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Feasibility and Environmental Impact of NOM Reduction by Microfiltration at a Finnish Surface Water Treatment Plant

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Abstract: Low-pressure membranes (including microfiltration and ultrafiltration) for natural organic matter removal in drinking water treatment have gained increasing interest in the Nordic countries. Microfiltration can produce stable water quality and requires less space than conventional treatment. Hollow fibre microfiltration coupled with chemical coagulation was tested at a Finnish surface water treatment plant to study its feasibility and environmental impact compared to clarification, rapid sand filtration and ozonation. Microfiltration improved both physical and chemical water quality, while natural organic matter removal was similar to that of conventional treatment. Membrane treatment would increase operational costs by 2.5–3.5 euro cents per m^3. Most of the costs derive from chemicals used in membrane cleaning. Membrane treatment is an energy-intensive process, but energy production in the Nordic countries has a low emission factor. Greenhouse gas emissions from operating microfiltration are estimated at 16 g CO_2-eq./m^3 of permeate. Lowering chemical consumption and using renewable energy in production could decrease total emissions.

Keywords: microfiltration; natural organic matter; environmental impact; drinking water treatment; surface water; retrofit

1. Introduction

Drinking water production in Finland is dependent on surface water. A total of 54% of the national drinking water is purified in surface water treatment plants (41%) and artificial groundwater plants (13%) [1]. Finnish surface waters generally have low turbidity, alkalinity and hardness but an elevated concentration of colour and natural organic matter (NOM) [2]. NOM itself is not a direct concern in drinking water. However, when combined with chlorine, NOM forms disinfection by-products [3]. Additionally, biodegradable organic matter intensifies microorganism regrowth in the distribution network [4], causes taste and odour problems [5], forms complexes with heavy metals [6] and affects the performance of unit processes.

Typically, NOM is removed by conventional treatment processes including coagulation, clarification and media filtration. Changes in raw water quality, especially seasonal variation, increase the stress on the purification process, as conventional treatment plants are slow to respond to quality changes. Alarmingly, several researchers have reported increasing NOM concentrations in surface waters across Northern Europe [7–9] and a proliferating number of extreme weather events [10] that can suddenly deteriorate water quality. For municipal water treatment plants that purify drinking water with conventional treatment, the ascending NOM content could increase treatment costs due to the increased consumption of coagulants [11] and/or decrease capacity by shortening the backwash interval for filters.

Membrane filtration can partially or entirely replace the conventional treatment as the main NOM removal step of a water treatment process. Moreover, membrane filtration produces stable water quality and can even act as a microbiological barrier. In Norway, nanofiltration is widely used for NOM removal, but these are small-scale treatment plants in remote areas that use only single-stage filtration [12], which leads to high operating costs. Thus, the treatment scheme may be inapplicable and economically unfeasible for large-scale water treatment plants. Therefore, research on the removal of NOM by membrane filtration has gained significant interest in the Nordic countries during the past decade [11,13,14].

Although membrane filtration could reduce treatment costs by decreasing the amount of coagulant required [15,16] and improving the multi-barrier effect against a microbial breakthrough [17,18], large initial investment and operational costs can be expected [19–21]. Beyond costs, however, treatment plants should also consider the environmental consequences of their treatment process. The operational setting of membrane filtration can significantly affect the environmental impact [22,23]. Furthermore, the decision to either replace existing steps [24] or add membrane filtration as an additional step [20] has a considerable effect on the environmental impact.

The use of micro- and ultrafiltration for the removal of NOM has been studied extensively [25–27]. This paper evaluates the feasibility of retrofitting a Finnish water treatment plant with microfiltration from both the water quality and operational costs perspective and what the effects on the environmental impact of the treatment process would be. The potential of the microfiltration process for replacing clarification and rapid sand filtration (RSF) is studied with the aim of improving treated water quality while minimising operational expenses and environmental impacts associated with microfiltration.

2. Materials and Methods

2.1. Feed Water

This study was conducted at the Pitkäkoski water treatment plant (WTP), a surface water treatment plant located in Helsinki, Finland. Raw water going to the WTP is taken from Lake Päijänne, which is about 100 km north of the treatment plant. Lake Päijänne is the second largest lake in Finland, with a catchment of 26,460 km^2. Raw water is taken from the southern part of the lake, which can be described as an oligotrophic lake with an elevated total organic carbon (TOC) concentration varying between 6.8 mg/L and 8.2 mg/L. The water flows by gravity to the treatment plant through a 120 km rock tunnel.

The treatment process at Pitkäkoski WTP consists of chemical coagulation with ferric sulphate, sedimentation, RSF, ozonation, granular activated carbon filtration and UV disinfection. Prior to distribution, the water is disinfected with monochloramine chlorination, which inhibits microbial growth in the network. Finally, limewater and CO_2 are added to remineralise the water to decrease pipe corrosion.

