

Article

## Ultrafiltration (UF) Pilot Plant for Municipal Wastewater Reuse in Agriculture: Impact of the Operation Mode on Process Performance

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**Abstract:** Following increasing interest in the use of UltraFiltration (UF) membrane processes as an alternative advanced disinfection technique, the performance of a UF pilot plant was investigated under two opposite operating conditions (“stressed operating condition” *versus* “conventional operating condition”). The results indicate that for both conditions, the reclaimed effluent complied with the Italian regulations for unrestricted wastewater reuse (*i.e.*, Total Suspended Solids (TSS) < 10 mg/L; Chemical Oxygen Demand (COD) < 100 mg/L and *Escherichia coli* < 10 CFU/100 mL). On the other hand, when compared with the Title 22 of the California Wastewater Reclamation Criteria, only the effluent produced under the “conventional operating condition” met the stipulated water quality standards (*i.e.*, TSS and turbidity undetectable and total coliforms < 2.2 CFU/100 mL). It should be noted that, in spite of the nominal cut-off size, total coliforms breakthrough was indeed occasionally observed. A localized membrane pore micro-enlargement mechanism was hypothesized to explain the total coliforms propagation in the ultrafiltered effluent, as monitoring of the membrane permeability and transmembrane pressure highlighted that gel/cake formation had only a minor contribution to the overall membrane fouling mechanism with respect to pore plugging and pore narrowing mechanisms.

**Keywords:** ultrafiltration; wastewater reclamation; membrane fouling; process monitoring

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## 1. Introduction

Interest in the use of UltraFiltration (UF) membrane processes as an alternative technology to conventional disinfection techniques, designed to achieve the restrictive agriculture wastewater reuse standards, is growing [1-3]. To this aim, the application of UF membrane processes has proved to be competitively priced and the possibility of easily installing the UF equipment into the current wastewater treatment facilities is a further key element [4,5].

From a pathogen removal point of view, previous researches underlined that microbial indicators such as *E. coli*, fecal coliforms or total coliforms were effectively blocked by the UF membrane surface [6,7]. However, more contradictory results were obtained when smaller pathogens like *Cryptosporidium*, *Giardia* and MS2 were considered [5,8,9].

Moreover, the overall UF process performance is heavily affected by the progressive fouling of the membrane surface. The literature reports several different direct methods (*i.e.*, inside gas sparging, outside aeration, *etc.*) and indirect ones (*i.e.*, changing operating modes, membrane surface modification, *etc.*) potentially able to minimize fouling deposition on UF membranes [10-13]. However, the impact of such membrane fouling on the microbial and physical-chemical characteristics of the effluents produced by UF systems is not fully cleared to date.

On the basis of the above results, the objective of this study was to investigate the application of the UF membrane process for the treatment of a secondary settled effluent in order to achieve the Italian unrestricted agriculture wastewater reuse standards. In particular, a UF pilot plant was realized and fed with the wastewater coming from the secondary sedimentation unit of the City of Taranto (South Italy) Municipal Wastewater Treatment Plant (TMWTP). The equivalence of the effluent produced by such an UF pilot plant with the strict standards required by the Title 22 Water Recycling Criteria ruled by the State of California was also investigated.

In particular, the above objectives were investigated under two opposite operating conditions, *i.e.*, “stressed operating condition” *versus* “conventional operating condition”, in order to characterize the microbial and physical-chemical characteristics of the final effluent produced in both situations. In addition, the membrane fouling mechanism occurring under each working condition was explored.

