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Wastewater reuse by means of UF membrane process: a comparison with Italian provisions

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ABSTRACT

Membrane filtration can represent a valid solution to water scarcity. In this paper, a study has been carried out about water reuse aimed to industrial and agricultural purposes, starting from a real wastewater coming from Ponte Rosarolo plant located in the Centre of Italy. Wastewater has been treated by means of ultrafiltration membrane process. Results obtained have shown that permeate flux meets provisions in terms of drain water species concentration stated by Italian regulation (D.Lgs 152/99), but there are still unsolved problems as for water reuse limits (D.M. 15/2003): particularly, a final disinfection stage seems to be necessary to lower the value of coliforms content to 0.

Keywords: Water reuse; Membrane filtration; Flux decay; Coliforms; COD; BOD_5

1. Introduction

Scientific community indicates water scarcity as the most important global problem of the present century.

A lot of countries have difficulties to satisfy the growing demand of drinking water, because of the increasing pollution of ground and superficial waters. In some of them, the problem becomes worsen as drinking water availability is a function of seasonal variations, together with drought and flooding events. Moreover, it must be considered that following the current demographic trajectory, the human population will surpass 10 billion by the year 2030 [1]. This trend, coupled with water consumption estimates, that ranges from 1,382 km^3/year in 1950 to 5,235 km^3/year by 2025 [2], states that by 2025 water availability will be about 872 m^3/capita/year, well underneath the limit of 1,000 m^3/capita/year, which defines the people who live in water scarce regions [3].

Water reuse can give a decisive contribute to solve this problem. Nowadays, reuse of wastewater is a very common practice worldwide. Recent studies have demonstrated that membrane separation technology can be successfully applied to purify wastewaters released from all kinds of factories. For example microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) processes have shown good efficiency to achieve water reuse requirement in textile [4], leather [5], food and beverage [6], electronic [7], diary industries [8], and municipal wastewater [9]. The success of membrane process is due to their low energy consumption [10] absence of phase change,

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lack of chemical addition [11,12], easy to operate plants [13], with respect to traditional process.

The objective of this paper is to verify if a real wastewater, particularly coming out from Ponte Rosarolo municipal wastewater treatment plant located in the Center of Italy (AQ, Italy), being processed with a UF tangential membrane, is able to meet Italian water reuse regulation, that is D.M. 185/2003. Results obtained have been compared with D.Lgs 152/99 specifics, too, which deal with the Italian water protection law against pollution.

Basically, Ponte Rosarolo plant is a traditional municipal wastewater treatment plant, with a preliminary pretreatment (coarse screening and fine screening) followed by a biological treatment, which consists of an activated sludge oxidation stage followed by a sedimentation step. The final disinfection step is carried out by using sodium hypochlorite as a disinfectant agent.

2. Materials and methods

2.1. Apparatus description

Experimental studies have been carried out with an UF tangential flow Membralox XLAB 3 (Exekia, Bazet, France) laboratory pilot plant with a single tube Membralox® (Fig. 1). The membrane is made of zirconium oxide and its molecular weight cut-off is 1,400 kDa (≈50 nm).

The recirculation pump has a nominal flux of 1 m^3/h and assures a fixed tangential velocity of 7 m/s.

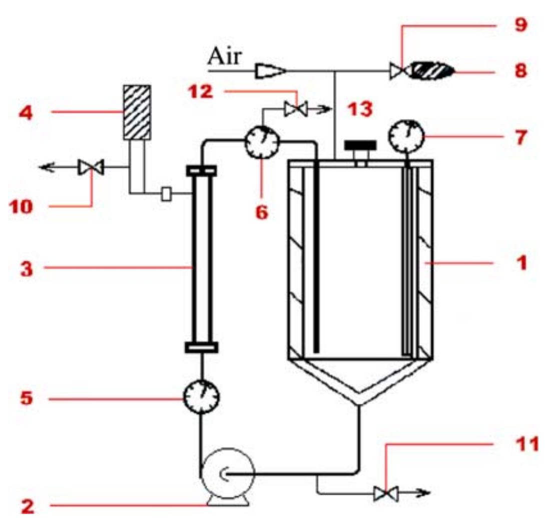

Fig. 1. Experimental apparatus used for UF process. 1: Jacketed feed tank; 2: pump; 3: membrane module; 4: back-flush device; 5, 6: manometers ($0–4 \times 10^5$ Pa); 7: temperature gage; 8: muffler; 9, 10, 11: valves; and 12: air purge valve.

Experimental runs have been carried out at room temperature. Cleaning procedure has been performed at 40°C; during this stage, temperature has been controlled by tank jacket connected to a Crioterm 10–80 thermostat.