2.2. Pilot Experiment

Commercial outside-in hollow fibre membrane in dead-end mode was trialled in this study (Figure 1). The membrane was produced by Pall at a pore size of 0.1 μm. The membrane material was polyvinylidene fluoride with modified hydrophilic surface characteristics and a pH range of 1–10. One module contained 140 fibres. The inner and outer diameters of one fibre were 0.7 mm and 1.4 mm, respectively. The total membrane surface area in the module was 0.26 m^2.
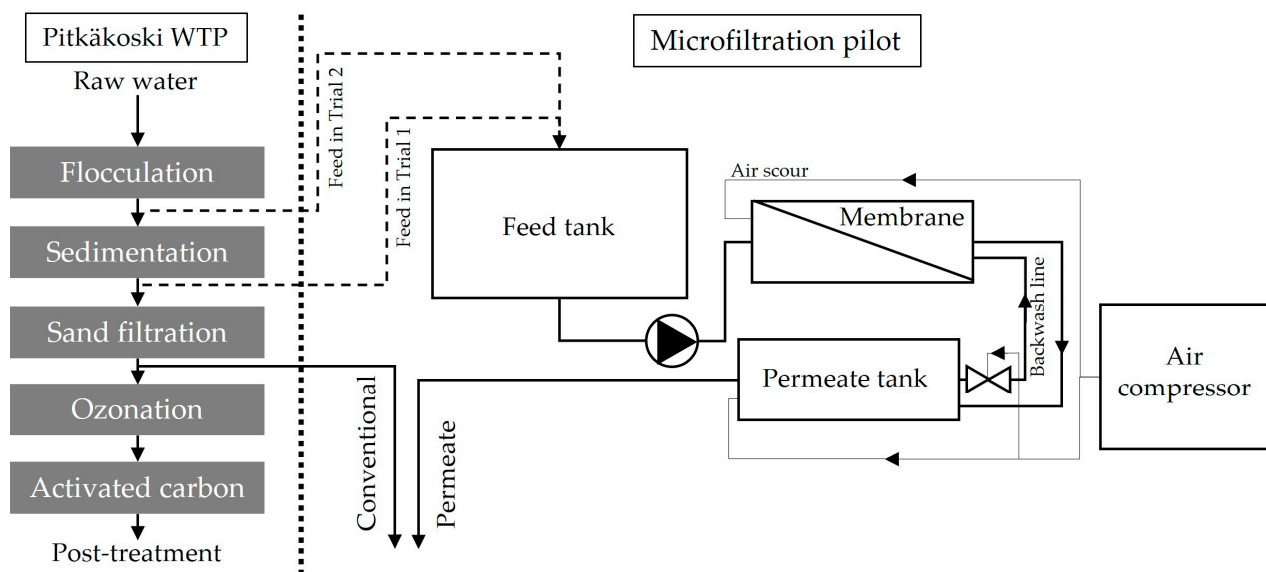
Figure 1. Process diagram of the membrane trials. Feed water in Trial 1 was taken after clarification, while in Trial 2 feed water was taken before the clarification step.

Two pretreatment arrangements were applied prior to the membrane filtration to study the necessary pretreatment prior to microfiltration. First, raw water was precipitated with ferric sulphate (PIX-322, Kemira, Helsinki, Finland), after which flocculation took place in quadripartite flocculation chambers, each equipped with a vertical mixer. The total flocculation time was 30 min, enough for effective NOM removal prior to membrane filtration [28,29]. In Trial 1, water was further treated with sedimentation, and feed water was taken from the collecting channel. In Trial 2, feed water was taken after the last flocculation chamber.

Membrane performance was measured as flux through the membrane. Due to changing raw water temperature during piloting, normalized flux was used to compare different operational settings and to aid in the comparison with studies in the literature. Normalised flux at 20 °C (J_{20}) is calculated as:

$$J_{20} = J_M \times (\mu_M / \mu_{20}) \ (L/m^2h) \tag{1}$$

where J_M is the calculated flux at temperature M (L/m^2h), μ_M is the dynamic water viscosity at temperature M (Pa·s) and μ_{20} is the dynamic water viscosity at temperature 20 °C (Pa·s).

Membrane flux and backwash intervals were altered to find the economically most feasible operating conditions with a high recovery percentage (Table 1). The membrane flux alternated between 90 and 110/130 L/m^2h, while the backwash interval varied from 20 to 30 min. The backwash consumed the same amount of water each time, but the backwash flux changed between 1000 and 1300 L/m^2h. When using a lower backwash flux of 1000 L/m^2h, a longer, less aggressive backwash was conducted. The higher backwash flux of 1300 L/m^2h produced a more rapid, high-pressure backwash. Permeate was used for the membrane backwash, and at the same time pressurised air was utilised to air scour the membrane feed side. Before each setup change, normalized water permeability (NWP) was verified by measuring the tap water flux under standard pressure to ensure a similar initial NWP for each operating condition.

Table 1. Different operational conditions were studied during the trials. Membrane flux, backwash interval and backwash flux were altered to find the optimal conditions for running the pilot. Settings 7 and 8 were only used in Trial 1, while the lower flux during Trial 2 was chosen in settings 5 and 6.

Parameter	Setting 1	Setting 2	Setting 3	Setting 4	Setting 5	Setting 6	Setting 7 [†]	Setting 8 [†]
Flux (L/m^{2}h) at 20 °C	90	90	90	90	130/110 *	130/110 *	130	130
Backwash int. (min)	20	30	20	30	20	30	20	30
Backwash flux (L/m^{2}h)	1000	1000	1300	1300	1000	1000	1300	1300

Note: * Lower flux during Trial 2 [†] The setting applied only with clarified water (Trial 1).