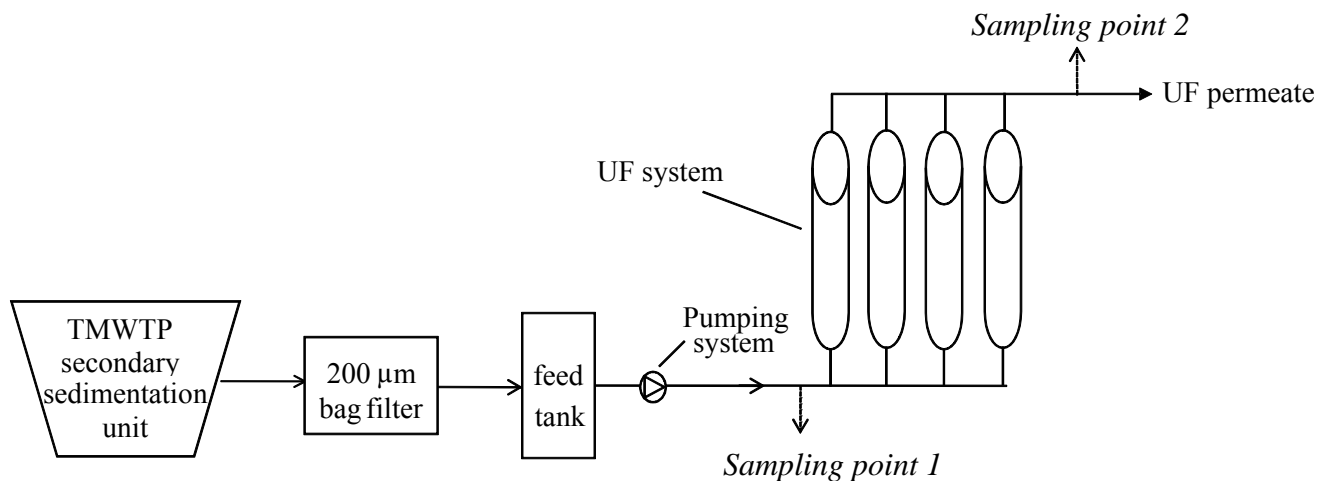
## 2. Materials and Methods

### 2.1. Experimental Facilities

The pilot plant used in this research is located in the TMWTP. The TMWTP consists of a conventional sewage treatment system, *i.e.*, mechanical screening and sedimentation followed by activated sludge oxidation and secondary sedimentation. Then, the secondary settled effluent (SSE) is disinfected with sodium hypochlorite in order to achieve the Italian regulation standards for wastewater discharge.

The pilot plant worked 24 hours/day and was fed with the SSE coming from the TMWTP. A scheme of the UF pilot plant configuration is shown in Figure 1.

**Figure 1.** Flow diagram for the ultrafiltration (UF) treatment system.



The SSE was passed through a 200  $\mu\text{m}$  bag filter and collected in a feed tank upstream of the UF system. The bag filter (Everblue Co., Italy) was used to remove extremely large particles that could damage the membrane fibers.

From the feed tank, a pumping system (Lowara Co., Italy) was used to distribute the SSE into the UF system. The crossflow UF treatment system consisted of four polysulfone hollow fiber membrane cartridges (Dizzer 5000+, Inge Co., Germany). The filtration surface of each hollow fiber membrane cartridge was 50  $\text{m}^2$  with a specific maximum flow rate of 140  $\text{L h}^{-1} \text{m}^{-2}$ . The fibers have an interior diameter of 0.9 mm, a nominal pore size of approximately 0.01  $\mu\text{m}$ , and a molecular weight cutoff of 200,000 Daltons.

The feed water was distributed uniformly among the four cartridges by a common header into the lumen of the fibers. The SSE travelled through the lumen of the fibers and then permeated through the pores of the thin skin to the outside of the fibers. Thus, a further common header collected the whole permeate produced by the four membrane cartridges.

Two sampling points were used during this experiment to collect the wastewater samples. The first sampling point was located immediately before the four membrane cartridges, the second immediately after. Accordingly, the sampling point 1 and point 2 permitted collection of the UF inlet and UF outlet, respectively.

## 2.2. The UF System Key Parameters

The typical process cycle of a conventional UF system consists of a production period, a backwash period and a fast flush period. During the production period, permeate is produced by the UF system. In this experiment, there was no discharge of the wastewater flowing in the interior of the lumens as the whole inlet flow rate was permeated through the membranes. During the backwash period, permeate mixed with sodium hypochlorite is pumped into the permeate ports of the cartridges. The resulting solution flows from the outside to the inside of the membrane fibers. Backwashing allows

solids to be lifted off and flushed out of the membrane fibers, whilst the sodium hypochlorite should prevent the formation of fouling on the membrane pores. At the end of the backwash period, the fast flush period begins: feed water is pumped through the membrane cartridges at a high velocity in order to remove those particles that were not removed during the backwash period.

Transmembrane pressure (TMP) and membrane permeability (MP) are the parameters usually considered to characterize the typical process cycle. TMP is generally determined by subtracting the permeate pressure from the average of the inlet flow rate and discharge pressure. As no discharge was produced in this investigation, TMP was calculated as follows:

$$TMP = P_{feed} - P_{perm} \quad (1)$$

where  $P_{feed}$  is the feed pressure and  $P_{perm}$  is the permeate pressure.

