

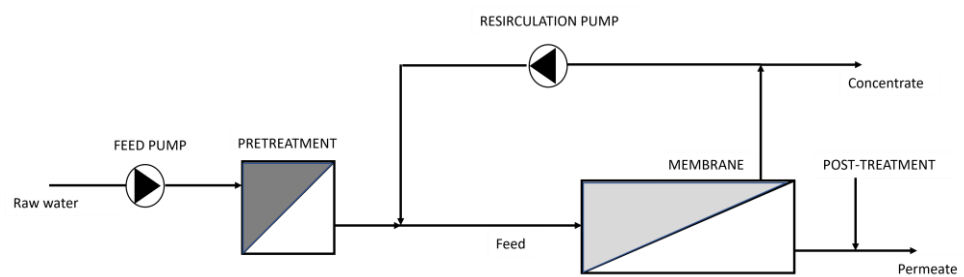
Report

Membrane filtration of coloured surface water

A Norwegian survey

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ABSTRACT

Today around 160 small and medium-sized waterworks in Norway are using membranes for treatment of raw water with high content of natural organic material (NOM). Most of the membrane-based waterworks use nanofiltration (NF) as treatment process. The first NF facilities were commissioned late 1980s, and it is anticipated that significant experience and knowledge on operation of membrane facilities exists. A survey has been conducted with the objective of mapping the status regarding design and operation of membrane facilities. The key findings from 18 plants answering the survey demonstrate that membranes provide efficient NOM removal, and that most plants operate with few reported challenges. However, some facilities suffer from performance decline due to fouling. Nevertheless, given optimal process design and operation, the use of membranes is considered well suited for treatment of coloured surface water for drinking water production.

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1 Introduction

Membranes are to an increasing extent used for drinking water treatment worldwide. In Norway membranes are primarily used for the treatment of raw waters with high content of natural organic matter (NOM). Sweden, Canada, Ireland, Scotland, Southern Germany and South Australia have similar requirements [1], and recent research shows a growing interest in using nanofiltration and ultrafiltration for NOM removal [2-9]. Nanofiltration has also experienced a growing interest as a treatment method for retrofitting existing facilities for enhanced NOM removal [10-12].

The first Norwegian waterworks using membranes as a treatment process were commissioned in the late 1980s. The use of membranes quickly became popular for small and medium-sized waterworks, and today, around 160 facilities using membranes are registered in Norway [13]. About 30% of the membrane facilities have a reported production smaller than 100 m³/d, 55% in between 100 and 1000 m³/d, and approximately 15% of the plants reports a production above 1000 m³/d. Only a few facilities have a production capacity above 5000 m³/d. Until recently nanofiltration was the only applied membrane process, however, ultrafiltration has become increasingly popular. Due to the large number of membrane facilities and the fact that several plants have been in operation for more than 30 years, significant knowledge and operating experience have been gained. However, existing knowledge is not compiled, and is mainly found locally at each waterworks. Thus, to fully utilize this knowledge, there is a need for collecting and systematizing existing operating experience and making this knowledge available to waterworks and operators around the country.

Relatively few literature references exist on operating experiences of membrane facilities for drinking water treatment in Norway. Most of the existing literature have been published by Thor Thorsen, who was central in the development of the nanofiltration process for drinking water treatment during the 1980s and 90s [14,15]. The "Norwegian" nanofiltration process is also presented in an international text book on nanofiltration applications [1]. In 2008, a survey was conducted on experiences of using nanofiltration systems as a hygienic barrier [16]. The study also addressed operational experiences and challenges related to fouling. The work was carried out as part of the EU project TECHNEAU and showed that 27% of the 37 facilities that responded to the survey faced problems with one or more barrier failures in the period 2001-2006. Furthermore, the production decline was linked to fouling [17]. The mentioned publications also formed the basis for a Norwegian guideline for nanofiltration plants [18].

Since most of the "Norwegian" literature on water treatment by membranes are more than 10 years old, a new survey of Norwegian waterworks was conducted in 2018 with the objective of mapping the status on design and operation of membrane facilities, including cleaning protocols and raw water quality. Furthermore, the waterworks were requested to report operational challenges of any kind. This paper presents the main findings from the survey.

2 Methods

During autumn 2017, a questionnaire was prepared and distributed by email to waterworks applying membranes as the main treatment process [13]. The email was sent to the waterworks' (municipality's) post office with a request to forward the information to the technical manager, VA manager or operations manager for the waterwork. An email with the survey and cover letter attached was sent the first time on 2017-10-24, the second time on 2017-11-14 and the third and last time on 2017-12-05. The last time, the response deadline was also extended by a few weeks. A total of 117 questionnaires were sent out and 18 responses were received. This corresponds to a response rate of 15.4%, which is considered disappointingly low. Questionnaires were only distributed to the facilities reporting production data to the National Water Facility Register during the last couple of years. It was assumed that the registered facilities (168) that didn't perform recent reporting to the register was no longer in operation. However, it was later realized this assumption was incorrect, and that most of the registered plants not performing recent reporting were still in operation. Thus, the number of operating membrane facilities for drinking water production is assumed to be closer to the registered number (168) than the number of facilities performing recent reporting (117). Several waterworks were contacted by telephone as an additional measure to increase the response rate, however, without success. It can be noted that the feedbacks from e-mail correspondence and telephone contact were mostly positive, however, in practice it was challenging for the waterworks to allocate time responding to the survey.

