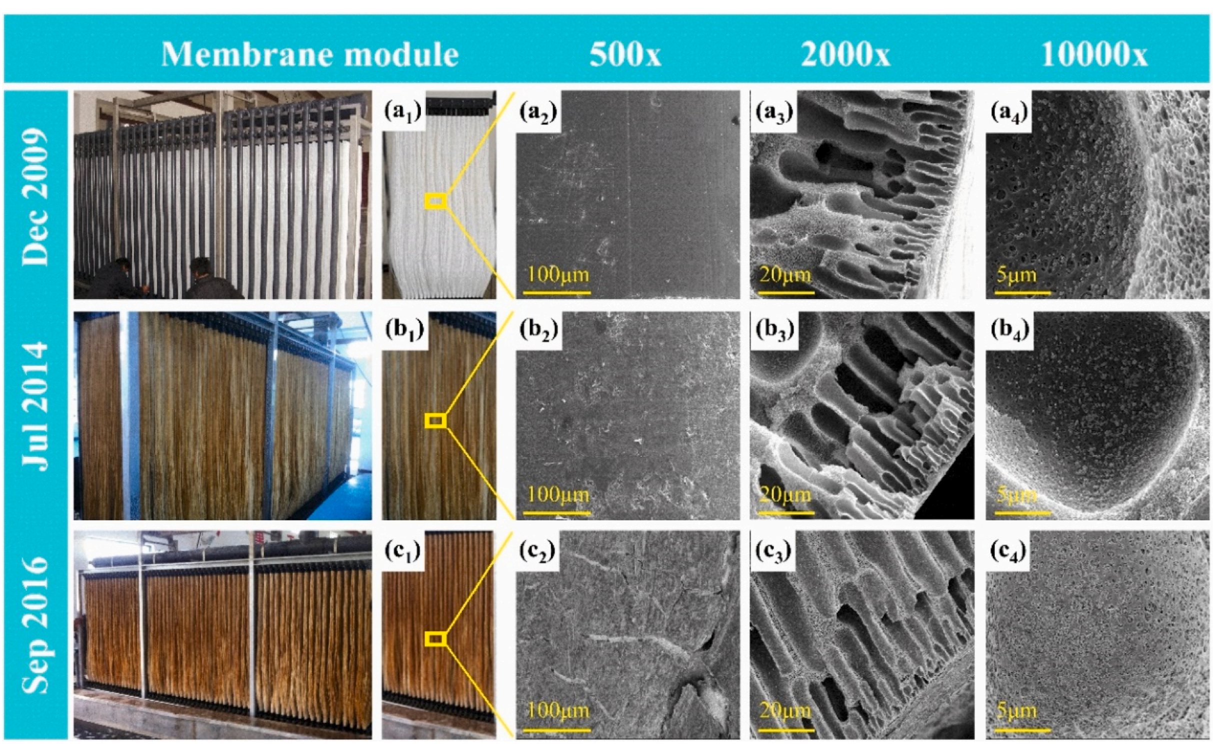

Fig. 8. Visual inspection and surface properties by scanning electron microscope analysis of the UF membrane fibers which were obtained in (a) December 2009 (the time of installation of the UF system), (b) July 2014 (during maintenance service) and (c) September 2016 (during maintenance service).
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could be observed for the full-scale application of UF membranes. For chemical cleaning, it is helpful to remove foulants from membrane after several operation cycles. The cleaning using chemical agents can be divided into maintenance cleaning (MC, or chemical enhanced backwash) and chemical recovery cleaning (CC). The purpose of maintenance cleaning is to slow down the deep fouling of UF membrane so as to extend the chemical recovery cleaning interval. Table S6 (Supporting Information) summarizes the part of chemical cleaning strategies of UF membrane in DWTPs in China, and the key parameters including cleaning interval, cleaning duration and NaClO concentration are illustrated in Fig. 9 when NaClO was used in maintenance cleaning. As presented in Fig. 9, the maintenance cleaning interval was at the range from 1 to 120 days, and the time was under 30 days for most DWTPs. For the cleaning duration, the shortest cleaning time was 15 min (e.g., Binhai Water Supply Plant) and the longest one was 24 h (e.g., Nanjiao Phase II Water Supply Plant and Qingtai Water Supply Plant). In the choice of cleaning agents, the general use was the combined acid and alkali cleaning. To be specific, the commonly used alkaline cleaning agents were NaClO and NaOH, while acidic cleaning agents are HCl and citric acid (Table S6, Supporting Information). The concentration of NaClO for maintenance cleaning was from 100 to 490 mg/L (Fig. 9). Chemical recovery cleaning removes the pollutant which cannot be removed by hydraulic cleaning and maintenance cleaning. As listed in Table S6 (Supporting Information), alkaline cleaning and acidic cleaning in chemical recovery cleaning were similar with maintenance cleaning, but a higher NaClO concentration was used in chemical recovery cleaning (e.g., a common NaClO concentration of 1000 mg/L) (e.g., Xujing, Hongyanchi, Qingtai, Beijing 309, Sanhe, Luoqiao and Zhaoqing High-tech Zone). The interval of chemical recovery cleaning was much longer than maintenance cleaning, generally from several months to 2 years, and the duration of chemical recovery cleaning ranged from 2 to 16 h.