MP is defined as the volume that passes through a given membrane area under the TMP established during a certain time interval. As a consequence, MP was calculated according to the following equation:

$$MP = \frac{Q_{perm}}{TMP \cdot A} \quad (2)$$

where  $Q_{perm}$  is the permeate flow rate,  $A$  is the total filtrating surface area and  $TMP$  is the transmembrane pressure.

### 2.3. Experimental Design

The research was conducted in two phases. Table 1 summarizes the characteristics of the UF typical process cycle investigated in each phase.

**Table 1.** Experimental plan and typical process cycle characteristics for the two experimental phases.

Period	Phase	Number of trials	Inlet Flowrate [m <sup>3</sup> /h]	Production Period [s]	Backwash Period [s]	Backwash Flowrate [m <sup>3</sup> /h]	Fast flush period [s]
9/13–10/2	<i>Stressed Operating Conditions (SOC)</i>	3	10	1,200	85	5	5
10/7–10/16	<i>Conventional Operating Conditions (COC)</i>	1	10	1,200	85	25	5

The production period was 20 minutes. Then, a backwash period took the next 85 seconds, while a fast flush period of five seconds concluded the typical process cycle. For the backwash period, the permeate was mixed with sodium hypochlorite (14% NaClO, Merck Co., NJ, USA) in order to achieve a free chlorine concentration of 100 mg/L in the backwashing water. The above conditions were chosen based on the results obtained in previous research carried out using similar feeding wastewater [6,7].

In order to investigate the UF system performance under extremely different operating conditions, two different backwashing conditions were realized in phase 1 and phase 2. In phase 1, the so-called “*stressed operating condition*” (SOC) phase, the effect of a poor backwashing was investigated: the

backwashing flow rate was half of the feeding flow rate. In phase 2, the so-called “conventional operating condition” (COC) phase, the backwashing flow rate was twice the feeding flow rate as per manufacturer suggestions.

For each trial carried out in both phases, the pumping system prevalence was set in order to attain an initial flow rate of 10 m<sup>3</sup>/h during the production period. Accordingly, the TMP ranged from 30–40 kPa in both the SOC and COC phase. Then, the flow rate and TMP were not controlled and they were allowed to fluctuate in response to the quality of the feed and the fouling of the membranes. In order to preserve membrane integrity, the pilot plant operation was stopped when the TMP achieved 120 kPa and the membranes were chemically cleaned.

For chemical cleaning, the following solutions were preliminarily prepared using part of the permeate produced during the production period: (1) chlorine caustic solution: caustic soda (30% NaOH, Solvay Co., Belgium) and sodium hypochlorite were added to the permeate in order to prepare a solution of pH 10 and with a concentration of 100 mg/L of free-chlorine; (2) hydrochloric acid solution: hydrochloric acid (33% HCl, Solvay Co., Belgium) was added to permeate in order to achieve a solution of pH 2.5; (3) sodium hypochlorite solution: sodium hypochlorite was added to the permeate in order to prepare a solution with concentration of 100 mg/L of free-chlorine. The membranes were first backwashed for 30 minutes with the chlorine caustic solution and afterward soaked in such solution for the next six hours. Then, the above procedure was serially repeated with the hydrochloric solution and sodium hypochlorite solution. The chemical cleaning ended with rinsing of the permeate pipeline of the UF system with the sodium hypochlorite solution for one hour.

The UF typical processing cycles were characterized and the UF system key parameters described in Section 2.2 were determined. For this purpose, the following data were recorded from the UF system control panel three-times a day: inlet pressure, permeate pressure, and permeate flow rate. Then, the TMP and MP were calculated according to equations (1) and (2), respectively. Moreover, the physical-chemical characteristics of the wastewater before and after the UF treatment were also considered. To this aim, 500 mL-UF inlet sample was collected daily, immediately before entering the UF system using the sampling port 1 (see Figure 1) and another 500 mL-UF outlet sample using the sampling port 2 (Figure 1). Before collecting the samples, both sampling ports were left opened for at least four hours. Then, the following analyses were performed on each sample according to Standard Methods [14] immediately after collection: chemical oxygen demand (method 5220D), turbidity (2130B), and suspended solids (2540D). Furthermore, pH and temperature of the UF-inlet sample were also measured using a WTW-multiline-F instrument (WTW Co., Weilheim, Germany). The spectrophotometer used for the above analyses was a HACH DR 2010 (Calverton, VA, USA).