The questionnaire consisted of two parts, a simplified part with general information, and additionally a more comprehensive part. Part 1 of the survey consisted of 16 questions, while part 2 consisted of 43 questions. This gives a total of 59 questions for the entire survey, sorted into subcategories such as "water quality", "process design" and "operation". It was also possible to submit attachments to the survey, *e.g.*, analysis results of raw water quality or product sheets from the membrane manufacturers.

The waterworks that replied to the survey were randomly assigned an ID number (from 1-18).

3 Results and Discussion

The results from the survey are reported and discussed under the following sub-headings.

3.1 Process design

Nanofiltration (NF) is by far the dominating treatment processes in Norwegian waterworks. However, over the last years both ultrafiltration (UF) and microfiltration (MF) have been selected for new facilities. What distinguishes the various membrane processes is primarily the membrane characteristics. Due to very small pores, in the order of 2-5 nm, nanofiltration membranes have excellent retention of dissolved material in the raw water. Ultrafiltration membranes have significantly larger pores (typically 10-100 nm), and will therefore have higher permeability, but also poorer retention of dissolved compounds in the raw water. MF membranes have even larger pores than UF membranes, typically > 100 nm. Thus, to increase retention of dissolved organic material (NOM) by UF and MF membranes to desired levels it will be necessary to apply coagulation as pre-treatment upstream the membrane stage.

Although NF and UF facilities use membranes as the main treatment stage, both the process design and the operating strategy are significantly different. A typical process design for nanofiltration facilities is shown in Figure 1 and consists of a feed pump, particle removal (pre-treatment), NF membranes and post-treatment. The retentate stream is split into concentrate bleed and a recycle stream that is returned to the feed side of the NF unit by means of a recycle pump. A simple pre-treatment and a high degree of recycling are special for Norwegian nanofiltration facilities and are sometimes referred to as the "Norwegian process for color removal". High recycling rate ensures high crossflow velocity through the membrane elements, which is beneficial to avoid fouling deposits on the membrane. This is related to different forces that are acting on the NOM particles within the boundary layer next to the membrane surface. In the boundary layer the flux will cause a drag on the NOM particles towards the membrane surface, whereas the crossflow velocity will cause diffusion of particles from the membrane surface due to inertial lift diffusion and shear force diffusion. Both mechanisms depend on the hydrodynamic diameter of the particles and the crossflow velocity. By balancing the flux and the crossflow velocity, one can avoid that accumulation of a critical amount of NOM materials on the membrane surface and, hence, irreversible fouling. A more thorough discussion about the NOM fouling mechanism can be found elsewhere [15]. The pre-treatment generally consists of simple filtration through a pressure sieve or cartridge filters with a pore opening in the range of 50 μm . The post-treatment usually consists of an extra hygienic barrier and pH adjustment/alkalisation.

The NF membrane systems included in this review used 8 inch spiral elements of 1.0 or 1.5 m length, assembled in 6 m long pressure vessels. A desired number of pressure vessels is installed in parallel to form a unit or section. Recently a nanofiltration membrane using hollow fibre configuration has been launched, however, this type has so far not been applied at a larger scale in Norway (status as of January 2020).

The operating strategy of nanofiltration plants typically involves continuous operation at stable production, however, interrupted by approximately one hour of downtime per day to carry out a membrane cleaning. The daily cleaning solution is often a proprietary product provided by the system supplier, frequently in combination with sodium hypochlorite. A standard cleaning sequence typically comprise the following steps, 1) Continuous circulation of cleaning solution without transmembrane pressure, 2) soaking of cleaning solution without circulation, and 3) flushing with permeate to displace the cleaning solution. Typically, the initial permeate production after membrane cleaning is discharged to avoid that cleaning chemicals or redissolved deposits can contaminate the permeate. Alternatively, short periods of pressure relaxation (no production or circulation) are used in combination to the generic procedure described above. Short periods with no flux will

cause a transport of particle away from the membrane surface due to Brownian diffusion which is caused by the concentration polarization within the boundary layer, hence, decreasing the pressure will decrease the NOM load on the membrane [15].