A chemically enhanced backwash (CEB) was performed every other day to clean the membrane to guarantee stable operating conditions. One CEB cycle began with an acid CEB at pH 2 (0.5% citric acid) for 12 min to remove inorganic foulants such as iron from the coagulant. Between acid and oxidising CEB, the membrane was rinsed with tap water. Rinsing was followed by an oxidising CEB at pH 11 (0.1% NaOCl) for 12 min and another rinse with tap water. The CEB was also conducted in reverse order, but performance recovery was better with acid CEB first.

2.3. Water Quality Analyses

Water quality analyses were carried out to compare the microfiltration permeate with the conventional treatment effluent after sand filtration. Feed waters for Trial 1 and Trial 2 for the membrane were taken from the same treatment line at Pitkäkoski WTP. The feed sample was taken from the pilot feed tank, while the permeate sample was collected from the pilot-unit effluent. The conventional treatment sample was collected after RSF from another treatment line. Although the treatment lines are identical, small differences in produced water quality are possible, especially in turbidity and NOM concentration.

TOC and dissolved organic carbon (DOC) were measured three times a week with a TOC analyser (TOC-V$_{CPH}$, Shimadzu, Kyoto, Japan). Prior to analysis, the samples for DOC were filtrated through a 0.45 μm polyethersulfone filter. UV absorbance at a wavelength of 254 nm (UVA$_{254}$) was detected daily with a spectrophotometer (Lambda UV/Vis, PerkinElmer, Waltham, MA, USA). Specific UV absorbance (SUVA$_{254}$) was defined as UVA$_{254}$ divided by DOC.

The composition of the organics present in the water sample was characterized once a week by liquid chromatography–organic carbon detection (LC-OCD) according to Huber et al. [30]. The sample was eluted through a column (Toyopearl HW-50S, Tosoh, Tokyo, Japan). First, the UVA$_{254}$ of the sample was determined using a fixed wavelength detector (K200, Knauer, Berlin, Germany). Second, the organic carbon of the eluted sample was decomposed to CO_2 in a reactor (Gräntzel-reactor, DOC-Labor, Karlsruhe, Germany), and the volume of formed CO_2 was measured with an infrared detector (Ultramat 6, Siemens, Munich, Germany). A software (FIFFIKUS, DOC-Labor, Karlsruhe, Germany) was employed to separate DOC into five major subfractions: biopolymers, humics, building blocks, low molecular weight (LMW) acids and LMW neutrals.

Turbidity was measured daily with a turbidity meter (2100AN, Hach, Ames, IA, USA) while iron concentration was quantified three times a week with a spectrophotometer (Lambda UV/Vis, PerkinElmer, Waltham, MA, USA). Heterotrophic bacteria were enumerated by a spread plate technique, with R2A agar incubated at 20 °C for 7 days according to Greenberg et al. [31] (HPC). Assimilable organic carbon (AOC) was determined according to Miettinen et al. [32]. Two bacterial strains, namely *Pseudomonas fluorescens P17* and *Aquaspirillum NOX* were used simultaneously. The growth yield of *P. fluorescens P17* was determined using sodium acetate (Merck, Darmstadt, Germany) as a substrate, while sodium oxalate (Merck, Darmstadt, Germany) was used to determine the growth yield of *Aquaspirillum NOX*. Both strains were cultured (spread plating) on R2A agars. A second-generation adenosine triphosphate (ATP) analysis was performed with a water test kit (LumiKem, Kemira, Helsinki, Finland). The luminescence of a sample was measured with

a luminometer (C-100, Kikkoman, Tokyo, Japan). HPC, AOC, and ATP were analysed once a week. Turbidity and AOC in feed water were analysed only in Trial 1 due to the high iron content of flocculated water in Trial 2 disturbing the measurement. Similarly, the iron concentration in Trial 2 was known to be 6.25 ppm Fe, as this is the dose applied in the purification process.

2.4. Cost Calculation and Environmental Impact Assessment Methodology

The cost estimation of a membrane process retrofit was based on the data gathered from the pilot study and from Pitkäkoski WTP. Four operating conditions were selected for closer study. The selection of operating conditions was based on permeate production and fouling characteristics during operation. The results are calculated for the production of 1 m^3 filtrated permeate.

Energy and chemical consumption are estimated for the cost calculation. Additionally, membrane modules are included as they are considered consumables. The operating costs at Pitkäkoski WTP were used as the base calculations, and the operating costs of the membrane treatment were compared to the base costs. Costs for energy and chemicals were collected in the year 2020. However, due to significant volatility in electricity prices in Northern Europe, energy costs are also estimated based on the electricity futures for the year 2024.

The environmental impact of microfiltration was estimated from numbers found in the literature. The calculations were based on data for chemical consumption and energy usage gathered from the pilot results and operating conditions at Pitkäkoski WTP, both of which are presented in the economic analysis. All numbers are calculated for the production of 1 m^3 of filtrated water. The methodology is influenced by the life cycle assessment methodology, but only the greenhouse gas (GHG) emissions presented as CO_2-eq. are considered. Only the operating conditions are included in the assessment. Construction of the microfiltration unit and supporting equipment is excluded. However, membrane modules are included, like in the cost calculation. Additionally, the energy required for backwash wastewater treatment is estimated.