In regard to the microbial determinations, two further 500 mL samples were contemporaneously taken from sampling port 1 and sampling port 2. The collection frequency was twice a week for each sample and the days of collection were chosen in order to be representative of the progressive fouling of the UF system (this point is discussed further in Section 3). Each sample was taken in a Sterilin bottle and stored at 4 °C. Total coliform and *E. coli* indicator numbers were enumerated within four hours from collection and realized according to membrane-filtration-based procedure no. 9222D from Standard Methods [14]. Such indicators were detected because they are standard indicator organisms regulated in Italy and elsewhere for agriculture wastewater reuse.

### 3. Results and Discussion

#### 3.1. Feed Water Characteristics

Table 2 compares the analyzed parameters of the SSE along with the mandatory standards established in Italy and in the State of California (U.S.) for unrestricted agriculture wastewater reuse. The State of California's agriculture wastewater reuse regulations were considered because most developed countries defined their wastewater reuse standards on this basis.

**Table 2.** Characteristics of the secondary settled effluent (SSE) coming from the City of Taranto (South Italy) Municipal Wastewater Treatment Plant (TMWTP).

Parameter	unit of measurement	min	max	average	MAC <sup>a</sup>	
					Italy <sup>b</sup>	State of California <sup>c</sup>
pH		6.3	7.5	7.2	6–9.5	N.S. <sup>d</sup>
Temperature	°C	19	25	23	N.S.	N.S.
Conductivity	µS/cm	1,584	1,950	1,800	N.S.	N.S.
Turbidity	NTU <sup>e</sup>	1	7	5	N.S.	2
Total Suspended Solids (TSS)	mg/L	1	8	3	10	N.S.
Chemical Oxygen Demand (COD)	mg/L	26	69	49	100	N.S.
<i>E. coli</i>	CFU <sup>e</sup> /100 mL	3,000	36,000	17,000	10	N.S.
Total coliforms	CFU <sup>e</sup> /100 mL	9,100	96,000	33,334	N.S.	2.2

<sup>a</sup> Maximum Allowable Concentrations (MAC) for unrestricted agriculture wastewater reuse

<sup>b</sup> in decree number 185/2,003

<sup>c</sup> in Water recycling criteria (Title 22 Regulations)

<sup>d</sup> N.S. = Not specified

<sup>e</sup> Abbreviations: Nephelometric Turbidity Unit (NTU), Colony Forming Unit (CFU)

The SSE characteristics of the TMWTP were quite constant during the experimentation. The concentrations of *E. coli* and total coliforms were always higher than the respective regulatory targets. As a consequence, a treatment specifically aimed at assuring compliance with the microbial standard regulations was required.

With regard to the other physical-chemical parameters, the SSE was already in compliance with the Italian regulation, whilst the wastewater turbidity was higher than the State of California's threshold. This is in agreement with the requirement of an oxidized, coagulated, clarified and filtered wastewater to be in compliance with Title 22 of the California Wastewater Reclamation Criteria.

The UF membrane cut-off is potentially able to contemporaneously remove suspended particles and coli-organisms from the SSE. Achievement of the above regulatory targets was firstly evaluated using SOC and then in COC in order to deeply investigate the UF performance.

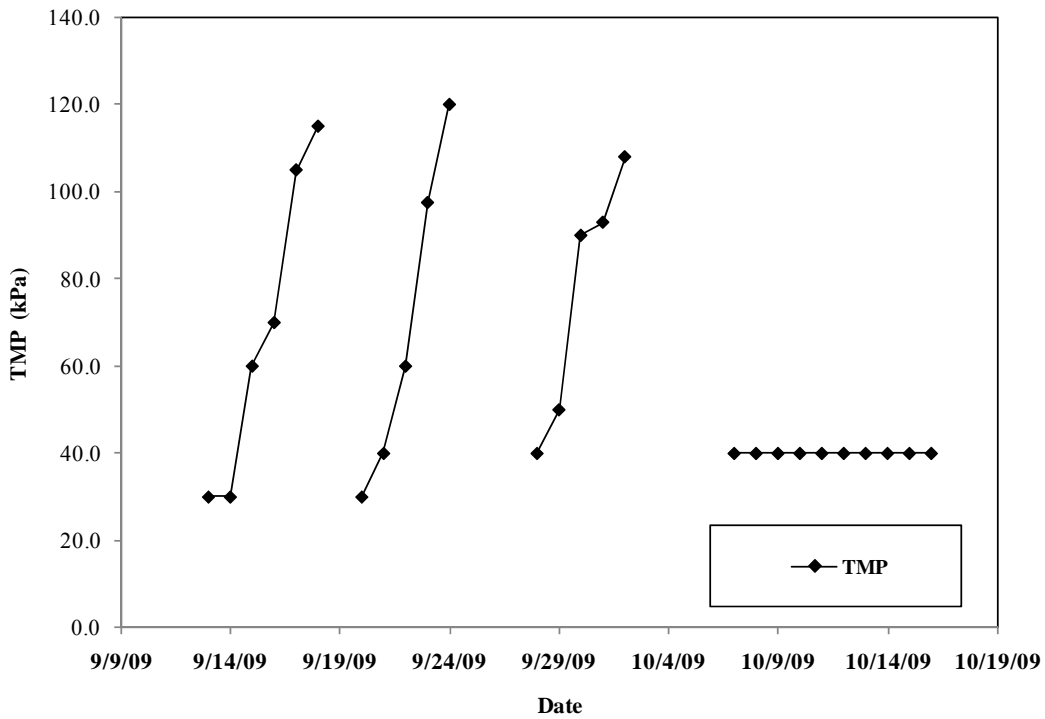
#### 3.2. Stressed Operating Conditions versus Conventional Ones: Effect on the UF System Key Parameters