Membrane cleaning is an important operation parameter to maintain the long-term operation of UF membrane, while cleaning efficiency may decrease as time elapsed, and inappropriate cleaning methods would lead to severe membrane fouling. Taking Dongying Nanjiao DWTP as an example, severe membrane fouling (i.e., transmembrane pressure (TMP) or resistance) occurred, although the interval of hydraulic cleaning (i.e., air-assisted backwash) decreased from 5–6 to 1.5–2 h in 7-year operation (Fig. 10a). Thus, frequent chemical cleaning was employed, and the total water treatment cost in the UF system increased by 55 % (i.e., from 0.24 to 0.37 CNY/m^3) [28]. The authors determined the key membrane fouling factors to be operation time and hydraulic cleaning via multiple

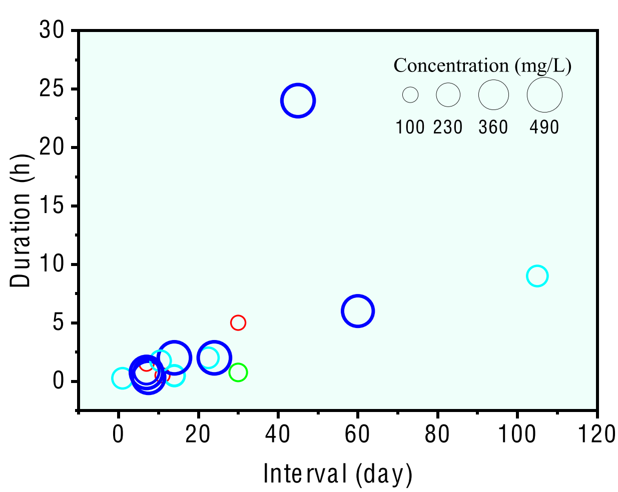

Fig. 9. Summary of key parameters for maintenance cleaning for UF membranes in full-scale DWTPs in terms of cleaning interval (day), cleaning duration (h) and NaClO concentration (mg/L). The data are summarized in detail in Table S6 (Supporting Information).

statistical approaches. As presented in Fig. 10b, the use of BAC filter increased the UF flux by 18.5 % and extended the filtration run by 45 %, while there was no increase in TMP even with less frequent membrane cleaning in the presence of BAC [34]. The results demonstrated the importance of selecting appropriate pretreatment and cleaning methods. In a hybrid short-length sedimentation/UF process (i.e., Process 2) (Dongying Nanjiao II DWTP), it was vital important for periodic hydraulic cleaning (1.5–2 h) to ensure the long-term stable operation of UF membranes [35]. However, the hydraulic cleaning efficiencies decreased as time elapsed, so frequent maintenance cleaning (14–24 days) and chemical recovery cleaning were required. Moreover, membrane material also took a great role in cleaning efficiency, and higher cleaning efficiencies (by 10 %–23.9 %) were obtained for PVC composite membranes (Fig. 10c). Thus, optimization of the combination of different cleaning methods is helpful to ensure the stability of UF performance in large-scale DWTPs.