3. Results and Discussion

3.1. Water Quality

3.1.1. Natural Organic Matter Removal

Water quality results (Table 2) indicate that microfiltration with chemical pretreatment is capable of competing with the conventional treatment in NOM removal. After chemical coagulation, particulate organic matter is removed well, but the dissolved organic matter remains in the water, regardless of the treatment selected. This indicates that the removal of NOM is dependent on the coagulation rather than the type of filtration. The average residual TOC in effluent water was comparable between microfiltration and conventional treatment in both trials with the difference between the two treatment options being 0.09 mg/L and 0.13 mg/L for Trial 1 and Trial 2, respectively. The UVA$_{254}$ was equal between microfiltration and conventional treatment throughout the piloting.

High hydrophobicity and aromaticity are associated with SUVA$_{254}$ above four [33]. In this study, the SUVA$_{254}$ results revealed that the remaining NOM for both permeate and conventional effluent was mainly hydrophilic. SUVA$_{254}$-values indicate that further improving NOM removal requires other treatment methods besides coagulation. Studying DOC and UVA$_{254}$ removal rates confirms this. Raw water had a TOC of 7.5 mg/L and UVA$_{254}$ of 0.203 cm^{-1}. Around 80% of UV-absorbing compounds were removed from treatment plant raw water, compared to approximately 65% of raw water DOC, indicating higher removal of hydrophobic NOM. The low aromaticity of permeate was also confirmed by the LC-OCD results.

Table 2. The results of water quality analyses show that microfiltration improves microbial and physical water quality compared to conventional treatment. Microfiltration is also capable of removing NOM to the same extent as conventional treatment.

Analysis NOM	Feed water	Trial 1 Permeate	Conventional	Feed water	Trial 2 Permeate	Conventional	n
TOC (mg/L)	3.3 ± 0.17	2.6 ± 0.20	2.5 ± 0.20	7.0 ± 0.10	2.5 ± 0.07	2.4 ± 0.10	13
DOC (mg/L)	2.7 ± 0.14	2.7 ± 0.19	2.5 ± 0.13	2.7 ± 0.19	2.7 ± 0.08	2.4 ± 0.07	13
UVA$_{254}$ (cm^{-1})	0.117 ± 0.006	0.042 ± 0.002	0.043 ± 0.005	0.096 ± 0.035	0.044 ± 0.003	0.044 ± 0.005	23
SUVA$_{254}$ (L/mg·m)	4.9 ± 0.27	1.8 ± 0.09	2.0 ± 0.21	4.0 ± 0.80	1.7 ± 0.12	1.8 ± 0.20	13
Microbiological		**Trial 1**			**Trial 2**		**n**
HPC (cfu/mL)	115 ± 47	7.1 ± 0.0	71 ± 18	120 ± 42	9.9 ± 6.8	20 ± 16	3
ATP (pg/L)	5.7 ± 2.7	2.2 ± 1.1	3.4 ± 1.0	25 ± 8.7	5.3 ± 3.8	7.8 ± 7.5	3
AOC (µg AOC-C/L)	32 ± 16	50 ± 24	31 ± 21	-	17 ± 0.8	20 ± 4.6	3
Physical Quality		**Trial 1**			**Trial 2**		**n**
Turbidity (FTU)	1.17 ± 0.1	0.06 ± 0.01	0.18 ± 0.05	-	0.06 ± 0.01	0.18 ± 0.17	22
Iron (µg Fe/L)	920 ± 46	42 ± 6.3	109 ± 23	6250	26 ± 6.6	74 ± 83	11

LC-OCD results (Figure 2) showed that microfiltration removed more biogenic NOM (comprising of biopolymers and LMW compounds). The total removal of chromatographic NOM was equal, 16% and 20% in Trial 1, and 47% and 48% in Trial 2 for permeate and conventional treatment, respectively. LMW acids in particular were removed more effectively. Biogenic NOM has been connected to biofilm growth in the distribution network, as this fraction is the smallest and thus the most available as a nutrient for bacteria [34]. However, both treatments removed mostly humic substances and their degradation products, as this is the fraction most amenable to coagulation [33].
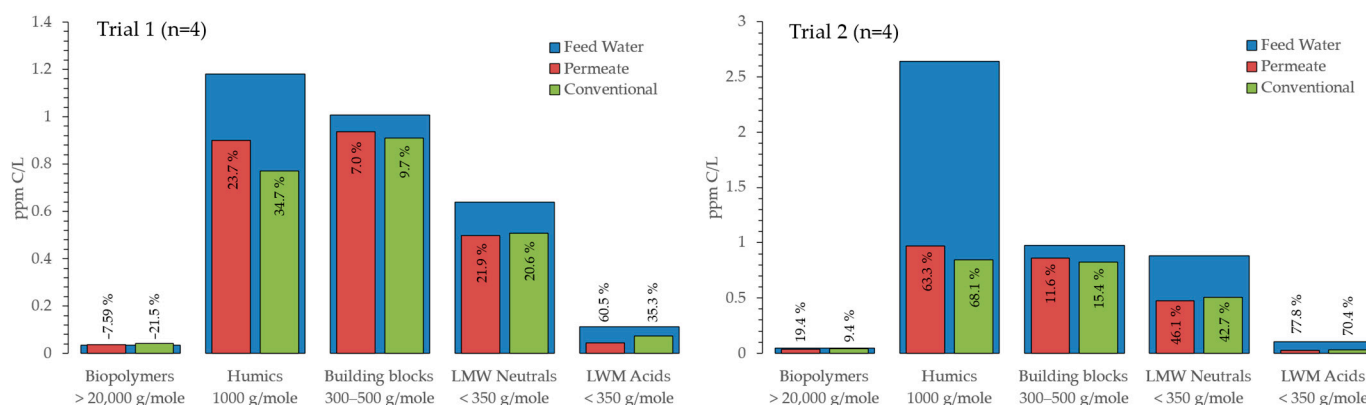
Figure 2. LC-OCD results showed that conventional treatment was more effective in removing humics, but microfiltration could reduce the amount of biogenic NOM, including biopolymers and LMW compounds. The numbers indicate the percentage of substances removed from the feed water.