In the first part of the experiment (from 13 September to 2 October), a poor backwashing flow rate was applied during the backwash period (SOC phase). On the contrary, a higher flow rate was used to

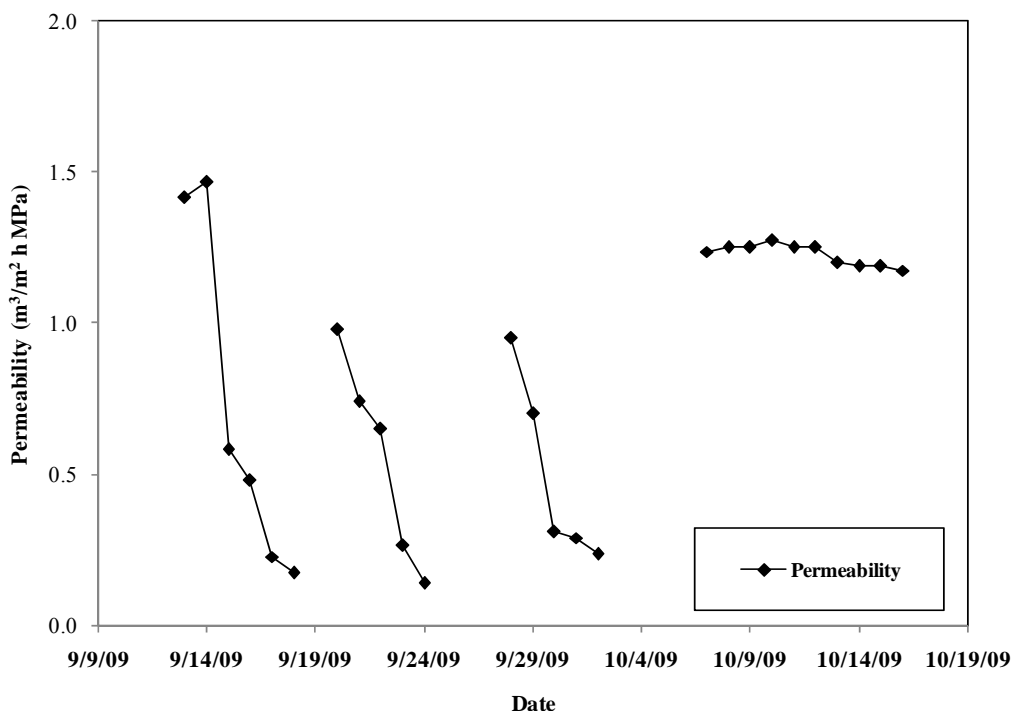
backwash the membranes in the second part of the experiment (from 7 October to 16 October; COC phase).

The UF process cycle performance in both experimental phases was assessed by monitoring TMP and MP. The variation of TMP and MP over time is shown in Figure 2 and 3, respectively.

**Figure 2.** Variation of the daily average TMP data over time calculated according to equation (2).



**Figure 3.** Variation of the daily average MP data over time calculated according to equation (3).



During the SOC phase, the TMP rapidly increased and the maximum TMP value (120 kPa) was achieved within five days of operation. Therefore, the pilot plant was stopped and the chemical cleaning procedure was applied. After each chemical cleaning, the TMP recovered to the initial value showing that the chemical cleaning successfully restored the initial cleaned membrane surface condition. In the COC phase, the TMP remained constant during the whole production time. With such conditions, the overall UF membrane operation period was nine days. Then the pilot plant was stopped even if the TMP data indicated that the UF pilot plant could continue to work indefinitely.