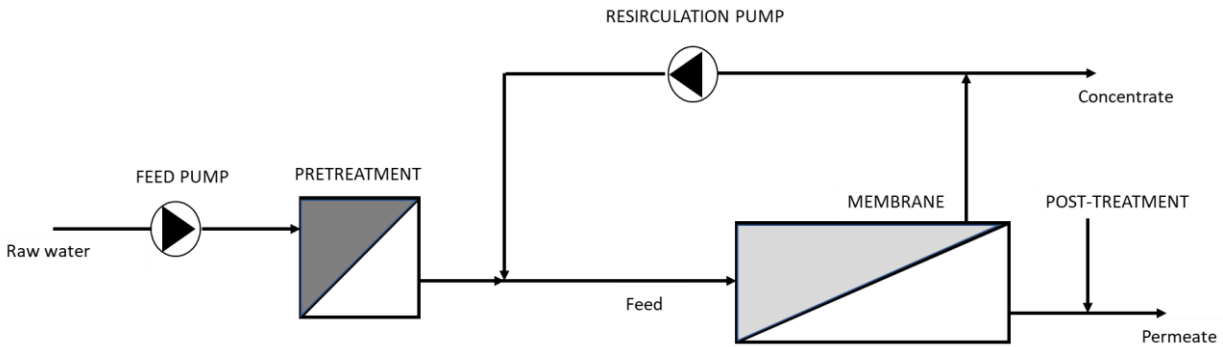


Figure 1. Typical process design of Norwegian waterworks using NF.

Figure 2 shows a typical process design for ultrafiltration facilities including, pre-treatment, pH adjustment, addition of coagulant, membrane filtration and post-treatment. The pre-treatment ensures coarse particle separation by use of pressure sieves or cartridge filters with a light opening in the range 50 - 300 μm , depending on the raw water characteristics. Furthermore, coagulant is added upstream the membrane to form flocs of sufficient size to be retained by the pores of the ultrafiltration membrane. pH adjustment is usually necessary to optimize the conditions for coagulation and formation of stable flocs. Both pH and coagulant dose should be optimized for each facility according to the raw water characteristics. The final treatment typically consists of an extra hygienic barrier (chlorination or UV), and possibly further pH adjustment for corrosion control.

UF membranes for application in drinking water treatment are hollow fiber membranes operated in "dead-end" configuration, *i.e.*, all the water entering the membrane on the feed side is forced through the membrane to the permeate side, while the particles larger than the pore size are retained by the membrane, forming a layer of deposits on the membrane surface. The formed deposits will increase the resistance to water transport through the membrane and must be removed frequently. This is done by backwashing the membrane for short periods by pumping clean water (usually permeate) in the reverse direction of the permeate flow. The filtration time between two backwashes is typically in the order of 1 hour. After a certain number of filtration and backwash sequences, a chemically enhanced backwash (CEB) is performed. The CEB frequency is typically once a day.

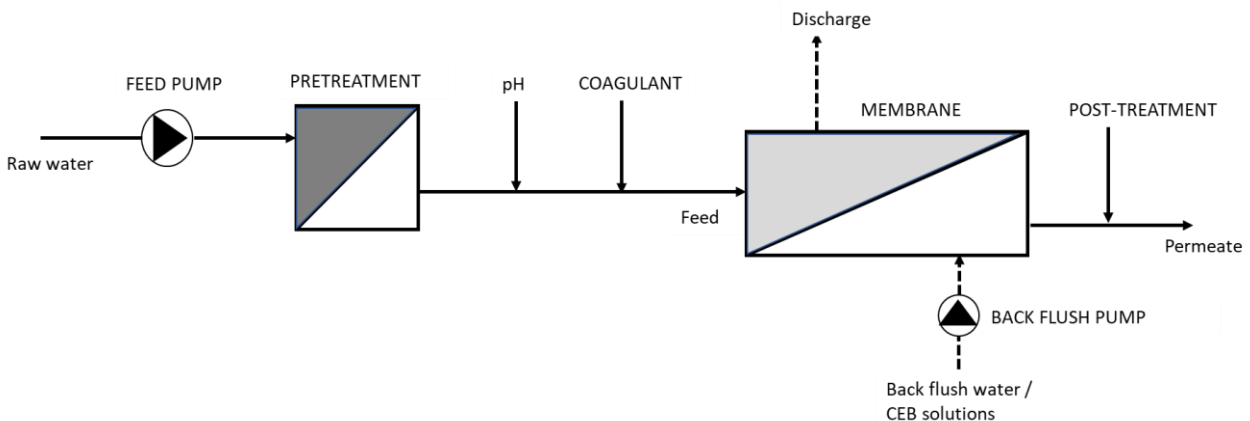


Figure 2. Typical process design of drinking water plants using UF.

3.2 Characteristics of Norwegian membrane-based drinking water plants

The two oldest waterworks participating in the survey were commissioned as early as 1991, while the newest waterwork started production in 2007. The age distribution of the waterworks that replied to the survey is assumed to give a representation of the Norwegian membrane facilities.

Figure 3 summarizes the key characteristics of Norwegian membrane-based drinking water facilities, including the distribution of membrane processes and type of membranes used. The figure also shows the distribution between different membrane materials. Of the 18 waterworks that responded to the survey, 17 waterworks use nanofiltration, while only one of the waterworks that responded to the survey uses ultrafiltration with pre-coagulation. Based on the dialogue with some of the 5-6 system suppliers in the Norwegian market, we have received information indicating that about 5 of the of the 160 membrane facilities for drinking water treatment that are in operation today are UF plants. In addition, one plant uses MF. This means that the large majority, *i.e.*, about 95% of the membrane plants for drinking water treatment, are NF plants.