3.1.2. Physical and Microbiological Quality

The physical and microbiological quality of permeate was better than with the conventional treatment (Table 2). The amount of residual iron and turbidity in permeate was always below the drinking water quality recommendations set by the European Union. Permeate quality fluctuated less than in conventionally treated water. The range of variation for turbidity was 0.06–0.1 FTU and 0.11–0.38 FTU for the permeate and the conventional treatment, respectively.

Microbiological analyses imply that microfiltration effectively removes microbes from water; very little growth was observed in cultivation. ATP concentration was also smaller compared to conventionally treated water. However, it remained unclear whether the microbial activity was diminished by the membrane or the coagulant, as both can effectively extract microbes from water. The improved microbiological quality strengthens the multi-barrier system against microbiological outbursts in the treatment plant.

Nevertheless, the AOC results suggest that microfiltration is incapable of reducing the biofilm formation potential in the distribution network where AOC is easily utilised by bacteria. The inability of microfiltration to remove AOC in comparison to NOM is due to the molecular weight of AOC, which is typically below 1000 Da [35].

3.2. Water Production between Settings

Microfiltration with eight operational settings was tested in Trial 1. Setting 1 was used as a baseline for comparing other settings. Approximately 40.5% of NWP was lost after a 42 h filtration period (Figure 3). Taking into account downtime and permeate used for backwashing, the average permeate production was 10.6 L/h during the 42 h (Table 3). NWP was fully recovered with a CEB. Reducing the CEB interval to 24 h slightly increased permeate production, as loss in NWP decreased and less water was consumed for backwashing. Average permeate production with a 24 h CEB interval was 10.9 L/h.
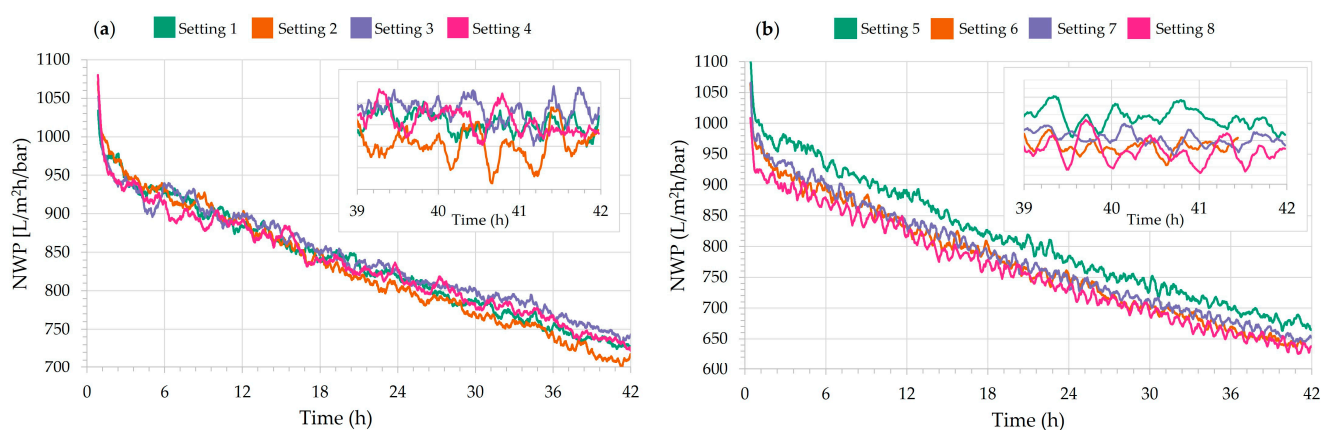
Figure 3. Normalised water permeability with (**a**) 90 LMH flux and (**b**) 130 LMH flux during Trial 1 with a 42 h filtration period.

A longer backwash interval was tested with settings 2, 4, 6 and 8. Prolonging the backwash interval to 30 min had a minor negative impact on average membrane NWP compared to a 20 min interval (settings 1, 3, 5 and 7), as the NWP loss after 42 h increased 0.6–2.0% with the longer backwash interval. However, average permeate production increased by 8.0–16.0% with all tested longer backwash interval settings because less permeate was used for backwashing.

Backwash efficiency was tested by changing the backwash flux. The same amount of water was consumed in each backwash, but the duration of the event changed. Operating with a lower backwash flux meant that the duration of the backwash was 20 s. Higher flux resulted in a shorter backwash event that lasted only about 15 s. As the results in Table 3 show, the more aggressive but shorter backwash (settings 3, 4, 7 and 8) had an insignificant effect on membrane performance compared to a lower flux backwash (settings 1, 2, 5 and 6). Similar permeate production was measured with both high and low backwash flux.