Since the SSE characteristics were constant during the experiment (see Table 2), such a difference in the TMP variation over time between the two phases can be explained by the different backwash efficacy achieved in the SOC and COC phase, respectively.

The increase in TMP over time in the SOC phase could be associated with the limited solids removal obtained during the backwash period. Coherently, once the backwashing flow rate was increased in the COC phase, the backwash period was more effective in removing the particulate matter deposited on the membrane surface (and consequently the TMP was constant). The particulate matter term generally refers to the aggregation of suspended solids and natural organic matter initially present in the SSE that remains blocked on the membrane surface. Since bacterial attachment to surfaces has been demonstrated in a wide variety of environments [15], such particulate matter could include also the presence of attached organisms hidden in particles.

The ineffectiveness of the backwash period in SOC to remove all particulate matter deposited on the membrane surface accelerated the progressive fouling phenomenon over time. This is confirmed by the variation in MP over time for the SOC phase as shown in Figure 3. As more and more particulate matter accumulated on the membrane surface during the production period, the MP of the UF membrane also reduced. Because of the progressive particulate matter deposition, the pore size became more and more plugged during the SOC phase. On the contrary, the MP slightly decreased during the COC phase. Therefore, a minor plugging of the membrane pores can be hypothesized.

Cheryan [16] reported that membrane fouling is caused by deposition and accumulation of particulate matter on the membrane surface and/or within the membrane pores. Ahn *et al.* [17] described the three deposition and accumulation mechanisms on the basis of fouling formation: gel/cake formation, pore plugging and pore narrowing. Gel/cake formation occurs when a large amount of matter accumulates on the surface due to size exclusion from the pores. Pore plugging occurs when particulate matter becomes stuck within the pores of the membrane. Pore narrowing consists of solid material attached to the interior surface of the pores.

Although the impossibility of performing particle size distribution of membrane foulants prevented a univocal definition of the fouling mechanism in the investigated conditions, the TMP and MP behaviors shown in Figures 2 and 3 are quite similar to those that Bourgeois *et al.* [6] found during the ultrafiltration of a filtered primary effluent with a well known particle size distribution. In particular, the rapid TMP increase and the contemporary MP decrease are mainly due to the attachment of particulate matter inside the pores that caused a large resistance to flow. Therefore, it is believed that the SSE also contained large amounts of molecules with sizes similar to, or smaller than the membrane pores (approximately 0.01  $\mu\text{m}$ ).

Accordingly, the pore plugging and pore narrowing seem to play the key role in the deposition and accumulation mechanism taking place in our investigation. On the contrary, the gel/cake formation has



only a minor contribution to the overall membrane fouling mechanism. Therefore, the effect of the poor backwashing flow rate applied during the SOC phase was to induce and promote a fouling mechanism on membrane surfaces mainly characterized by pore plugging and/or pore narrowing.

In the COC phase, TMP and MP remained constant, as shown in Figures 2 and 3, respectively. Due to the effectiveness of the backwash period, no cumulative fouling (resulting in increasing TMP and decreasing MP) occurred over time. Furthermore, there was no indication that this performance would not have continued indefinitely. As a consequence, only a small amount of gel/cake formation was hypothesized to have occurred under the investigated conditions and the backwash period was extremely effective at removing all the matter accumulated on the membrane surface, consequently preventing both pore plugging and pore narrowing.

The above results also underlined that, regardless of the UF operating conditions, the free chlorine concentration (100 mg/L) used to prevent the gel/cake biofouling might be reduced. This is relevant because reducing sodium hypochlorite dosage makes the UF membrane application more economically attractive.

### 3.3. Stressed Operating Conditions versus Conventional Ones: Effect on Wastewater Physicochemical Parameters

Besides the above UF system key parameter analysis, the UF performance in both the SOC and COC phase was also assessed by taking into consideration the impact of the membrane treatment on the physicochemical characteristics of the UF inlet and UF outlet samples.

Table 3 summarizes the turbidity, COD and TSS average values measured on the UF permeate samples collected during the SOC and COC phases.

**Table 3.** UF permeate wastewater characteristics during the SOC and COC phases.

Parameter	Unit of measurement	Min	Max	Average
Turbidity	NTU <sup>a</sup>		<0.2	
Total Suspended Solids (TSS)	mg/L		<0.2	
Chemical Oxygen Demand (COD)	mg/L	20	60	36

<sup>a</sup> Abbreviations: Nephelometric Turbidity Unit (NTU)

Turbidity and TSS determinations were always below the detection limits, in agreement with previous research results [6,18]. The complete removal of TSS occurred since the pore size of the UF membrane (*i.e.*, 0.01  $\mu\text{m}$ ) was approximately one-and-a-half orders of magnitude smaller than the filter used to perform the TSS test (*i.e.*, 0.45  $\mu\text{m}$ ). As particles generally increase the turbidity of water [19], turbidity values below the detection limit suggests that particles smaller than 0.45  $\mu\text{m}$  were also removed.

