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Removal of bacteriophages MS2 and ΦX174 in biotic iron and manganese filters and in ultrafiltration of iron and manganese containing backwash water

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ABSTRACT

Viruses pose risks to human health and might be of special concern in filter backwash water recycling, yet there are no published studies on the subject. The fate of the bacteriophages MS2 and ΦX174 during biological filtration was studied at pilot scale. Furthermore, the efficacy of phage removal in ultrafiltration of filter backwash water was investigated for four different ultrafiltration (UF) membranes at laboratory scale. During biotic iron and manganese removal from groundwater (2.1–2.7 mg/L Fe_{dis} and 0.4–0.7 mg/L Mn_{dis}), both phages were removed by less than one log_{10} unit at filtration velocities of 7.5 m/h and 15.0 m/h. Treatment of filter backwash water (3.9–7.9 mg/L Fe_{total}) with a polyether sulfone flat-sheet UF membrane (pore size 26 nm) yielded a removal of MS2 phages of 4 log_{10} units and of ΦX174 of approx. 3 log_{10} units. Using different UF membranes revealed no clear relation between pore size and phage retention, with phage retention reaching up to 7 log_{10} units with pore sizes larger than phage diameters.

1. Introduction

Deep-bed filtration is a common drinking water treatment process, in which 2 to 10 % of the treated water is lost as filter backwash water (FBW) [1,2]. To reduce water loss, FBW can be recycled into the raw water stream, if the drinking water (DW) quality is not negatively impacted. For a safe recycling, membrane materials including polyamide (PA) [3], polyacrylonitrile (PAN) [3], polyether sulfone (PES) [4], polyvinylidene fluoride (PVDF) [5,6], polypropylene (PP) [7], and polyvinylchloride (PVC) [8] have been examined for FBW recycling in surface water treatment plants. Ceramic membranes made of aluminium oxide (Al_2O_3) and silicon carbide (SiC) were investigated for FBW treatment [9,10] They might be another interesting option for water recycling as they are more resistant to chemicals [11] and organic fouling [9,12].

When viruses accumulate in FBW or enter FBW during collection in open basins, these microbes (that could be pathogens) might be recirculated to the raw water and potentially into DW. With respect to water treatment small viruses are of special interest as they very likely pass the filters more easily. As handling of pathogenic viruses can be difficult in lab settings, bacteriophages such as MS2 and ΦX174 are commonly used as surrogate viruses.

Sand filtration has been studied for the removal of viruses and phages. Aronino et al. [13] compared phage removal from DW and effluents of secondary wastewater treatment via sand filtration at different filter depths. In DW, MS2 and ΦX174 were removed by 52 % (0.32 log_{10} units) and 55 % (0.35 log_{10} units), respectively, at a depth of 100 cm, while in effluent of secondary wastewater treatment, removals of 47 % (0.28 log_{10} units) and 58 % (0.38 log_{10} units), respectively, were observed at the same depth. Retention correlated with organism size as the only relevant factor, while interactions between the phages and the filter material were negligible. Hambsch et al. [14] examined MS2 and ΦX174 retention in sand filters for groundwater (GW) and surface water treatment with and without flocculant addition in lab-scale and pilot-scale experiments. Without flocculant addition, no phage removal was achieved, while using ferric chloride as flocculant, MS2 were removed

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by 1.0 to 5.0 $\log_{10}$ units and ΦX174 by 0.2 to 2.4 $\log_{10}$ units [14]. Using polyaluminium chloride, MS2 were inactivated and ΦX174 were removed by 0.5 to 4.0 $\log_{10}$ units, and for both flocculants, removals were higher in surface water and increased with flocculant dose [14].To improve virus retention in sand filters, Scott et al. [15] coated quartz sand with iron aluminium hydroxides and reported a change in the zeta potential of the filter medium from negative (−40 mV) to positive (45 mV) at a pH between 6 and 9. In lab scale experiments at a filter velocity of 3.6 m/h, there was no MS2 removal in uncoated sand filters, while in metal hydroxide coated sand filters, around 52 % (0.32 $\log_{10}$ units) of spiked phages were removed [15]. You et al. [16] compared removals of MS2 and ΦX174 in lab scale sand filters and zero-valent iron filters shortly after start-up and after 10 days of operation. In the sand filters, no MS2 removal was achieved, while ΦX174 were removed by 49 % after start-up and by 42 % after ten days [16]. This difference in removal between the two phages was proposed to stem from the higher isoelectric point (IEP) of ΦX174 and thus lower net charge of ΦX174 under the experimental conditions, which might have caused weaker electrostatic repulsion between filter material and phages and therefore stronger adsorption due to Van-der-Waals attraction [16]. In contrast to the sand filters, both phages were removed by 4 $\log_{10}$ units at the beginning and by over 5 $\log_{10}$ units after ten days of operation in the zero-valent iron filters, indicating different interactions between the filter and material and the phages [16]. Following a study in which sand aquifer material coated with oxyhydroxides was investigated regarding viruses removal, Ryan et al. [17] proposed that viruses and phages were attached to ferric oxyhydroxides due to electrostatic attraction, then inactivated at the surface and lastly released as non-infectious, disintegrated particles. Nieto-Juarez and Kohn [18] demonstrated that phage removal by different iron minerals in batch tests was mainly caused by adsorption, with only a fraction (7–22 %) being inactivated.

When deep-bed filters are backwashed, viruses might be transferred into FBW. While microfiltration might be insufficient to remove all pathogens during treatment, the usage of nanofiltration or reverse osmosis are not suited due to economic feasibility and particle-rich water matrix of FBW. Ultrafiltration (UF) of FBW, however, showed reduction of total bacteria concentrations to raw water concentrations [5] or even below drinking water limits [3], and complete removal of coliform bacteria [4,5,7]. Viruses are 1 to 2 $\log_{10}$ units smaller than bacteria and might pass through UF membranes. To the authors knowledge, the removal of viruses from FBW has not been studied yet.

For surface water, UF membranes have shown different removals for different viruses such as adenovirus (> 4 $\log_{10}$ units), murine norovirus (around 3 $\log_{10}$ units), and hepatitis A virus (> 5.0 $\log_{10}$ units) [19,20]. Several membrane studies have been conducted with surrogates like MS2 and ΦX174, which are similar to viruses in size (around 30 nm in diameter). However, MS2 and ΦX174 were often removed to different degrees by UF, indicating other characteristics than size to play important roles. ElHadidy et al. [21] studied the removal of both phages from deionized water with a PVDF membrane (hydrophobic, negative surface charge of −55 to −57 mV at pH 7 to 9) at different pH values to investigate the influence of phage size, surface charge, and hydrophobicity on phage removal. At neutral pH, MS2 were removed by 3.7 $\log_{10}$ units, while ΦX174 removals decreased from 4.7 to 3.0 $\log_{10}$ units over three filtration cycles [21]. The removals of both MS2 and ΦX174 were higher than the removal predicted from a mass transport model removals derived from the pore size distribution of the membrane [21]. For MS2, electrostatic repulsion of the negatively charged phages and the negatively charged membrane surface was proposed [21]. For ΦX174, repulsion likely played a smaller role as they carry no charge at neutral pH, and thus adsorption to the membrane surface was proposed [21]. Furthermore, ΦX174 removals decreased over multiple filtration cycles, and it was hypothesized that adsorption sites on the membrane surface were occupied [21].