Table 3. Performance indicators and the amount of permeate produced with each setting during the experiment.

Setting	Average Flux (L/m²h)	Average TMP (bar)	Average NWP (L/m²h/bar)	NWP Loss (%)	Average Permeate in 42 h (L/h)	Average Permeate in 24 h (L/h)
			Trial 1 (Clarified Feed Water)			
1	91.2	0.110	836	40.5	10.6	10.9
2	91.2	0.111	830	41.1	12.1	12.5
3	90.4	0.108	844	39.4	10.7	10.8
4	91.9	0.111	835	41.4	12.3	12.5
5	128.1	0.159	812	43.4	16.3	17.2
6	127.9	0.167	775	43.9	17.8	18.6
7	129.4	0.167	781	44.8	16.7	17.5
8	130.2	0.173	760	43.0	18.4	19.2
Setting			**Trial 2 (Flocculated Feed Water)**			
1	87.5	0.174	505	37.8	10.0	10.1
2	87.8	0.180	491	40.8	11.6	11.8
3	88.3	0.188	472	41.8	10.1	10.2
4	86.2	0.188	459	44.5	11.3	11.7
5	107.9	0.248	438	42.8	13.2	13.5
6	105.3	0.266	397	45.6	14.3	14.7

Increasing the membrane flux by 45% to 130 L/m²h resulted in more fouling and higher average transmembrane pressure (TMP). However, permeate production increased by 47–56%. A daily CEB could increase permeate production. However, this would increase the consumption of cleaning chemicals.

Flocculated water was utilised in Trial 2. Increased NWP loss was expected as the feed water had a higher suspended solid content. The rate of membrane fouling increased significantly, as the TMP was on average 60% higher in all settings compared to Trial 1 (Figure 4). However, NWP loss after 42 h of filtration was similar to Trial 1.
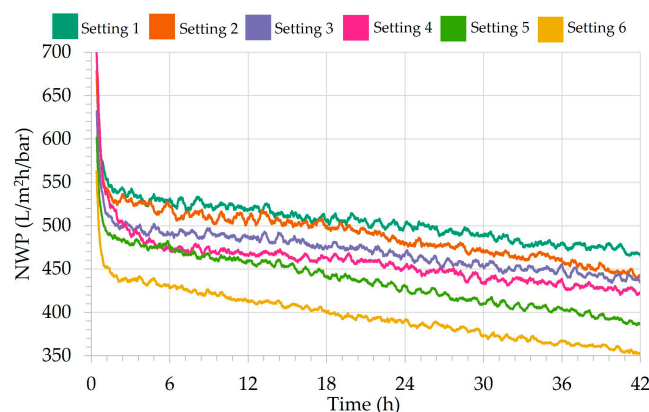
Figure 4. Normalised water permeability during Trial 2 with a 42 h filtration period.

Again, backwash intensity had an insignificant effect on membrane production. Having a longer backwash interval did increase permeate production by 11–16% when lower membrane flux was applied and by 8% when operating with higher flux. Significantly, shortening the CEB interval from 42 h to daily CEB had a very small effect on permeate production.

Based on the performance results, four settings were selected for a closer cost calculation and environmental impact assessment. The selection was based on NWP loss and permeate production capability. These factors were assumed to reflect lower operational expenses and environmental impact. Setting 6 from both Trial 1 and Trial 2 was selected based on permeate production, and Setting 2 from Trial 1 was also chosen for further evaluation. Finally, Setting 6 from Trial 1 with a shorter CEB interval of 24 h was selected. Higher

backwash flux was estimated to consume more energy per m^3 of permeate. Therefore, insignificant improvement in membrane performance excluded these for further studies.

3.3. Cost Comparison

3.3.1. Membrane Treatment Process Composition

The current treatment process at Pitkäkoski WTP consists of chemical coagulation (dose 6.25 ppm Fe), flocculation, clarification, RSF, ozonation, activated carbon filtration, UV-disinfection and post-chemicalisation. Based on the permeate quality results, microfiltration can replace RSF, also clarification can possibly be omitted. Microfiltration also effectively removes pathogens and bacteria, which supports the exclusion of ozonation. The permeate quality results did not indicate that a significantly lower coagulation chemical dose could be used with microfiltration in contrast to what has been reported in the literature [15,16,36].

3.3.2. Energy Consumption

A dead-end microfiltration process uses relatively low pressure and requires only a pressurising pump on the feed side. The energy consumption by the feed pump (P_{feed}) was calculated from:

$$P_{feed} = (Q_P \times \Delta P)/(\eta \times 3.6 \times 10^4) \text{ (kWh)} \tag{2}$$

where Q_P is the average permeate flow (L/h), ΔP is the feed pressure (bar) and η is the assumed pump efficiency.

The amount of air consumed during air scour could not be measured. However, based on the manufacturer's recommendation, an airflow of 0.1 m^3/m^2/h was assumed. The energy consumption of an air blower was estimated at 0.025 kWh/m^3 of air.