Therefore, the size-cut operated by the UF membranes effectively removed the TSS initially present in the feed water and such size-cut was independent of the specific phase investigated (*i.e.*, the SOC or COC phase). Consequently, the TSS removal effectiveness was not affected by the specific fouling condition established on the membrane surface during the operation.

With regard to the COD parameter, the average percentage of removal achieved was 30% and this result was analogous in both the SOC and COC phase. Indeed, the organic compounds (*i.e.*, simple organic molecules) with molecular weights smaller than the membrane pore size easily permeate the membrane. As highlighted in similar experiences, such organic compounds are normally found in secondary settled effluent [6,18]. Therefore, the COD removed is the fraction associated with TSS that remains blocked on the membrane surface.

Different from the above parameters, the schedule of the microbial samplings was developed in order to take into consideration the progressive membrane fouling described above. Accordingly, in agreement with the TMP and MP variation with time, shown in Figures 2 and 3, respectively, the sampling days were as follows:

- for each one of the three trials carried out in the SOC phase, the day after the first pilot plant working-day and the day immediately before the pilot plant shut down
- for the unique trial in the COC phase, the day after the first pilot plant working-day and then every four days

Thus, *E. coli* and total coliforms analyses were performed on the two samples taken for the UF inlet and UF outlet. Table 4 summarizes the *E. coli* and total coliforms microbial densities measured during the SOC and COC phase.

**Table 4.** *E. coli* and total coliforms microbial densities measured for the UF inlet and UF outlet samples during the SOC and COC phase.

Parameter		Phase (sampling date)								
		SOC						COC		
		(9/14/09)	(9/17/09)	(9/21/09)	(9/23/09)	(9/29/09)	(10/1/09)	(10/8/09)	(12/8/09)	(10/16/09)
UF inlet	<i>E. coli</i> CFU/100 mL	8,600	15,400	36,000	25,000	10,000	26,000	3,000	17,000	12,000
	Total coliforms CFU/100 mL	9,100	40,500	96,000	40,000	15,000	48,000	18,000	9,500	24,000
UF outlet	<i>E. coli</i> CFU/100 mL	0	0	0	0	0	0	0	0	0
	Total coliforms CFU/100 mL	0	7	0	8	0	0	0	0	0

When the pilot plant worked in the SOC phase, *E. coli* were always completely removed from the feed water. On the contrary, total coliforms were completely removed from all samples taken the day after the first working pilot day, while positive results were found in two of the three samples taken the day before the pilot plant shutdown.

When the pilot plan worked in the COC phase, no positive microbial density was measured for both microbial indicators and for all samples taken.

As the membrane pore size (0.01  $\mu\text{m}$ ) in a 200,000 molecular weight cutoff membrane is much smaller than the coliform bacteria, the membrane should block the coli-species. Therefore, a positive result was not expected in any experimental condition and for both investigated microbial indicators.

One can theorize that a well maintained UF system without membrane defects or equipment leakage should prevent passage of bacteria. However, the experimental conditions seem to exclude membrane damage or equipment leakages. Indeed, in the presence of any of such issues, the positive results for the microbial densities should be recurrent for every sample taken, independent of the specific

experimental phase. Similarly, an accidental sample contamination is unlikely as the positive microbial density results are not randomly distributed with respect to the sampling day or the microbial indicator considered.

Indeed, the total coliforms positive results were measured in the SOC phase and, specifically, the day immediately before the pilot plant shut down. As shown in Figure 2 and 3, UF membranes were working in extremely severe process conditions characterized by TMP close to its threshold and low MP due to the heavy pore occlusion. On the contrary, no positive coliform count was measured during COC, generally characterized by lower and constant TMP and MP.