Looking at membrane selection sorted by membrane manufacturers, it is seen that 44% of the waterworks use NF membranes from Hydranautics, while 22% and 16% respectively use membranes produced by Microdyn Nadir and Koch. 18% of the waterworks that responded to the survey did not report membrane type.

The membranes from Hydranautics are the so-called thin-film composite membranes where the active layer is made of sulfonated polyethersulfone (PES). Here, two subtypes are used, namely HydraCoRe 10 and HydraCoRe 50, having molecular weight cut-off (MWCO) of 3000 kDa and 1000 kDa, respectively. Thus, the latter one has smaller pores, and therefore better color removal at the expense of lower permeability compared to HydraCoRe 10.

The membranes from Microdyn Nadir and Koch constitute the second main type of NF membranes that are typically used for color removal in Norway today. These are asymmetric membranes of cellulose tri-acetate (CA). ROGA 8133 UF Magnum from Koch is no longer in sale but is still in use in some plants where membrane replacement has not been necessary after Koch stopped selling this product. Until recently, Microdyn Nadir has offered two types of nanofiltration membranes consisting of cellulose acetate, Trisep SBNF and Trisep SBUF, respectively, which have replaced the ROGA element in the Norwegian market. However, Microdyn Nadir has recently decided that they will no longer produce Trisep SBUF. Thus, Trisep SBNF will, to our knowledge, be the only cellulose acetate-based membrane that will be available in the market in the future. Trisep SBNF has a MWCO of 2000 kDa, *i.e.*, in between the two Hydracore membranes.

In addition to sulfonated PES and CA membranes, also polyamide-based membranes exist in the NF segment. However, we have not received information indicating that polyamide-based NF membranes are in use in drinking water facilities in Norway.

All NF membranes used in Norway today are spiral elements. In contrast, MF and UF membrane elements apply hollow fiber configuration.

The UF plant that answered the survey uses hollow fiber elements from X-flow of the type XIGA. The membrane material here is a mixture of PES and polyvinylpyrrolidone. Based on information provided by system suppliers of membrane systems in Norway, we know that hollow fiber elements from INGE are also in use in UF systems in Norway. These consist of the so-called multibore fibers, where 7 single fibers are cast together. The membrane material of these membranes is modified PES.