In a recent study on wastewater treatment using a ceramic dead-end membrane (α-Al_2O_3), Ahmadi et al. [22] examined the effects of PACl coagulant, membrane surface condition, and operation time on the removal of MS2 and ΦX174 bacteriophages. They observed high MS2 removal (~8 LRV) with pristine membranes, though performance declined over time. PACl enhanced MS2 removal, likely by increasing the virus's surface charge via Al^{3+}, promoting aggregation [22]. These larger aggregates were more effectively removed through size exclusion and hydrophobic interactions with the membrane. ΦX174 removal also decreased over time, starting at 6–8 LRV with moderate PACl doses and pristine membranes, but falling below 2 LRV later [22]. However, PACl had a lesser effect on ΦX174, possibly due to differences in hydrophobicity or aggregation behaviour. With pre-fouled membranes, fouling dominated phage removal, obscuring the roles of electrostatic interactions and aggregation [22]. Overall, MS2 removal exceeded that of ΦX174, likely due to MS2's greater hydrophobicity and stronger adhesion to the fouled layer [22]. Furthermore, MS2 retention has been shown to increase with increasing flux and transmembrane pressure to enhanced aggregation [23].

Stored filter backwash water, especially if the water is exposed to the environment, might be contaminated by animals and recycling within the drinking water treatment chain could cause a risk to human health. The overreaching aim of this study is to mitigate the risk to human health caused by pathogen contamination of FBW and subsequent recycling to the influent of the drinking water treatment plant. The objective is to evaluate ultrafiltration as a barrier for pathogens, focussing on bacteriophages as surrogates for human viruses. The research focus so far was FBW recycling from surface water treatment plants. Investigation of the retention of viruses in GW filters in combination with FBW recycling after treatment was identified as research gap and is the focus of this study. FBW from the GW filters was treated using PES membranes to evaluate phage retention, and differences in phage removals using different UF membranes were studied in regards to pore size, surface charge, hydrophobicity, and water matrix.

2. Material and methods

2.1. Phages and phage analyses

MS2 and ΦX174, which are commonly used as model organisms for non-enveloped viruses like adenoviruses and noroviruses (characteristics see Table 1), were cultivated and analysed by double layer agar methods according to International standards [24,25]. Feed samples were diluted up to factor 10^{-4}, while permeate samples were analysed undiluted. The (diluted) samples were mixed with a host culture (Table 1) in liquid agar, and poured onto modified scholten agar agar (ΦX174) or tryptone yeast glucose agar (MS2, ΦX174) plates. Plaques were counted after an incubation period of 18 (± 2) h at 36 (± 2)°C, and phage concentrations were determined in plaque forming units (PFU) with eq. 1.

$$c_{phage} = \frac{N}{\sum_{1}^{i} n_i V_i F_i} \tag{1}$$

c_{phage}: phage concentration (PFU/mL)
N: counted number of plaques (−)

Table 1
Phage characteristics according to ElHadidy et al. (2013) and host bacteria.

Phage	Outer-capsid diameter	Genetic material[a]	IEP	Host bacterium
MS2	27.4 nm	ssRNA	3.3	*E. coli* WG5
ΦX174	27.6 nm 33 nm including spikes	ssDNA	6.6	*Salmonella typhimurium* WG49

[a] ssRNA: single-stranded ribonucleic acid, ssDNA: single-stranded deoxyribonucleic acid.

n: number of repeat measurements per dilution (−)

F: dilution factor (−) (e.g. 0.1 for 10 times diluted, 0.001 for 1000 times diluted)

V: sample volume (mL).

The logarithmic removal value (LRV) was calculated with eq. 2.

$$LRV = log_{10}\left(\frac{c_0}{c}\right) \tag{2}$$

c_0: phage concentration at the beginning of the experiment (in the feed) (PFU/100 mL)

c: concentration at the end (in the permeate) (PFU/100 mL).

If the measured feed concentration differed from the calculated feed concentrations by more than 1 $\log_{10}$ unit, the calculated concentration was used to determine LRV.

2.2. *Pilot plant for GW treatment*

The fate of phages during DW treatment was studied in a pilot plant for biological iron and manganese removal (Fig. 1). Two filter columns with expanded polystyrene filter material were operated up-flow in series. The influent of each filter was aerated with technical oxygen. The oxygen concentration in front of the first filter was regulated according to the redox potential (target value of 30 mV) and was between 0.00 and 0.51 mg/L. The oxygen concentration before the second filter was set to ca. 0.84 mg/L. The control system was setup to ensure that the first filter removed mainly iron, and the remaining iron and the manganese were removed in the second filter. Samples were collected at different points along the pilot plant to monitor iron, manganese, and phage removal. More detailed descriptions of the system were provided by [26,27].

2.3. *Membrane test rigs*

Retention of phages by membranes was tested in three different lab-scale test rigs (Fig. 2). The test rig LSta80 (SIMA-tec, Schwalmtal, Germany) for flat-sheet ultrafiltration membranes consists of stainless-steel components and flexible tubing (PTFE and FEP) (Fig. 2A). The test rig was operated in cross-flow mode (cross-flow velocity 0.16, 0.37, or 0.62 m/s) with flat sheet membranes (membrane area 85 cm^2). Experiments were run using a modified PES membrane with a molecular weight cut-off (MWCO) of 15 kDa (Mann+Hummel, Ludwigsburg, Germany) at a temperature of 15 °C.

The second test rig was operated in dead-end mode under negative pressure with submerged out-in membranes (PVDF or SiC) (Fig. 2B). The ultrafiltration occurred in a T-shaped acrylic container, where two stirrers homogenized the test water in the upper part of the container and the membrane was situated in the lower part. Additional test water was continuously supplied from a mixed refill container. The T-shaped container was equipped with a water cooler, experiments were run at 15 °C, and the permeate flow was continuously monitored for pressure and volume flow. The third test rig (Convergence Inspector Hydra, Demcon Convergence, Enschede, Netherlands) was operated pressure-driven in dead-end mode with in-out filtration (Fig. 2C). Permeate was collected behind the membrane and monitored for pressure and flow. The samples of test rigs B and C were analysed on site for all monitored parameters except for the phage concentrations. Membrane characteristics for the membranes used in this study are summarized in Table 2. Test rigs B and C were backwashed with permeate at 200 L/(m^2•h). The membrane areas employed were comparable between all test rigs.

2.4. *Fate of phages during GW treatment*

In three filtration experiments, MS2 and ΦX174 were spiked separately into the influents of the pilot plant for iron and manganese removal. An overview of the filtration experiments at the pilot plant for GW treatment is given in Table 3.

Samples were analysed for infectious phage concentration (in PFU), Fe, Mn, and organic carbon. At the end of each experiment, both filters were backwashed separately with treated water and the FBW was collected for further experiments.

2.5. *Impact of FBW and membrane characteristics on phage removal*

The influence of flux, water matrix and membrane characteristics on phage removal was examined in a number of experiments (Table 4). Removal of MS2 phages at different membrane fluxes was studied at test rig A with a flat-sheet membrane (experiments PES-1, PES-2, and PES-3). Fe-FBW containing MS2 was treated at fluxes of 25, 65, and 100 L/(m^2•h) for one day each. Feed and permeate samples were taken at the beginning and at the end of each day.