An explanation of such sporadic total coliform counts could be due to isolated pore micro enlargements under the more severe UF membrane working conditions. Indeed, the majority of the membrane pores are blocked and the measured TMP is close to its manufacturing threshold. Under such stressed conditions, it was hypothesized that the TMP on the membrane surface can locally achieve values even higher than that measured. Accordingly, the permeation of the smaller coli-species could be locally possible as a consequence of membrane pore enlargements resulting from such high pressure. The consistent measurements of TSS below the detection limits (*i.e.*, 0.45  $\mu\text{m}$ ) highlights the little importance (*i.e.*, micro) of pore enlargements that preclude particles passing the UF membrane while only the smallest bacteria are able to permeate. Other authors also reported the presence of species related to the group of coliform microorganisms (*i.e.* *Aeromonas hydrophila* and *Pseudomonas*) in permeate samples of well-maintained UF systems [6,7]. Further research is in progress in order to definitively clear the cause(s) of microbial contaminations.

### 3.4. Stressed Operating Conditions versus Conventional Ones: Effect on Agriculture Wastewater Reuse Regulations

As reported in Table 2, the SSE was not in compliance with *E. coli* MAC established by the Italian agriculture wastewater reuse regulation while total coliforms and turbidity were higher than the standards set by the State of California for unrestricted agriculture wastewater reuse.

Regardless of working conditions (*i.e.*, SOC or COC), the initial *E. coli* population was completely removed (see Table 4). Therefore, UF membranes proved to be a reliable technology to achieve the Italian agriculture wastewater reuse standards.

If the UF system worked in COC, total coliforms were always below the related standard established by the State of California. As the UF membranes represented an effective barrier to the overall particulate matter (see Table 3), turbidity was also lowered under the target. Finally, the membrane blocked the organic content associated with the particulate matter, consequently lowering the biological oxygen demand (BOD) of the UF permeate. Therefore, the UF system produced an effluent equivalent to those of an oxidized, coagulated, clarified, and filtered wastewater as per Title 22.

When the UF system worked in SOC, the total coliform counts in the permeate could be higher than the related standard established by the State of California, while turbidity always remained under the MAC. Therefore, operators of wastewater facilities should prudentially avoid working the UF system under too severe conditions. In particular, in order to identify and differentiate between SOC and COC, the operators should carefully monitor the TMP value achieved during the production period. Before

achieving the maximum TMP value as per manufacturer instructions, a stronger backwash period or membrane chemical cleaning should be applied so that membrane operators can prevent functioning under “stressed” membranes.

Finally, the above results confirm that the UF membranes cannot be considered a complete barrier to pathogens. As a consequence, the integration of the UF system in an overall disinfection strategy based on a multibarrier disinfection approach and long-term residual disinfection strategy should be taken in consideration.

#### 4. Conclusions

Ultrafiltration membrane processes have been recently employed as an alternative technology to the conventional disinfection techniques in order to achieve the restrictive agriculture wastewater reuse standards.

The importance of considering the ultrafiltration system’s key parameters monitoring, *i.e.*, transmembrane pressure and membrane permeability, was underlined by the necessity to characterize the progressive fouling deposition on the membrane surface. The gel/cake formation had only a minor contribution to the overall membrane fouling mechanism with respect to pore plugging and pore narrowing mechanisms. Such a result highlights that the free-chlorine concentration (100 mg/L) applied in this investigation during the backwash phase to prevent biofouling could be significantly reduced.

In most cases, the ultrafiltration membranes represented a reliable barrier achieving the complete removal of *E. coli* and total coliforms. Such indicator bacteria were considered in this investigation because they are controlled by the Italian agriculture wastewater reuse regulation (*i.e.*, *E. coli* < 10 CFU/100 mL) and the State of California regulation (*i.e.*, total coliforms < 2.2 CFU/100 mL). In particular, the ultrafiltration system generally produced effluents equivalent to those of an oxidized, coagulated, clarified and filtered wastewater (*i.e.*, <0.2 mg/L TSS; <0.2 NTU turbidity and 1 mg/L BOD) as per the State of California Title 22 regulation. However, when the membranes were operated in the so-called “stressed operating conditions”, low fecal coliform concentrations (<10 CFU/100 mL) were sporadically found in the permeate. On the contrary, *E. coli* were always undetected in the experimental conditions investigated, assuring consistent respect of the standard required by the Italian regulation.

A localized membrane pore micro-enlargement mechanism was hypothesized to lead to the above total coliforms drawbacks. Accordingly, operators of wastewater facilities adopting an ultrafiltration membrane process to achieve the restrictive agriculture wastewater reuse standards should prudentially avoid working in “stressed operating conditions”. In particular, the process parameter that most needs careful monitoring in order to prevent operation under “stressed” membranes is transmembrane pressure. Before achieving the maximum transmembrane pressure value as per manufacturer instructions, a stronger backwash period or a chemical cleaning of membranes should be prudentially applied.

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