With an average flux of 90 L/m^{2}h and a feed pressure of 0.73 bar (Setting 2) and accounting for the amount of energy and produced permeate used in backwashing, the energy consumption totalled 0.061 kWh/m^3. Increasing the flux to 130 L/m^{2}h improved energy efficiency, as the total energy consumption was 0.054 kWh/m^3 despite the increase in average feed pressure to 0.83 bar (Setting 6). If the operation period with Setting 6 was shortened to 24 h, the energy consumption was 0.053 kWh/m^3. In Trial 2, a higher feed pressure was required to produce a flux of 110 L/m^{2}h, which meant that the energy consumption increased to 0.064 kWh/m^3.

Throughout the trial the water temperature remained stable, ranging between 3.8 °C and 5.0 °C. This is lower than the average yearly temperature of 8 °C experienced at Pitkäkoski WTP, meaning that a slightly lower total energy consumption is to be expected.

3.3.3. Chemical Consumption

A CEB was conducted every other day, except in the 24 h Setting 6 test. When operating under Setting 2 conditions and producing on average 12.1 L/h, the chemical consumption of the oxidizing CEB was 0.044 mole NaOCl/m^3, and the acid cleaning required 0.007 mole citric acid/m^3 of permeate. With the higher flux used in Setting 6, the chemical consumption decreased to 0.030 mole NaOCl/m^3 and 0.005 mole citric acid/m^3, respectively.

A shorter CEB interval of 24 h improved permeate production to 18.6 L/h, while chemical consumption also increased to 0.050 mole NaOCl/m^3 and 0.008 mole citric acid/m^3, respectively. In Trial 2 with Setting 6, the permeate flow was 14.3 L/h. The lower average water production compared to Trial 1/Setting 6 meant that 0.037 NaOCl/m^3 and 0.006 mole citric acid/m^3 were required for CEB.

3.3.4. Required Membrane Surface Area

The choice of flux and feed water temperature, which affects viscosity, has a significant impact on the number of modules required. With a normalized flux of 90 L/m^{2}h in Trial 1/Setting 2 and excluding downtime and backwash, approximately 21.49 m^2 of membrane is required to produce 1 m^3/h of permeate.

Increasing the flux to 130 L/m^{2}h decreases the required membrane surface area to 14.61 m^2. With a shorter CEB interval of 24 h, the average permeate production is increased and the required membrane surface area to produce 1 m^3/h reduces to 13.96 m^2. When treating flocculated water, the membrane surface area demand is 18.18 m^2. Pitkäkoski WTP has a capacity of 7500 m^3/h. Therefore, the treatment plant would need 2190–3225 modules, with one hollow fibre module having a surface area of 50 m^2.

The whole study period lasted six months. Due to the short duration of the study, membrane lifetime is difficult to assess. Based on the feed water conditions, a five-year lifetime was assumed for Trial 2 conditions, while the membrane treating the clarified water in Trial 1 was assumed to have a lifetime of seven years.

3.4. Economic Summary

The costs of chemicals and electricity are based on prices at Pitkäkoski WTP in 2020. As previously mentioned, the cost of electricity has significantly increased since 2020. Therefore, energy costs were also calculated based on the energy futures for 2024. Table 4 shows that approximately 60% of the operational costs for 1 m^3 of treated water derive from the chemical usage for CEB. If the electricity price for 2024 is used, the chemical costs would constitute approximately 55% of the total costs. The estimated costs of every operational setting are in Table S1.

Table 4. Cost of operating microfiltration at different settings in EUR/m^3. The chemicals used in CEB account for approximately 60% of the operational costs. The energy expenses would double if energy futures for 2024 are used.

Trial	Setting	Citric Acid	NaOCl	Energy	Modules	Total	Total (2024 Energy)
1	2	0.0124	0.0090	0.0033	0.0105	0.035	0.038
1	6	0.0084	0.0061	0.0029	0.0071	0.025	0.027
2	6	0.0105	0.0076	0.0035	0.0125	0.034	0.037
1	6 (24 h)	0.0141	0.0102	0.0029	0.0068	0.034	0.037

The retrofitting membrane treatment would increase the operational costs of a WTP from 0.025 EUR/m^3 to 0.035 EUR/m^3. Liden et al. [11] calculated the costs of operating a hollow fibre ultrafiltration membrane for NOM removal in a Swedish surface WTP and reported similar total costs for ultrafiltration. Slightly higher operating costs were reported for smaller WTPs [37].

While Trial 2 suggested that clarification improved permeate quality only a little, the costs of running membrane filtration were around 36% higher. Even if a similar membrane lifetime was assumed, the increase would be 23%. Running with a lower flux of 90 L/m^{2}h (Setting 2) was slightly more expensive than utilising a daily CEB routine with a higher flux.

3.5. Environmental Impact of Microfiltration

GHG emissions from electricity produced in Finland have steadily decreased. However, Finland is not self-sufficient in electricity production, and foreign electricity is also used. Therefore, the GHG emissions of electricity usage are calculated based on the emission factors of electricity used in Finland [38], which in the year 2020 were 72 g CO_2-eq./kWh. CO_2-eq. emissions from energy usage for microfiltration ranged from 3.8 g CO_2-eq./m^3 to 4.6 g CO_2-eq./m^3, with the highest impact produced in Trial 2 conditions.