The influence of FBW origin on ΦX174 removal was studied at test

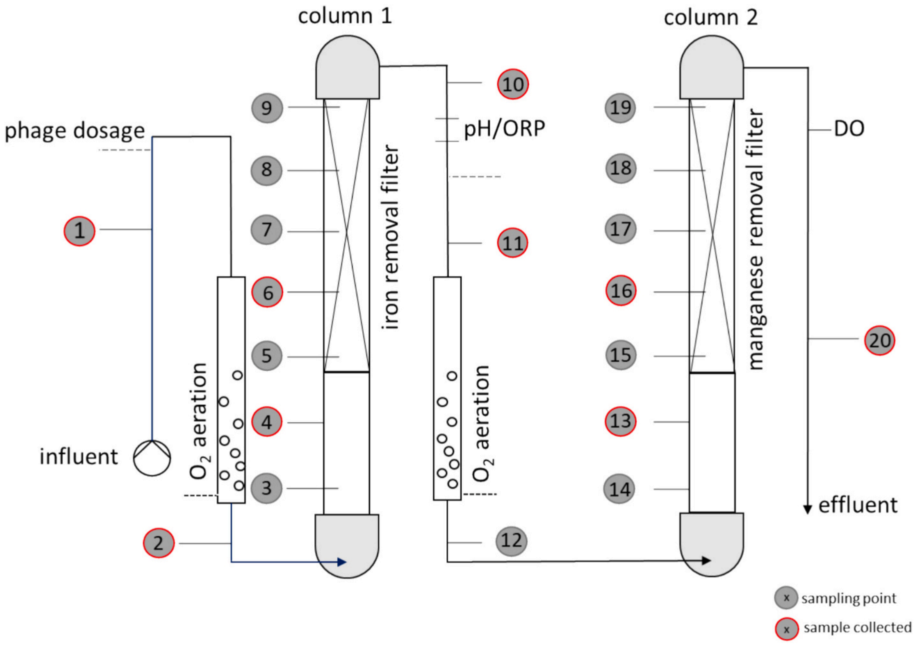

Fig. 1. Scheme of the pilot plant for biological iron and manganese removal, adapted from Mahringer et al. (2020).

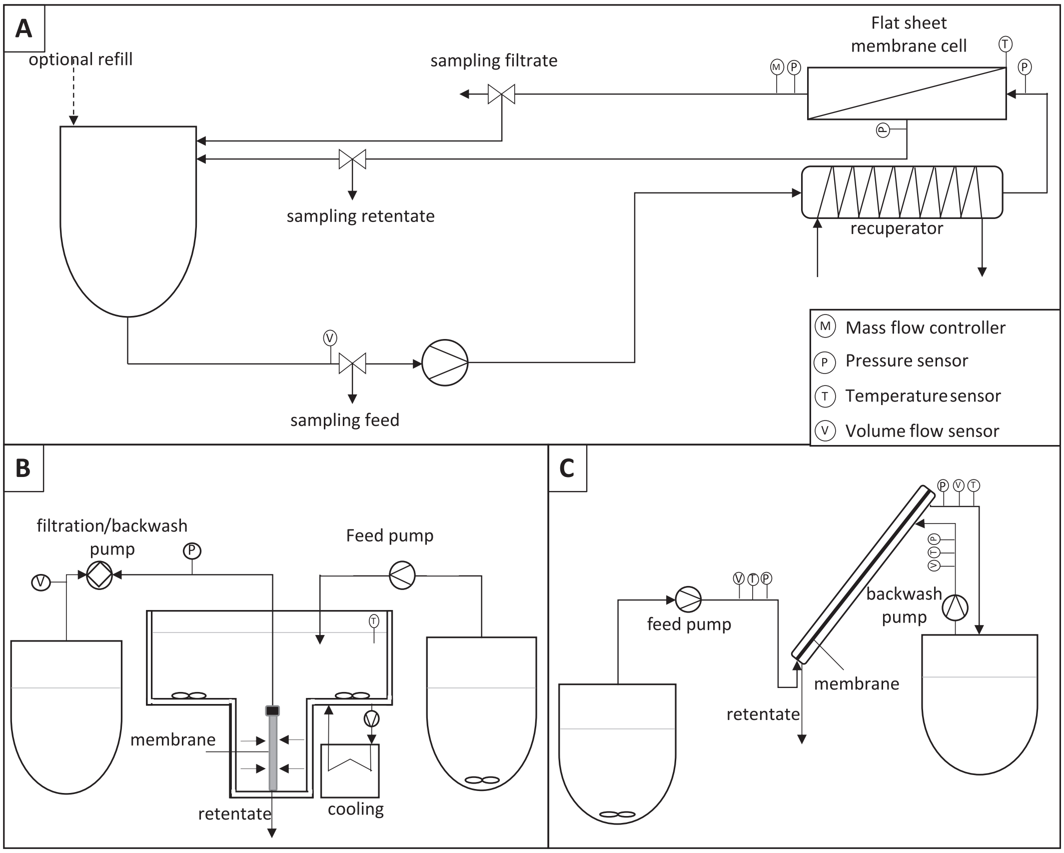

Fig. 2. Schemes of ultrafiltration devices for (A) flat-sheet cross-flow membranes, (B) submerged dead-end membranes, and (C) in-out dead-end membranes.

Table 2
Characteristics of all membranes employed throughout all membrane experiments.

Material	PES	PES	SiC	PVDF
Brand and manufacturer	Multibore® 1.5 (Inge DuPont)	UP 150 P (Mann+Hummel)	Cembrane	ZeeWeed 500 (Veolia)
Module type	Hollow fibre	Flat sheet	Plate	Hollow fibre
Pore size	20 nm[a]	26 nm[b]	100 nm[a]	40 nm[a]
MWCO	82 kDa[b]	150 kDa[a]	370 kDa[b]	270 kDa[b]
Zeta potential at pH 7	−52 [28]	−36 mV [29]	−58 [28]	−36 [28]
Mode of filtration	Dead-end In-out	Cross-flow	Dead-end Out-in	Dead-end Out-in
Membrane area	0.0086 m^2	0.0085 m^2	0.0103 m^2	0.0107 m^2

[a] Manufacturer information.
[b] calculated according to [30]: pore diameter (nm) = 0.11•MWCO0.46 with MWCO in Da.

Table 3
Overview of filtration experiments.

Experiment	Phage	Filtration velocity (m/h)	Sampling times (h after start of experiment)	Duration (h)
MS2–1	MS2	15.0	2, 24	24
MS2–2	MS2	7.5	2, 24	24
ΦX174–1	ΦX174	7.5	2, 24	24

rig A using a flat sheet PES membrane (experiments PES-4 and PES-5). Fe-FBW and Mn-FBW containing ΦX174 was treated at 100 L/(m^2•h) for one hour. Tests were conducted in duplicate and samples were taken at the beginning and at the end of the filtration period. Membranes were cleaned with membrane cleaner (P3-ultrasil 075 containing HNO_3 and H_2SO_4, SIMA-tec GmbH, Schwalmtal, Germany) and ultrapure water between duplicate runs, and exchanged when the feed water was exchanged. Each new membrane was conditioned with ultrapure water in the test rig overnight before the start of the experiment.

The influence of membrane characteristics on the removal of MS2 and ΦX174 from FBW was tested in experiments PES-6, PES-7, PVDF-1, PVDF-2, SiC-1 and SiC-2. Membranes were operated for one hour at 100 L/(m^2•h) using the test rigs B (experiments PVDF-1, PVDF-2, SiC-1 and SiC-2) and C (experiments PES-6 and PES-7) (Fig. 2B and C) with sampling at the beginning and end of each experiment. Each membrane experiment was conducted as duplicate carried out within one day. Membranes were cleaned with hydrochloric acid and sodium hypochlorite between runs. PES membranes were flushed in-situ for 10 min with each solution, while PVDF and SiC membranes were removed from the test rig and soaked for 30 min in each solution. Membranes were rinsed with ultrapure water until the pH value was stable before resuming experiments. The FBW was produced by diluting concentrated FBW (2 g/L TSS) from a full-scale water treatment plant (production of drinking water from groundwater in Hamburg, Germany) with tap water

Table 4
Overview of UF membrane experiments.