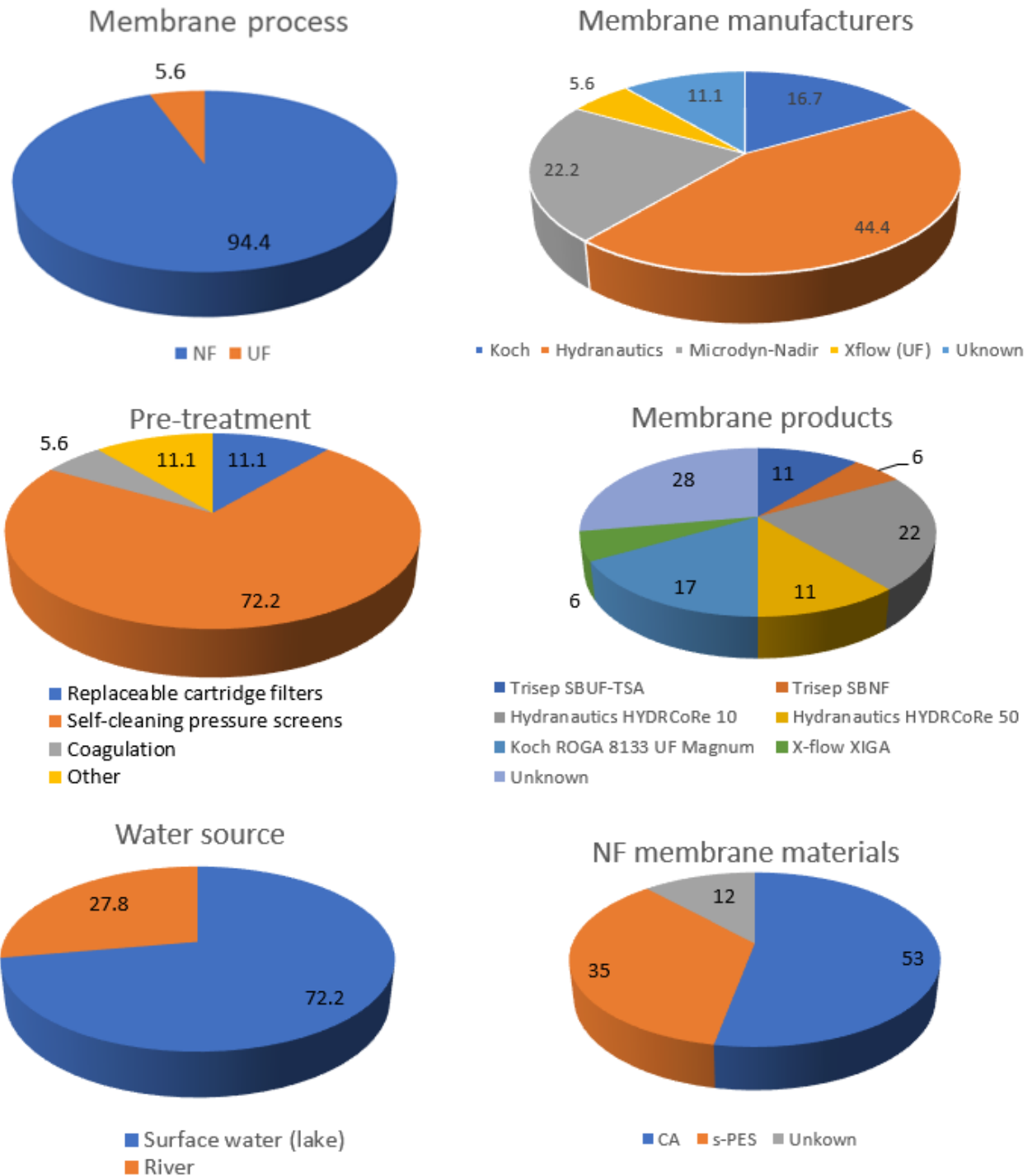


Figure 3. Distribution of the 18 water works responding to the survey.

Furthermore, Figure 3 shows the distribution of different types of pre-treatment among the waterworks that participated in the survey. Self-cleaning pressure screens are used by 72% of the facilities that responded. 11% of the waterworks use replaceable cartridge filters, 11% use multi-media filters ("other"), while the ultrafiltration plant uses coagulation as pre-treatment. Some waterworks did not indicate the light opening of

the applied filters. Of those who answered this question, 12 waterworks use filters having pore opening of 50 μm , while 2 waterworks used 25 μm . The ultrafiltration system used pre-filters with 50 μm pore opening.

The dominant source of raw water is surface water from lakes. As many as 72% of the waterworks that responded to the survey use surface water as raw water source. 28% of waterworks use rivers as a source of raw water, while none of the respondents uses groundwater.

All facilities report that they use post-treatment in one form or another. The primary purpose of the treatment is stated to be disinfection, where 11 waterworks use UV, 6 waterworks use chlorination, and one waterwork uses ozonation. Furthermore, 16 waterworks report that they use pH adjustment as a corrosion control before the water is entering the distribution network, of which 4 waterworks use limestone filters. In addition, one facility has reported use of water glass for corrosion control.

3.3 Water characteristics

Figure 4 reports the color of raw water and permeate, as well as the relative rejection of color by the membrane for the different waterworks participating in the survey. Two of the waterworks, ID9 and ID10, did not report any values, whereas the other waterworks reported only an annual average. Hence, no information of seasonal variation can be extracted. The color in the raw water varies from around 15 mg Pt/l up to 70 mg Pt/l, while the colour in the permeate is less than 6 mg Pt / ml. This gives a range of color retention from 79% to 97%. Figure 5 shows the turbidity in the raw water and the permeate. It is observed that the turbidity in the raw water is generally low (<1 NTU). The retention of turbidity varies typically in the range 75% to 95% between the waterworks. It is assumed that the waterworks that report the same turbidity in raw water and treated water are due to erroneous reporting.

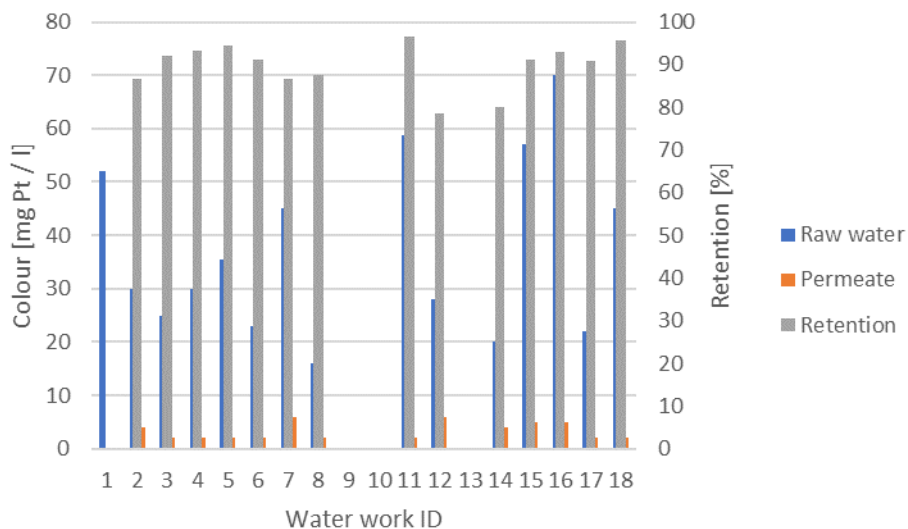


Figure 4. Average color in raw water and permeate, and rejection of color by the membrane.