3.3. *Membrane lifetime analysis*

Due to the characteristics of membrane cleaning, membrane lifetime can be evaluated in different ways, such as the change of resistance of membrane, the increasing of fouling rate, the decreasing of cleaning efficiency and the appearance of membrane leakage or infrastructure deterioration [32]. Resistance could be considered as a membrane's impedance to fluid flow, and the increase of resistance indicated the membranes age due to the materials leading to irreversible fouling. Fouling rate is usually linked with changing physical and chemical characteristics of the membranes, and it increased for aged membranes, thus when fouling rate reached a certain value meant the end of membrane lifetime. There is a correlation between age and membrane leakage because higher TMP is likely to occur later in membrane life. Cleaning rate decreased as membrane aged, and it is difficult to restore the membrane performance at the end of membrane lifetime. Infrastructure was important to support membrane [30], and the deterioration of infrastructure led to replacement of membrane, regardless of the condition of membrane fiber. Fig. 11 shows the operation time and lifetime of UF membranes in full-scale DWTPs in China. Due to the rapid increase in the development of UF membranes in China within the last 10 years, many DWTPs were established or upgraded in less than 10 (or even 5 years). As presented in Fig. 11, about half of the DWTPs using UF membranes were operated for less than 4 years, and there were 8.77 % and 12.31 % of DWTPs for operation time of 5 and over 10 years, respectively. With replacement of the membranes, only 7.58 % of DWTPs had a full replacement cycle, and it is a challenge to make an accurate determination of membrane lifetime. As shown in Fig. 11, the shortest membrane life was 5 years for the part of DWTPs with membrane replacement, and the DWTPs with a 7-year membrane life accounted for 37 % of total replaced capacity, with the longest membrane life above 10 years (13.79 %). A longer membrane life can reduce the frequency of membrane replacement and thus reduces investment.

4. Challenges and perspectives

In recent years, the number of DWTP using UF membrane has been growing at a faster rate. Although great efforts have been made for the application of long-term operation of UF membranes in full-scale DWTPs, there are some challenges to be addressed for the sustainable application of UF membranes shown in Table 1. As a summary, the main problem is the maintenance of UF membrane during the application due to its susceptibility to fouling, which has a risk of substandard UF permeate. Selection of appropriate treatment process, cleaning methods and cleaning agents can make a contribute to reduce the possibility, and the more experience can support more precise control. Thus, it remains important for timely summary and analysis of long-term DWTPs with UF membranes operations and maintenance. Evaluating the evolution of UF performance in full-scale DWTPs during long-term operation would