Experiment	Flux (L/(m^2•h)	Water	UF-membrane	Test rig	Phage
PES-1	25	Fe-FBW	PES flat sheet	A	MS2
PES-2	60	Fe-FBW	PES flat sheet	A	MS2
PES-3	100	Fe-FBW	PES flat sheet	A	MS2
PES-4	100	Fe-FBW	PES flat sheet	A	ΦX174
PES-5	100	Mn-FBW	PES flat sheet	A	ΦX174
PES-6	100	FBW	PES hollow fibre	C	ΦX174
PES-7	100	FBW	PES hollow fibre	C	MS2 + ΦX174
PVDF-1	100	FBW	PVDF hollow fibre	B	ΦX174
PVDF-2	100	FBW	PVDF hollow fibre	B	MS2 + ΦX174
SiC-1	100	FBW	SiC plate	B	ΦX174
SiC-2	100	FBW	SiC plate	B	MS2 + ΦX174
PES-8	100	DW	PES hollow fibre	C	ΦX174
PES-9	100	DW	PES hollow fibre	C	MS2 + ΦX174
PVDF-3	100	DW	PVDF hollow fibre	B	ΦX174
PVDF-4	100	DW	PVDF hollow fibre	B	MS2 + ΦX174
SiC-3	100	DW	SiC plate	B	ΦX174
SiC-4	100	DW	SiC plate	B	MS2 + ΦX174

to 200 mg/L TSS, which is a typical concentration for FBW from that water treatment plant. Phage concentrations in the feed were adjusted by adding 1 to 2 mL phage stock suspension per litre feed. In membrane experiments PES-6, PVDF-1, and SiC-1, a ΦX174 stock suspension of 2•10^{8} PFU/100 mL was diluted to a feed concentration of 1.2•10^{6} PF/100 mL in FBW. In experiments PES-7, PVDF-2, and SiC-2, a ΦX174 stock suspension of 10^{12} PFU/100 mL and MS2 stock suspension of 2•10^{12} PFU/100 mL were diluted to feed concentrations of 9•10^{8} PFU/100 mL for ΦX174 and 2•10^{9} PFU/100 mL for MS2 in FBW. Spiked FBW was divided into thirds for the experiments PES-6, PVDF-1, and SiC-1, and experiments PES-7, PVDF-2, and SiC-2. Experiments were carried out on three consecutive days using one third of this feed stock per experiment and day.

The influence of the filter cake on MS2 and ΦX174 removal was examined using DW in experiments PES-8, PES-9, PVDF-3, PVDF-4, SiC-3 and SiC-4. Experiments were run analogous to experiments using FBW as water matrix.

2.6. Water analyses

Samples were analysed immediately for Fe, Mn, turbidity, and organic carbon (DOC and TOC). Phage samples from experiments PES-6, PES-8, PVDF-1, PVDF-3, SiC-1, and SiC-3 were stored at 4 °C and shipped over-night for immediate analysis. Phage samples of all other experiments were immediately frozen at −80 °C and sent to the analysing lab where they were thawed as a set and analysed.

Dissolved iron (Fe$_{\mathrm{dis}}$) and dissolved manganese (Mn$_{\mathrm{dis}}$) were determined photometrically after a colour reactions (Spectroquant® Iron or Manganese test kit, Merck KGaA, Darmstadt, Germany) using photometers (either Lambda35, Perkin Elmer, Waltham, Ma, USA or DR 5000 UV/VIS, Hach Lange, Düsseldorf, Germany) at a wavelength of 562 nm (Fe) or 505 nm (Mn). For total iron (Fe$_{\mathrm{tot}}$), samples were pre-treated (Spectroquant® Crack Set 10, Merck KGaA, Darmstadt, Germany) before measuring.

Total organic carbon (TOC) and dissolved organic carbon (DOC) were determined with carbon analysers (either vario TOC cube, Elementar Analysensysteme GmbH, Langenselbold, Germany, or TOC-L Total Organic Carbon Analyzer, Shimadzu, Kyoto, Japan), after acidification with HCl.

For DOC, Fe$_{\mathrm{dis}}$, and Mn$_{\mathrm{dis}}$ analyses, samples were filtered with syringe PET-filters (0.45 µm pore size, Chromafil, Macherey-Nagel, Düren, Germany). Turbidity was measured using a Turb® 555 turbidity photometer (WTW Xylem Analytics, Weilheim, Germany), pH was measured with a SenTix® electrode (WTW Xylem Analytics, Weilheim, Germany). TSS was determined gravimetrically and calculated with eq. 3.

$$TSS = \frac{m_{loaded} - m_{empty}}{V} \tag{3}$$

TSS: Total suspended solid concentration (mg/L)

m_{empty}: weight of filter before filtration (mg)

m_{loaded}: weight of filter after filtration and drying (overnight at 105 °C) (mg)

V: volume of the homogenized sample (L)

2.7. Data processing

For all samples and analytes, values below the limit of quantification (LOQ) were conservatively set to the LOQ. Mean values and standard deviations of phage removal were calculated after exclusion of outliers according to the Dixon test [31].

Feed samples were grouped per experiment, and permeate samples were grouped per experiment, membrane, and sampling time. If the sampling time did not significantly impact phage concentrations, permeate samples were grouped by experiment and membrane. All sample groups were checked for normality of variance according to the Levene test, and depending on the outcome, the appropriate test was chosen between either student-*t*-test and Welch test for two groups of samples, and ANOVA or Welchs ANOVA was used for comparison of three groups. Three-way comparisons were corrected with a Bonferroni-adjustment. All statistical tests were applied at a confidence level of 0.05 and conducted using the R outliers package and R Stats package.

It must be noted that the sampling size used for statistical evaluation was small, and in some cases, repeat measurements resulted in the same concentration (e. g., no PFU detected would be considered as the LOQ). In these cases, the calculated LRV were nearly identical and hence the standard deviations were almost identical. This small difference along with the small sampling size diminished the power of the statistical tests, so the results from these tests should be understood as indicators.

3. Results and discussion

3.1. Water quality parameters in filter and membrane experiments

The pilot plant was fed from different GW wells, so the composition of the raw water varied in terms of Fe$_{\mathrm{dis}}$ and Mn$_{\mathrm{dis}}$ concentrations (Table 5). In all experiments, Fe$_{\mathrm{dis}}$ and Mn$_{\mathrm{dis}}$ concentrations decreased significantly in the pilot plant, with Fe$_{\mathrm{dis}}$ being removed to values below the limit of quantification.

FBW from experiments MS2–1 and ΦX174–1 were used for membrane experiments PES-1 to PES-5. For all other experiments with FBW,

Table 5
Fe$_{\mathrm{dis}}$ and Mn$_{\mathrm{dis}}$ in raw water used for filtration experiments.