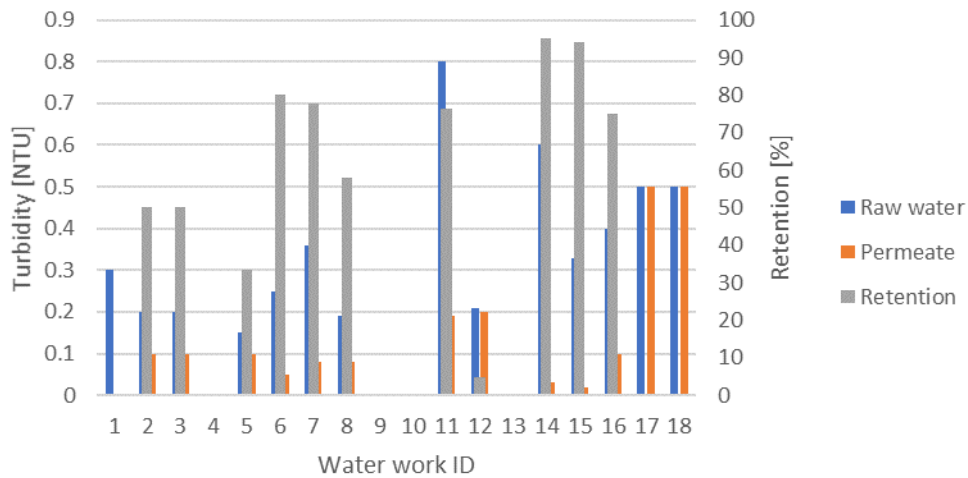


Figure 5. Average turbidity in raw water and permeate, and rejection of turbidity by the membrane.

3.4 Process operation and maintenance

There is a large spread in the production capacity of the waterworks that responded to the survey. The largest plant was designed for a water production equal to 500 m³/h, while the smallest waterworks was designed for a production of 2.7 m³/h. However, only a few of the waterworks are operated at design capacity. The average utilized capacity for the waterworks is 61%, while the lowest utilization is as low as 27%. No special challenges have been reported by this waterworks, and it is assumed that the redundant design capacity was chosen to account for future needs and/or seasonal variations.

The survey also asked for the design flux and if the water production varied according to the season. Only a few waterworks reported actual values of the design flux. The ultrafiltration plant reported as expected a significantly higher design flux of 80 l/m²/h, compared to the NF facilities that reported a design flux close to 15 l/m²/h. It is somewhat surprising that one of the nanofiltration plants reported a design flux as high as 25 l/m²/h, since it has been commonly accepted for a long time that the water flux should not exceed about 15 l/m²/h to avoid irreversible fouling [15]. The UF facility reported no difference in the actual production during summer and winter, however, the operating flux was lower than the design flux of 80 l/m²/h. The NF facilities reported higher production during summer, while 2 out of 3 NF facilities additionally reported the production during summertime was lower than the design flux.

The waterworks were also asked to report the cleaning regime. All waterworks used a variant of daily cleaning/flushing, but the type of chemicals, duration and concentrations varied. Most common is daily cleaning/flushing with water and chlorine. The most surprising finding is that one waterwork uses only daily rinsing without chlorine, but instead performs a weekly acid wash. An extended cleaning, typically soaking the membranes in cleaning solution over-night, is performed 1-4 times per year. The number of extended cleaning cycles per year typically reflects to what extent the membrane performance is observed to decline due to irreversible fouling.

The ultrafiltration facility uses a traditional cleaning regime, where the membranes are regularly backwashed with permeate, typically once or twice an hour. Backwashing is activated by either production time or accumulated water production, depending on which occurs first. After a certain number of backwashes, a

chemically enhanced backwash (CEB) is performed. The chemicals used in CEB alternate in this case between acid and caustic + chlorine.

It should be noted that cellulose acetate-based membranes and sulfonated PES membranes have very different pH and chlorine tolerance. The former can typically tolerate a pH range in the cleaning solution in the range of 2.5 -7.0 and 1 ppm chlorine, while sulfonated PES membranes can tolerate a wider range in pH (1-12) and up to 100 ppm chlorine.

3.5 Reported challenges

The survey also asked the waterworks to report operational challenges with the membrane facility based on the categories "impaired performance", "odour and taste" and "other problems". 8 waterworks have experienced reduced performance either permanently or for short periods under special conditions. Furthermore, 5 waterworks report challenges with odour and taste, but only one waterwork experienced this as a recurring problem due to the membranes. Amongst other problems reported, only ruptured membranes are directly related to the use of membranes. The remaining problems being reported such as frozen in-take pipe are general and can occur at any waterworks regardless of the process.

3.6 Operational costs

To get an overview of the most important operating costs, the waterworks were asked to state, annual energy consumption and required labour for operation and maintenance of the plant. The results are plotted as a function of reported capacity in Figure 6. The specific annual energy consumption decreases with increasing production and for water works with a production larger than 50 m³/h the energy consumption will be around 0.5 kWh/m³. A similar trend is observed for the required labour needed for operation and maintenance, indicating that for waterworks with a production larger than 50 m³/h the required percentage of a man-year is 0.5% per m³/h of production.