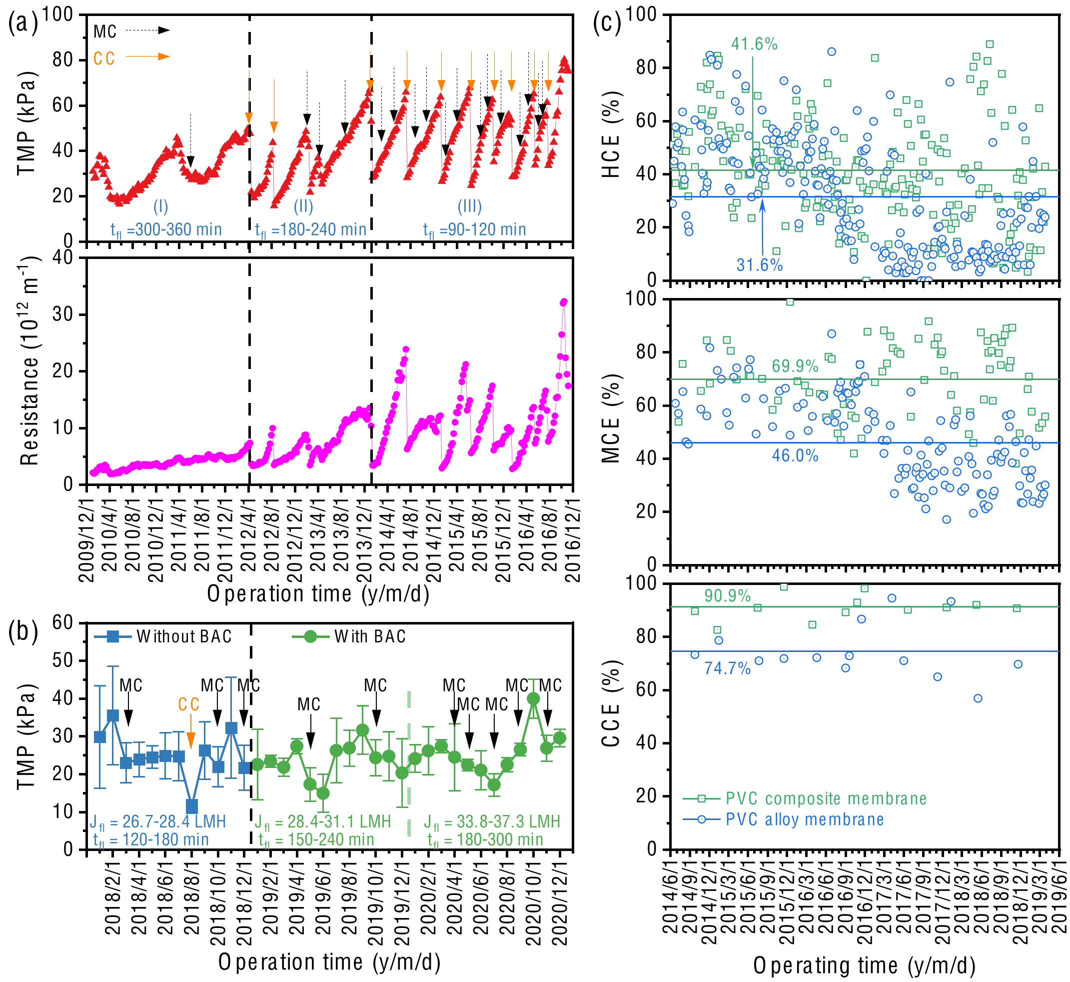

Fig. 10. Variation of the UF membrane performance over time during long-term operation: (a) transmembrane pressure (TMP) and resistance during 7-year operation, (b) membrane performance with and without BAC during 3-year operation (average TMP value is shown for each month), and (c) cleaning performance during the 5-year operation using PVC composite and PVC alloy membranes. Note: MC, maintenance cleaning; CC, chemical recovery cleaning; J_{fl}, filtration flux; t_{fl}, filtration length; HCE, hydraulic cleaning efficiency; MCE, maintenance cleaning efficiency; CCE, chemical recovery cleaning efficiency.
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provide guide for sustainable and rapid development of UF technology.

5. Conclusions

In this study, the application of DWTPs using UF membrane in China was systematically investigated. The characteristics including installed capacity, location, membrane materials and operation mode of UF membranes, and the diagnostic of membrane performance were analyzed.

The total capacity of the full-scale DWTPs using UF membranes has reached ~10 million m^3/d by 2020, 5.8 % of the total urban water supply production in China. Most of UF membranes were used in DWTPs in Eastern China, and PVDF membrane was the primary UF membrane material. The DWTPs using UF membranes under submerged mode (~60 %) were more than those under pressure mode.

Five typical water treatment processes involving UF membranes were employed in full-scale DWTPs, and the hybrid process of coagulation/sedimentation (and sand filtration or/and BAC filtration) were usually used coupling with UF membrane. All the hybrid UF processes could meet the requirement of drinking water quality, and the selection of suitable treatment process greatly depended on feed water quality.

The median filtration flux of UF membranes under pressure mode (70 LMH) were 2.33 times of that under submerged mode (30 LMH), while the median filtration length of UF membranes under pressure mode (30 min) was just 40 % of that under submerged mode (75 min). Similar backwash strength in terms of the ratio of backwash flux and filtration flux under both filtration modes were used.

The primary membrane foulants varied with elapsed time and were different with other DWTPs. The physical cleaning efficiency of (air-assisted) hydraulic backwashing would decrease with operation time, and it was necessary to employ maintenance or chemical cleaning to achieve the sustainable operation of UF membranes in long-term operation.

Half of the UF membranes in DWTPs have been operated for less than 4 years by 2020, only 1/8 of the membranes were operated for more than 10 years. Of the replaced UF membranes, more than 1/3 were replaced after operating for 7 years. As the operation time of most DWTPs was too short to reach the membrane replacement time, it is necessary to assess the membrane lifetime during long-term operation.

Moreover, the percent of DWTPs using UF membrane for urban water supply production in China is still low, and it is expected for rapid development in coming years. The evaluation of UF performance in full-scale DWTPs during long-term operation would provide guide for sustainable and rapid development of UF technology.