	MS2–1		M2S-2		ΦX174–1	
	Influent	Effluent	Influent	Effluent	Influent	Effluent
Fe$_{\mathrm{dis}}$ (mg/L)	2.16	< 0.03	2.07	< 0.03	2.7	< 0.03
Mn$_{\mathrm{dis}}$.(mg/L)	0.54	0.05	0.77	0.40	0.65	0.07

FBW was produced by mixing DW with a FBW stock. Water characteristics are summarized in Table 6. The waters used differed in their total Fe content, which was present as particles and could have impacted phage removal through the formation of a cake layer on the membrane. Dissolved parameters were similar across the different feed waters used.

3.2. Fate of phages during rapid filtration

The spiked phage concentration in the raw water was $2.2–7.4 \bullet 10^5$ PFU, which is higher than virus concentrations found in studies done in Barcelona, Spain (around $1 \bullet 10^{-5}$ gene copies per mL of the human adenovirus) [32] and Perth, Australia (no coliphages detected) [33], and similar to concentrations found in a study done in Colorado, USA ($8 \bullet 10^4–1 \bullet 10^6$ viruses, defined as DNase resistant, SYBR Green I fluorescent particles between 20 nm and 0.2 μm) [34]. Independent of filtration velocity, MS2 were not retained (0.01–0.1 $\log_{10}$ units) in the filters for biotic iron and manganese removal and phage concentrations in the effluent and in the FBW did not differ significantly from the spiked concentration in the raw water (Fig. 3). ΦX174 were removed by 0.9 $\log_{10}$ units at 7.5 m/h in the pilot plant (Fig. 3). However, the difference between influent and effluent concentrations was not significant ($p =$ 0.3). The concentration in the Mn-FBW was significantly ($p < < 0.05$) higher ($4 \bullet 10^5$ PFU) than the spiked raw water concentration ($3 \bullet 10^5$ PFU) and the effluent ($4 \bullet 10^4$ PFU), indicating a slight accumulation of phages in filter 2, most likely through adsorption, and a slight remobilisation of adsorbed phages during filter backwash. These results were in line with other studies, which have found rapid filtration to be an insufficient barrier for viruses, with removals being below one $\log_{10}$ unit [13,35,36].

The rapid filter used in this study employed polystyrene filter material covered with ferric oxide-hydroxides and manganese oxide-hydroxides after years of use and fresh precipitates formed during the filter experiments in this study. Ryan et al. [17] determined that viruses and phages adsorb onto ferric oxide-hydroxides due to electrostatic attraction, where they are subsequently inactivated at the surface and released as non-infectious, disintegrated particles. Nieto-Juarez and Kohn [18] showed MS2 removals above 3 $\log_{10}$ units within 2 h through adsorption onto amorphous ferric hydroxide ($Fe(OH)_3$). Scott et al. [15] showed an increase of MS2 removal by 0.22 $\log_{10}$ units by coating sand with ferric and aluminium chloride compared to uncoated sand. In this study, however, no MS2 removal and only 0.9 $\log_{10}$ units removal of ΦX174 were achieved using microbially produced iron and manganese oxyhydroxides. Potentially the surface characteristics differed from commercially available ferric materials used in other studies. Furthermore, the filter velocity of 7.5 m/h was comparably high, which might have led to reduced phage adsorption. Virus and phage adsorption onto manganese oxyhydroxides has not been studied as of yet. Column 2 of the pilot plant contained both iron oxide-hydroxides and manganese oxide-hydroxides. Consequently, the removal in column 2 cannot be attributed to manganese oxide-hydroxides alone and no clear adsorption onto manganese oxide-hydroxides can be derived from this data.

Table 6
Overview of water characteristics in membrane experiments.

	Unit	Fe-FBW	Fe-FBW	Mn-FBW	FBW	DW
Experiments		PES-1, PES-2, PES-3	PES-4	PES-5	PES-6, PES-7 PVDF-1, PVDF-2 SiC-1, SiC-2	PES-8, PES-9 PVDF-3, PVDF-4 SiC-3, SiC-4
TSS	mg/L	–	–	–	243.17	–
Turbidity	NTU	–	–	–	895.87	0.57
Fe_{total}	mg/L	7.86	3.88	17.76	80.72	–
Fe_{dis}	mg/L	0.03	0.10	0.13	0.04	–
Mn_{dis}	mg/L	0.46	0.08	0.03	–	–
pH	–	–	–	–	8.00	8.00
TOC	mg/L	–	–	–	20.51	0.50
DOC	mg/L	–	–	–	7.75	–

3.3. Influence of membrane flux and FBW origin on phage removal

During UF treatment of FBW in experiment PES-1, PES-2, and PES-3, no MS2 were detected in the permeate at any point during the experiment independent of the flux, resulting in a LRV of 3 to 4 $\log_{10}$ units (see Figure SI1 in the supplementary information). Further experiments were conducted at a flux of 100 L/(m^2•h).

In contrast to PES-3, where no MS2 were detected, in experiments PES-4 and PES-5, ΦX174 were determined in all permeate samples with LRV ranging from 1.9 to 3.2 $\log_{10}$ units (Fig. 4). ΦX174 removal was slightly higher in the first duplicate run than in the second run for both tested FBW. In the first run with Fe-FBW (PES-4) the removal was 1.3 $\log_{10}$ units ($p = 0.005$) higher than in the second run and with Mn-FBW the difference in removal was 0.8 $\log_{10}$ units ($p = 0.01$) between duplicate runs. The same membrane was used for duplicate runs, which could have influenced adsorption capacity in the second run. Though total iron concentrations varied greatly between the experiments, there was no significant difference in phage removals between the two FBW.

The difference of MS2 and ΦX174 removal via UF has been previously noted. While Arkhangelsky and Gitis [37] only showed a slight difference in removals using PES membranes (LRV of up to 3.18 $\log_{10}$ units for MS2 and of up to 3.15 $\log_{10}$ units for ΦX174), Kreißel et al. [38] showed a slightly higher difference of removals (up to 6 $\log_{10}$ units for MS2 and up to 4.5 $\log_{10}$ units for ΦX174). As the phages are similar in size, other mechanisms than size exclusion must have influenced the removal, such as electrostatic interactions [21,39]. The IEP of MS2 is lower than that of ΦX174 (Table 1), resulting in a more negative surface charge at neutral pH. PES membranes have a slightly negative surface charge [40], which might have caused a higher repulsion of MS2 than ΦX174 at the membrane surface, resulting in a higher removal. Furthermore, iron has been shown to adsorb and inactivate MS2 [17,18], which could have contributed to higher MS2 removal compared to ΦX174 removal.

In membrane experiments PES-4 and PES-5, the ΦX174 retention decreased between the first and the second duplicate run. Between these runs, reversible fouling was removed by rinsing the membrane with clean water. It is possible that adsorbed phages were not removed in this cleaning step, and the adsorption capacity of the membrane was already diminished at the beginning of the second run. The transmembrane pressure (TMP) at the start of each run was around 0.3 mbar with the exception of the second run using Mn-FBW, where the TMP was 0.1 mbar at the start. The TMP indicates membrane fouling, which in this case did not correlate with phage removal. ElHadidy et al. [21] described a drop in ΦX174 retention by PVDF hollow fibre membranes over time, ascribing the drop to the blockage of easily accessible adsorption sites.

3.4. Influence of water matrix on phage analyses

In DW, measured feed concentrations matched up well with initial concentrations calculated from the dilution of the stock suspension in the feed (Fig. 5). In FBW, measured feed concentrations were about 1 $\log_{10}$ units lower than the calculated concentrations in experiments PES-6, PVDF-1, and SiC-1, and 2 to 4 $\log_{10}$ units lower than calculated in experiments PES-7, PVDF-2, and SiC-2. Components in the FBW, such as ferric hydroxides and calcium, likely interacted with the phages and thereby might have influenced the recovery from FBW feed samples.