The reported consumption of chemicals is uncertain, partly because too few waterworks have responded, and partly because the answers are given on different units (kg, litres and NOK). In addition, different cleaning agents and different concentrations of chlorine solutions are used. The consumption of "chlorine" and "cleaning agent" is estimated only where the stated amounts of chemicals used, while different concentrations are not taken into account. This simplified estimate of consumption of chemicals is plotted in Figure 6 as a function of reported production. It can be observed that the consumption of chlorine and cleaning agent decreases with increasing production, and reaches a constant level for water works with production above 50 m³/h.

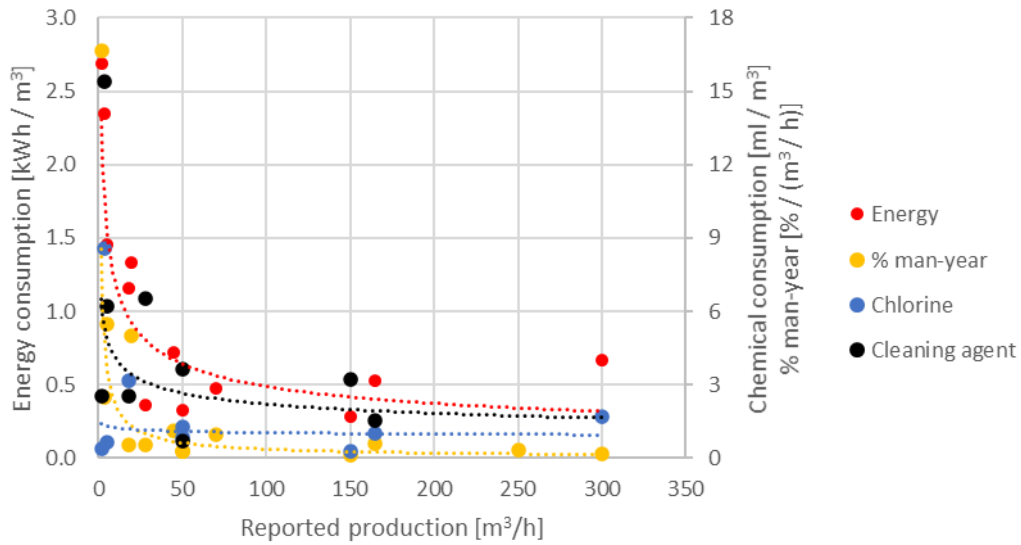


Figure 6. Energy, % man-year and chemical consumption versus reported production.

A significant factor contributing to the operating costs of membrane facilities is the costs associated with membranes replacement. The lifetime of the membrane is determined by several factors, where design flux and cleaning regimes are crucial for maintaining the membrane performance. Figure 7 shows the service life of membranes at the various waterworks before membrane replacement. The waterworks that lack data have either not replaced the membranes or have not provided this information. The service life of membranes is generally long, whereas 6 waterworks report a service life of 10 years or longer. The oldest membranes that were replaced were 17 years old. One waterworks replaced membranes already after 5 years, which is shorter than expected.

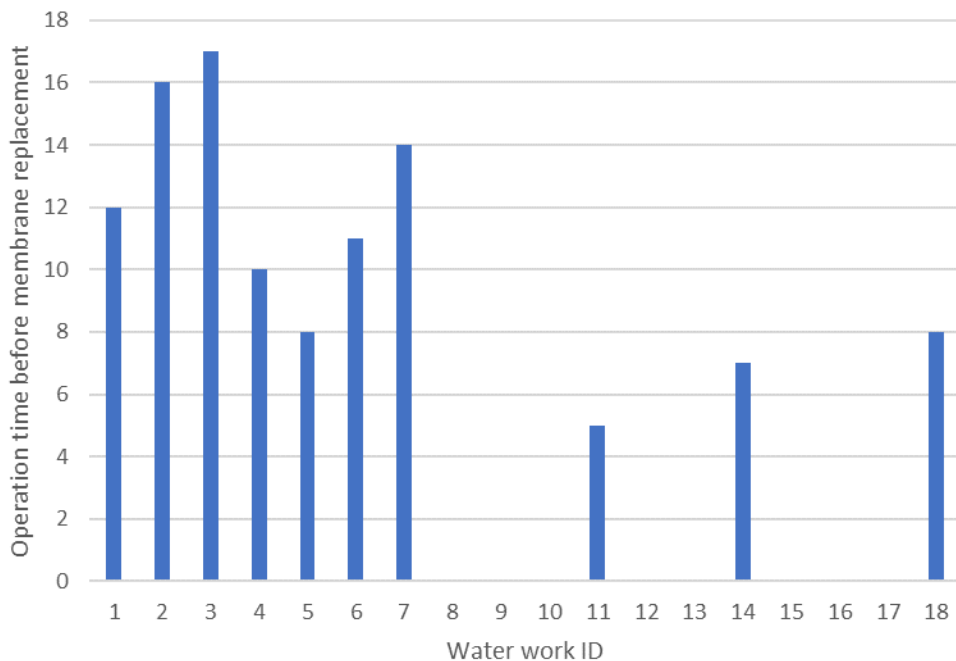


Figure 7. Lifetime of the membrane of last replacement.