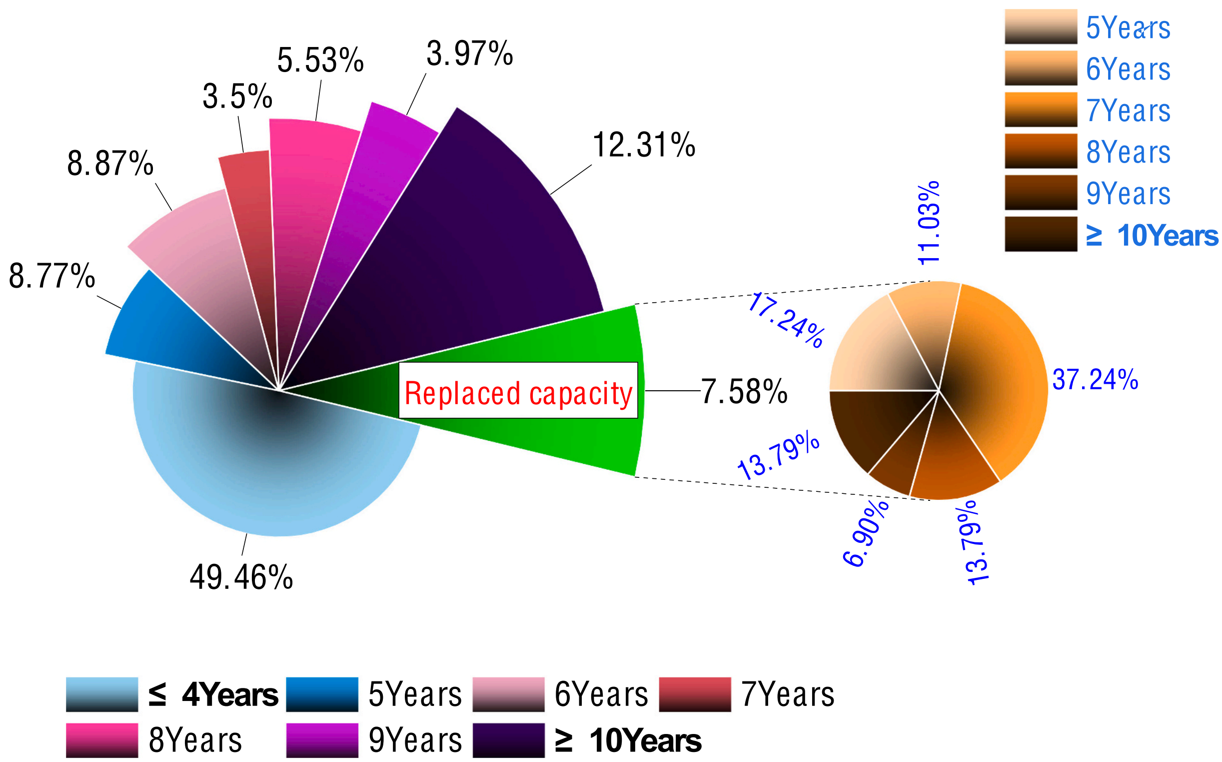

Fig. 11. Operation time of UF membranes in full-scale DWTPs and the replaced capability. The data are collected according to survey.

Table 1
Challenges and perspectives of UF membrane application.

Challenges	Perspectives
(1) Biological safety of UF permeate	➢ Monitoring variation of permeate quality ➢ Development of appropriate water treatment process
(2) Evolution of membrane fouling behaviors	➢ Evaluation of long-term operation of UF membranes ➢ Analysis of membrane foulant seasonally or annually
(3) Decrease in membrane cleaning efficiency	➢ Optimization of air-assisted hydraulic back-washing process ➢ Using appropriate agents of chemical cleaning for specific foulants
(4) Membrane ageing/ lifetime	➢ Establishment of membrane ageing evaluation methods ➢ Overcoming the tradeoff between membrane replacement and energy consumption due to fouling

CRediT authorship contribution statement

Haiqing Chang: Investigation, Methodology, Data curation, Formal analysis, Software, Project administration, Writing- original draft and editing.
Yingyuan Zhu: Software and Writing- original draft.
Haikuan Yu: Investigation, Data curation, Software and Writing-reviewing and editing.
Fangshu Qu: Writing-reviewing and editing.
Zhiwei Zhou: Writing-reviewing and editing.
Xing Li: Resources, Supervision, Writing-reviewing and editing.
Yanling Yang: Writing-reviewing and editing.
Xiaobin Tang: Writing-reviewing and editing.
Heng Liang: Resources, Supervision, Writing-reviewing and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.desal.2022.116122.

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