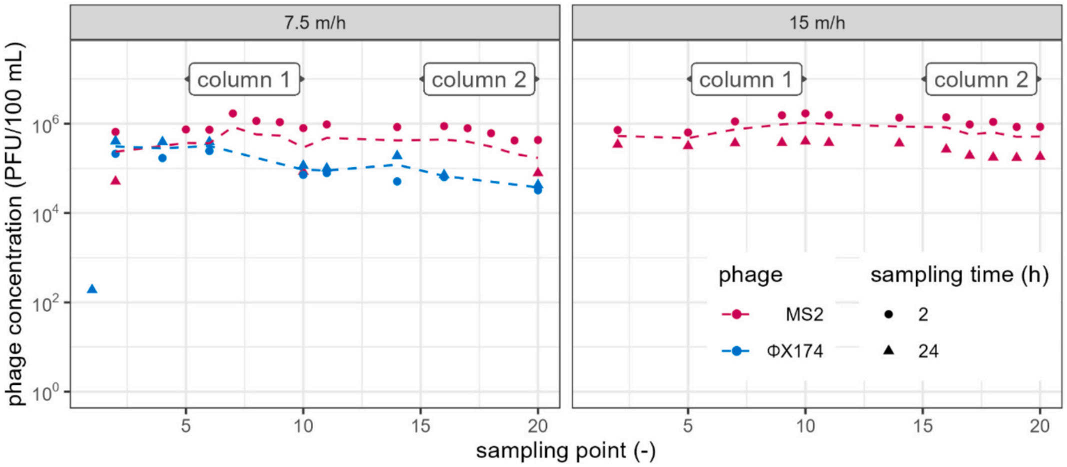

Fig. 3. Phage concentrations along the pilot plant with mean values of samplings after 2 h and 24 h depicted as dashed line.

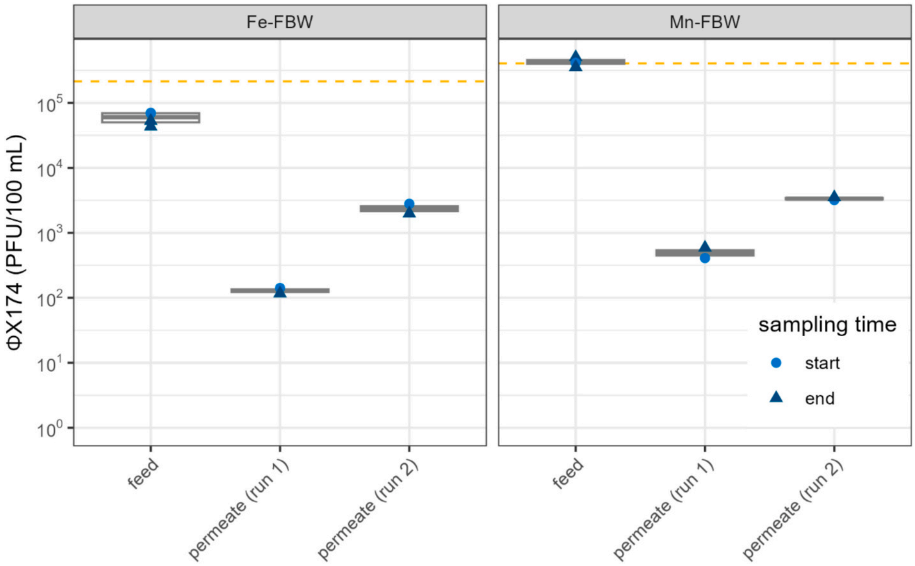

Fig. 4. Box-Whisker-Plot for ΦX174 of feed and permeate samples during UF filtration with flat-sheet PES membrane, overlayed with analysed concentration coded by sampling time point (start and end of the run) and separated for duplicate runs. Dashed lines depict starting concentrations as determined in backwash water right after filter backwash.

Ferric hydroxides have been shown to adsorb and inactivate MS2 phages [17,18], while calcium as Ca^{2+} was shown to form complexes with MS2 [41]. For a more accurate comparison of phage retention in both water matrices, calculated initial phage concentrations were used to determine LRV for all experiments.

3.5. Influence of membrane characteristics on phage removal from FBW

Between membrane experiments PES-6, PVDF-1, and SiC1, and experiments PES-7, PVDF-2, and SiC-2, ΦX174 removal differed when comparing different sampling times in a few cases, and the two experiments were kept separate in the discussion (Fig. 5). Differences between the retention of MS2 and ΦX174 from FBW were compared for experiments PES-7, PVDF-2, and SiC-2, and from DW in experiments PES-9, PVDF-4, and SiC-4. No significant difference in LRV of both phages was found for any tested membrane.

In most cases, the permeate concentrations at different sampling times did not differ significantly from each other, and were pooled for discussion (displayed as permeate (total)). However, if results differed between the start and end of the experiment, the data is shown separately (permeate (start) and (end)). Removals differed between the start and end in a few experiments, showing significant differences in ΦX174 retention in DW with PVDF ($p = 0.001$) in experiment PVDF-4, and in FBW with SiC ($p = 0.04$) in experiment SiC-4 (Fig. 5). In both cases, LRV was higher at the beginning than at the end (2.8 and 2.2 $\log_{10}$ units, respectively). Additionally, MS2 retention in DW with PVDF was higher at the beginning (6.8 $\log_{10}$ units) than at the end (4.3 $\log_{10}$ units) of the filter cycle ($p = 0.007$) in experiment PVDF-4 (Fig. 6). ElHadidy et al. [21] have shown that ΦX174 retention decreased over time as easily available adsorption sites on the membrane surface became occupied.

Several studies have found a correlation between membrane pore size and LRV for UF membranes [20,42,43]. The membranes used in this study varied not only in material, but also in pore size. The PES membrane had the smallest pore size of about 20 nm, and was the only membrane with pores smaller than the geometric diameter of the phages (Tables 1 and 2). The PVDF membrane had slightly larger pores (40 nm) and the SiC membrane had the largest pores of 100 nm. Based on pore size, it was hypothesized that PES should show the highest removal and SiC the least, but this hypothesis was negated. In terms of membrane