3.7 Investment costs and footprint

Some of the waterworks that responded to the survey reported the investment cost associated with the construction of the facility. The wording of the question was "what is the total investment cost associated with the design and construction of the membrane plant?", but whether this also includes buildings and intake systems was somewhat unclear. The reported numbers are also not index-regulated. With this as a starting point, the investment costs showed a clear linear relationship between investment costs and design capacity. Furthermore, the investment cost per design capacity showed a clear decreasing trend with increasing design capacity, *i.e.*, the investment cost per cubic meter decreases with increasing plant size as shown in Figure 8. For waterworks with a design capacity larger than 50 m³/h the estimated investment cost is approximately 0.1 MNOK per m³/h.

The footprint requirement may also be an important factor for the choice of treatment process. The survey asked for "total footprint for process equipment". The specific footprint is plotted as a function of the design capacity in Figure 8 and shows that for waterworks with design capacity larger than 25 m³/h the expected footprint will be less than 2 m² per m³/h installed capacity.

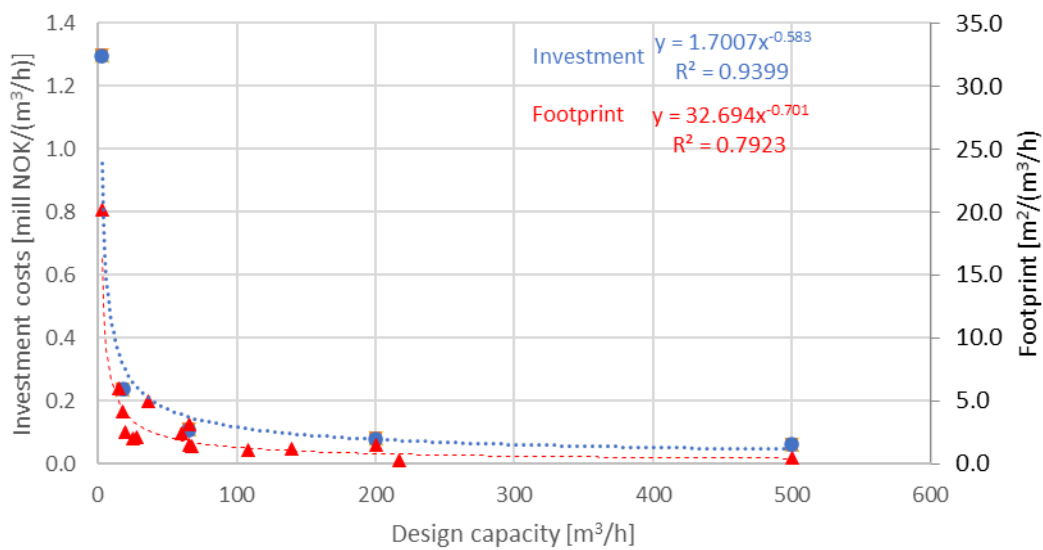


Figure 8. Investment costs and specific footprint.

4 Conclusions

Nanofiltration is by far the most used membrane process among Norwegian waterworks, counting more than 95% of the 160 facilities being registered in 2019. The remaining are UF and MF facilities using pre-coagulation. The same distribution between NF and UF/MF was observed among the waterworks responding to the survey. Many facilities operate at stable performance without considerable challenges. However, some facilities experience a significant decrease in performance over time due to membrane fouling.

The formation of fouling deposits on the membranes entails increased operating costs associated with increased energy consumption and more frequent cleaning requirements, as well as increased downtime and reduced production capacity. In addition, irreversible fouling leads to more frequent replacement of the membranes. Optimizing cleaning protocols and operating parameters such as cross flow and flux will help reducing operational challenges related to fouling. However, to avoid operational challenges related to fouling for new facilities it is important to obtain more knowledge about optimal selection of membrane process and membrane type for a given raw water characteristic.

Given optimal process design and operation, the use of membranes is considered well suited for treatment of coloured surface water for drinking water production.

Author Contributions: Conceptualization, Edvard Sivertsen and Willy Thelin; Data curation, Ingrid Selseth; Formal analysis, Edvard Sivertsen, Ingrid Selseth and Willy Thelin; Funding acquisition, Willy Thelin; Investigation, Edvard Sivertsen, Ingrid Selseth and Willy Thelin; Methodology, Edvard Sivertsen, Ingrid Selseth and Willy Thelin; Project administration, Willy Thelin; Software, Ingrid Selseth; Writing – original draft, Edvard Sivertsen and Willy Thelin. All authors have read and agreed to the published version of the manuscript.

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