Two chemicals were used in CEB. The GHG emissions during citric acid production were calculated using data from Wang et al. [39], while data from Alvarez-Gaitan et al. [40] were used for NaOCl. However, the GHG emissions for NaOCl are calculated in the Australian context. Depending on the state or territory, the GHG emissions of the Australian electricity grid [41] are 2 to 12 times higher compared to the Finnish grid. Because the

NaOCl used at Pitkäkoski WTP is produced in Finland, the GHG emissions of membrane treatment are also reported with the NaOCl emissions halved.

The GHG emissions for membrane replacement are calculated based on data from Bonton et al. [24]. They reported an emission factor of 3 g CO_2-eq./m^3 drinking water with an estimated lifetime of 10 years for nanofiltration membranes. However, in this work the lifetime is assumed to be 5–7 years. Accounting for the fact that the microfiltration membrane studied is around three times as efficient per surface area, 2 g CO_2-eq./m^3 was assumed for a 7-year lifetime. With a shorter lifetime of five years, 3 g CO_2-eq./m^3 was used.

Compared to conventional treatment, microfiltration with hollow fibre membranes produces significantly more wastewater. The produced wastewater, however, has low organics content, and CEB wastewater requires only the removal of residual chlorine and the neutralization of pH. Recovery ranged between 81.0% and 87.4% between the selected settings, meaning that approximately 126–190 L of wastewater per m^3 of permeate were produced. At Pitkäkoski WTP, all wastewater is pumped to a wastewater treatment plant (WWTP), where again pumps are required to lift the wastewater to the treatment plant. According to Mölsä [42], pumping energy usage at the WWTP causes an impact of approximately 10 g CO_2-eq. per m^3 of wastewater. When operating under Trial 1/Setting 2 conditions, energy usage at the WWTP causes 1.91 g CO_2-eq./m^3. For Setting 6, the higher recovery reduces the environmental impact to 1.32 g CO_2-eq./m^3. Shortening the operational period to 24 h, the wastewater treatment impact is even lower: 1.28 g CO_2-eq./m^3. Operating under Trial 2/Setting 6 conditions, the emissions from energy usage at WWTP are 1.65 g CO_2-eq./m^3.

The total environmental impacts of the selected four operating conditions are presented in Table 5, while emissions from every setting are listed in Table S2. The comparison is based solely on emissions from operating microfiltration as previous studies have shown that 90% of the environmental impact is attributed to the operational phase of the life cycle, and the rest is due to construction [24,43]. CEB chemicals account for 62–74% of the total CO_2-eq./m^3 emissions. If the emissions of NaOCl production are halved, approximately 53–66% of the total impact stems from chemical usage. Energy usage produces the second-most emissions, accounting for 14–20%. A similar share of emissions has been reported for ultrafiltration [44], although lower total CO_2-eq. emissions were reported.

Table 5. Summary of the total GHG emissions for microfiltration with different operational settings (g CO_2-eq./m^3). Operating with high flux and clarified water produces the lowest environmental impact, whereas the other operating conditions are closer to each other.

Trial	Setting	Citric Acid	NaOCl	Energy	Modules	Wastewater Treatment	Full NaOCl Impact	Half NaOCl Impact
							Total	
1	2	6.46	11.62	4.40	2.00	1.91	26.40	20.59
1	6	4.41	7.93	3.87	2.00	1.32	19.54	15.57
2	6	5.48	9.85	4.61	3.00	1.28	24.60	19.68
1	6 (24 h)	7.36	13.23	3.79	2.00	1.65	27.65	21.04

Vince et al. [45] studied the GHG emissions of different treatment processes in drinking water treatment and reported that CO_2-eq. emissions for ultrafiltration are similar when compared to a combination of RSF and ozonation. Liden et al. [11] compared hollow-fibre ultrafiltration to a combination of clarification, RSF and slow sand filtration and found that ultrafiltration produced lower total emissions. However, the lower GHG emissions were mainly due to operating the ultrafiltration with a different coagulant. Total emissions for membrane filtration can be decreased by optimizing the use of chemicals during CEB [44] or utilising electricity produced from renewable sources [23].

4. Conclusions

Microfiltration produces consistent water quality and can prevail over the conventional treatment process in turbidity and microbe removal. When combined with chemical coagulation, similar NOM removal is expected. Based on the water quality results, microfiltration can supplant a combination of RSF and ozonation at Pitkäkoski WTP. The water quality results also indicated that inline coagulation can be applied, as permeate quality was similar compared to when purifying clarified feed water. However, environmental impact analysis showed that applying the clarification step significantly decreased CO_2-eq. emissions. Pre-treating raw water with coagulation and clarification and operating microfiltration with high flux and a long backwash interval produced the lowest environmental impact. The environmental impact of microfiltration can be comparable to RSF and ozonation. However, the emissions heavily depend on the local electricity production method and usage of chemicals during the CEB. The operational costs of microfiltration are 2.5–3.5 euro cents per m^3 of drinking water and it improves the filtrated water quality compared to conventional treatment. Microfiltration can be a viable option if the treatment plant capacity is increased, as it requires a smaller footprint than RSF and ozonation and can even be retrofitted to existing RSF basins.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w15101822/s1, Table S1: Operational expenses of all studied settings in EUR/m^3, including cost estimation based on energy futures for 2024; Table S2: GHG emissions estimation for every operational setting (CO2-eq./m3).

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Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available due to including treatment-plant-specific information on costs and environmental impacts.

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