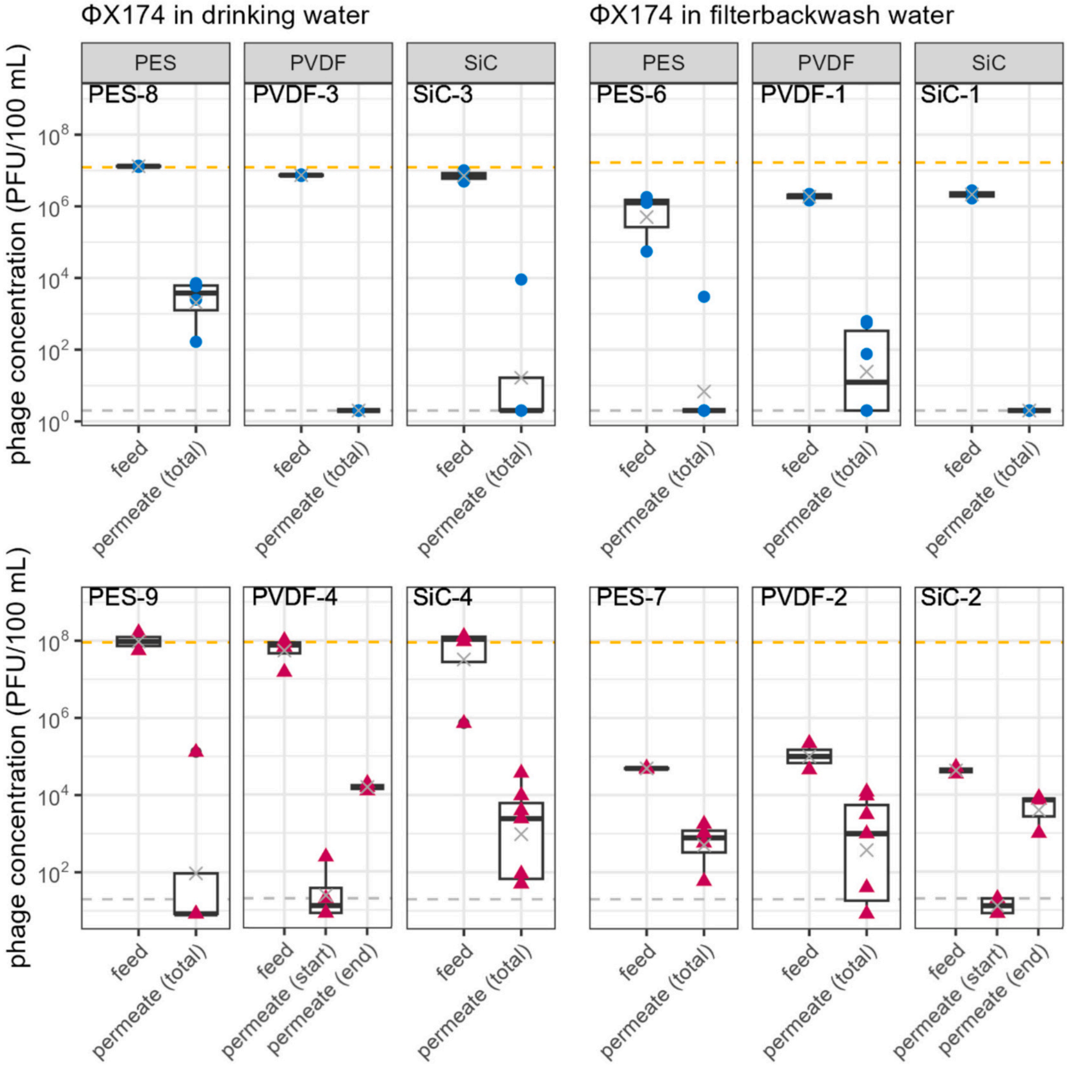

Fig. 5. ΦX174 concentrations (as PFU) over Box-Whisker-Plots per sampling point, with a grey cross marking the mean value. Permeate samples from two tie points were pooled, except in experiments were there was a significant difference in LRV according to the Student *t*-test. The yellow dashed line depicts the target concentration, the grey dashed line depicts the LOQ (2 PFU/mL to 20 PFU/mL depending on the analysis lab). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

material, no significant differences in LRV were shown for MS2 removals regardless of water matrix (4.7–5.3 $\log_{10}$ units in FBW and 5.7–6.1 $\log_{10}$ units in DW). MS2 removal was higher than in experiments done with comparable membranes by Hambsch et al. [42] and Arkhangelsky and Gitis [37], who achieved MS2 removals of 2.6 $\log_{10}$ units and 3.8 $\log_{10}$ units, respectively. In experiments were ΦX174 were examined without MS2 (PES-6, PES-8, PVDF-1, PVDF-3, SiC-1, and SiC-3), phages were significantly ($p = 0.01$) better removed from DW using the PVDF membrane (LRV 6.8 $\log_{10}$ units) compared to the PES membrane (LRV 3.8 $\log_{10}$ units), while the removal with the SiC membrane was not significantly different from either (LRV 5.9 $\log_{10}$ units). In experiments were both phages were added to the feed simultaneously (PES-7, PES-9, PVDF-2, PVDF-4, SiC-2, and SiC-4), there were no significant differences in LRV for ΦX174 removal between the three membranes. Therefore, pore size did not correlate with removal, and other factors such as electrostatic and hydrophilic interactions must have influenced phage removal. All three factors, size exclusion, electrostatic interaction and hydrophilic adsorption, might also be influenced by fouling [19,38,41] and are discussed for each membrane individually in the following paragraphs.

3.6. *Phage removal with PES membranes*

As the membrane pore size (20 nm, Table 2) was smaller than the size of each phage, complete removal was expected, but not observed (Figs. 5 and 6). The pore size is usually determined from scanning electron microscope (SEM) data or the MWCO, which is given by the substance that gives a 90 % rejection [44]. Accordingly, there are some pores that allow substances of greater molecular weight to pass the membrane.

The PES membrane has a zeta-potential of −52 mV at pH 7 (Table 2), and PES is generally considered hydrophobic [37,40,45]. ΦX174 has an IEP of 6.6 and is therefore nearly neutral or slightly negative at neutral pH [46], and was reported to be slightly hydrophilic [13,47]. Electrostatic repulsion of the membrane should be low due to the near-neutrality of ΦX174, and hydrophilic adsorption should not be expected on a hydrophobic membrane. Thus, size exclusion was likely the dominant removal mechanism for ΦX174 removal with PES membranes. In membrane experiments PES-6, ΦX174 removal from FBW was significantly higher than removal from DW in experiment PES-8 by 3.5 $\log_{10}$ units ($p = 0.007$). Particles in the water might have blocked pores which might have increased the removal further through size exclusion [46]. To conclusively investigate the impact of pore size and surface charge on phage removal, further experiments with PES membranes of

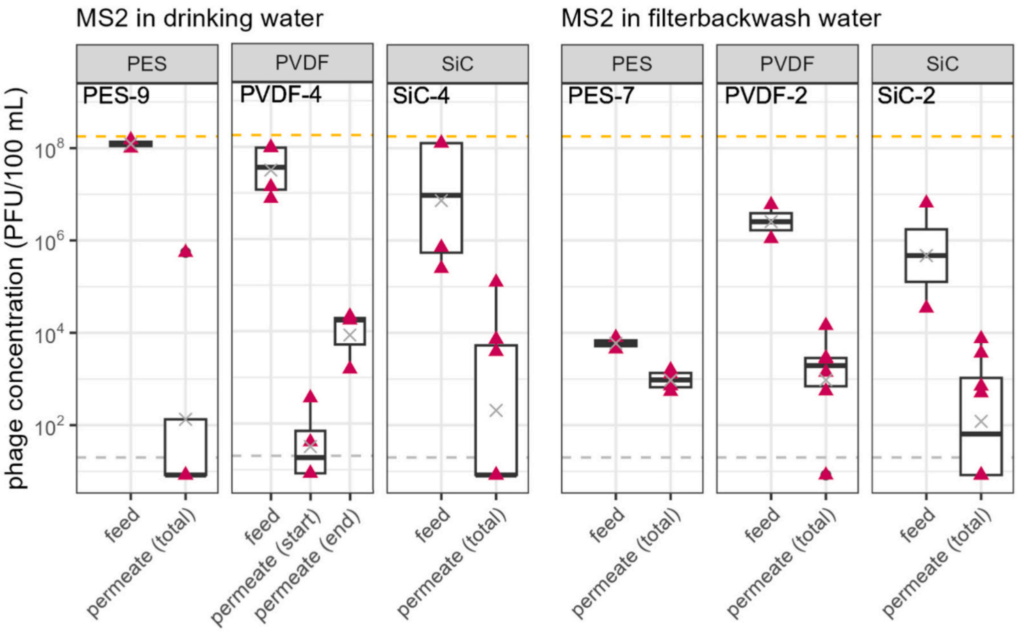

Fig. 6. MS2 concentration (as PFU) over Box-Whisker-Plots per sampling point, with a grey cross marking the mean value. Permeate samples from two tie points were pooled, except in experiments were there was a significant difference in LRV according to the Student *t*-test. The yellow dashed line depicts the target feed concentration, the grey dashed line depicts the LOQ. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

different pore sizes as well as removal at different pH values are necessary.

MS2 has an IEP of 3.5 [21], which makes it more negative at neutral pH, and is considered hydrophobic [48]. Besides size exclusion, electrostatic repulsion could have been a relevant removal mechanism for MS2. As the removal difference between the two phages was not significant (LRV in FBW 5.3 $\log_{10}$ units and 4.9 $\log_{10}$ units for MS2 and ΦX174, respectively, in experiment PES-9), size exclusion was probably the main removal mechanism.

3.7. Phage removal with PVDF membranes

The PVDF membrane was the least negatively charged of this study, with a zeta-potential of −36 mV, and is considered hydrophobic in literature (Table 2) [21,45].

As with PES, no hydrophilic adsorption was expected, and electrostatic repulsion should have had a negligible effect on ΦX174 removal. The geometric size of ΦX174 is smaller than the average pore size of the PVDF membrane. However, the hydrodynamic size of ΦX174 is around 60–80 nm and therefore bigger than the average pore size of the PVDF membrane [13]. Consequently, the hydrodynamic size might have caused a better removal than expected from the geometric size. Electrostatic repulsion of MS2 from PVDF should have been less pronounced than from PES due to the membrane surface charge. Therefore, LRV for MS2 of the PES membrane was expected to be higher than of the PVDF membrane. However, due to less repulsion, MS2 could possibly get closer to the membrane surface and adsorb, as both phage and membrane are hydrophobic, which likely increased LRV and could explain the similar LRV compared to PES membranes. As with PES membranes, experiments with PVDF experiments of different pore sizes and at different pH values would help elucidate removal mechanisms.

It was hypothesized that fouling would increase phage removal through complete pore blockage or attachment inside the pores, effectively decreasing pore diameter. However, removal did not differ significantly ($p > 0.05$) between drinking water and FBW. Indeed, mean removal values were higher in drinking water in most experiments (6.8 $\log_{10}$ units in PVDF-3, 5.4 $\log_{10}$ units in PVDF-4, and 5.7 $\log_{10}$ units in PVDF-5) than mean removal values in FBW (5.8 $\log_{10}$ units in PVDF-1, 5 $\log_{10}$ units in PVDF-2, and 4.7 $\log_{10}$ units in PVDF-6). Ahmadi et al. [22] suggested ΦX174 may have less hydrophobic interactions with the fouling layer than with the membrane, leading to less removal of fouled membranes in contrast to pristine membranes.

In drinking water, removal decreased from the beginning of the experiment (6.6 $\log_{10}$ units ΦX174 in PVDF-3 and 6.7 $\log_{10}$ units MS2) towards the end of the experiment (3.8 $\log_{10}$ units ΦX174 and 4.5 $\log_{10}$ units MS2). This behaviour has been described previously and attributed to an exhaustion of adsorption sites for phages on membranes [21]. In FBW, there is no difference between the phage concentrations in filtrate between the beginning, which could stem from the fouling layer hindering hydrophobic interactions [22].

While PVDF remains a common membrane material, its use is controversial. Due to highly toxic solvents used in production and sulfur dioxide emissions during production, PVDF poses significant environmental risks and adverse health effects [49]. To reduce the negative impacts, less toxic solvents and sustainable polymeric substances (such as cellulose acetate) should be considered as alternatives to fluoride containing membrane materials such as PVDF [49].

3.8. Phage removal with SiC membranes

The pore size of the SiC membrane was significantly larger than the geometric size of both phages, and also much larger than the pores of the other membranes, therefore highest phage breakthroughs were expected for SiC membranes. Counterintuitively, removals did not differ significantly from the other membranes. SiC was the most negatively charged membrane in this study, with a zeta-potential of −58 mV at pH 7 (see Table 2), and was the only hydrophilic membrane [12,50].

For ΦX174 retention, electrostatic repulsion was likely negligible and LRV could have been increased by hydrophilic adsorption of ΦX174 onto the membrane surface, while electrostatic repulsion should have increased MS2 retention [21]. If these effects impacted phage removal to a similar degree, that could explain similar retention of both phages.

Further experiments at different pH values could shed light on the impact of these effects. While size exclusion can be aided by fouling [46,47], there was no significant difference between phage removal in FBW (experiments SiC-1 and SiC-3) and DW (experiments SiC-2 and SiC-4). Ahamdi et al. [22] demonstrated that fouling could negatively impact phage removal by ceramic membranes, as the change of hydrophobicity may lead to less interaction between the membrane surface and phages, especially in the case of ΦX174.

4. Conclusion

In this study, the retention of phages during GW and FBW treatment was examined to address microbial safety concerns.

A GW treatment filter in pilot scale was evaluated for its potential to remove phages. At a filtration velocity of 7.5 m/h, no MS2 removal was achieved, while ΦX174 were removed by about 0.9 $\log_{10}$ units. MS2 concentrations in the FBW were comparable to the spiked raw water concentrations, while ΦX174 concentrations were slightly higher in FBW than in spiked raw water. The FBW was treated with a flat-sheet PES membrane in cross-flow, and all permeate samples were free of MS2 phages, resulting in a removal of 3 to 4 $\log_{10}$ units. ΦX174 phages were removed by up to 3.2 $\log_{10}$ units and no removal difference was detected between Fe-FBW and Mn-FBW. Removal efficiency decreased in repeat runs, indicating a loss in adsorption capacity on the membrane surface over time.

In lab-scale experiments, phage removal from two matrices (DW and FBW) using two hollow-fibre membranes and one plate system (PES, PVDF, and SiC), which represented different materials, pore sizes, and surface characteristics, were compared. Phage retention of at least 3.8 $\log_{10}$ units was achieved with all membranes, and retention could not be explained by membrane pore size alone. For membranes with pore sizes larger than phage diameter (PVDF and SiC), high phage retention of almost 7 $\log_{10}$ units could be achieved. Further studies are necessary to determine the exact removal mechanism. Retention in FBW did not significantly differ from retention in DW, showing no clear positive impact of the fouling layer.

Phage retention from feed samples was lower in FBW than in DW, implying FBW components interaction with phage particles. Overall, UF membranes were an effective barrier for phages in FBW reclamation, reaching LRV of over 4 $\log_{10}$ units with different membrane types.

CRediT authorship contribution statement

A. Steuer: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **C. Kast:** Writing – review & editing, Validation, Methodology, Investigation, Conceptualization. **D. Mahringer:** Writing – review & editing, Supervision, Resources, Funding acquisition, Conceptualization. **C. Förster:** Writing – review & editing, Validation, Resources, Investigation. **M. Ernst:** Writing – review & editing, Resources, Project administration, Funding acquisition. **A.S. Ruhl:** Writing – review & editing, Validation, Supervision, Resources, Funding acquisition.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.seppur.2025.136283.

Data availability

Data will be made available on request.

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