The plant is equipped with a back-flush system BF3, controlled by an electrovalve (pressure 7×10^5 Pa, re-injected volume 3 ml). Back-flush device has been activated during membrane cleaning procedure with manual frequency (about 2 min) and controlled length (about 1 s).

2.2. Feed water characteristics

In this paper, water has been sampled at the outlet of the municipal wastewater treatment plant of Ponte Rosarolo in L'Aquila (Italy), after the chlorination section. Each sample has been analyzed to evaluate total suspended solids (TSS) and total dissolved solids (TDS) by evaporation at 105°C according to the Standard Methods [14]; pH has been evaluated using a Backman Φ72 pH meter; electrical conductivity using a microprocessor LF196 conductivity meter; biological and chemical oxygen demand (BOD_5, COD) total phosphorous; Total Kjeldahl Nitrogen (TKN) have been determined using UV–vis spectrophotometer type Cadas 50 and its corresponding measurement kits (Dr. Lange) at various wavelengths. As for the biological indicator, total coliforms have been analyzed using colonies growth on a suitable nourishing ground, according to Standard Methods [12].

2.3. Experimental runs

Three experimental runs have been carried out in the present study: the first concerns flux decay experiment, the second and third regard concentration experiments (run 01 and run 02). In particular, concentration experiments have been divided into two different experimental runs:

- the first, named *run 01*, has been directly processed with the UF membrane;
- the second, named *run 02*, has been prefiltered with a screen of 0.45 μm rating, before processing with the UF membrane.

3. Results and discussion

3.1. Flux decay experiment

Flux decay experiment has been carried out using a wastewater sampled from the municipal wastewater treatment plant of Ponte Rosarolo in L'Aquila (Italy), after the chlorination process and before its entry in Aterno River. Wastewater has been analyzed in terms of TSS, TDS, turbidity, and coliforms, as reported in Table 1.

Later on this sample has been treated with a screen of 0.45 μm of rating, in order to separate coarse solids, avoiding the sudden membrane fouling.

Conditions of flux decay experiment have been described in Table 2.

After 3 h run, permeate flux decays more than 30% with respect to its initial value. This value is about twice as the one detected by Ferella et al. [15], which nevertheless have performed experimental runs with distilled water.

After 50 min run, permeate and retentate flux have been sampled to be analyzed in terms of TSS, TDS, turbidity, and coliforms. Results obtained are showed in Fig. 3, which reports the comparison with the feed values reported in Table 1.

Fig. 2 shows a flux decay that closely follows its typical trend [11].

UF membrane is very selective against coliforms: this behavior is confirmed by a very low coliform concentration in permeate flux, while retentate one is even higher than feed flux. These results demonstrate that micro-organisms do not adhere to membrane surface.

Differently from data obtained by Di Zio et al. [16], they have been transferred to retentate flux, increasing its bacteriological content.

Fig. 4 shows the comparison between the permeability of dirty membrane—after 6 h run—clean membrane and after washing with chemical solution. Dirty membrane has been cleaned using distillate water: this washing method has demonstrated to be very effective, as permeability of clean membrane is nearly superimposed the one obtained after chemical washing. Moreover, a linear trend is observed between permeability against the transmembrane pressure: this behavior has been confirmed during all the runs performed.

Table 1
Wastewater analysis

Parameter	Value
TSS, mg/l	6.0
TDS × 1E−03, mg/l	5.0
Turbidity, NTU	1.5
Coliforms × 1E−04, UFC/100 ml	1.43

Table 2
Conditions of flux decay experiment

Parameter	Value
Membrane rating	50 nm (≈1,400 kDa)
Temperature	Room value (25℃)
Speed filtration	7 m/s
Flux decay experiment length	6 h
Trans Membrane Pressure (TMP)	2.8 bar

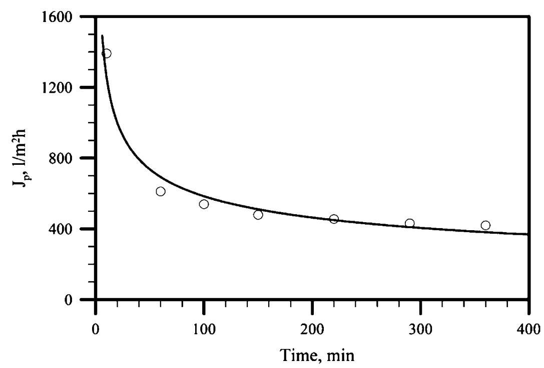

Fig. 2. Flux decay curve.

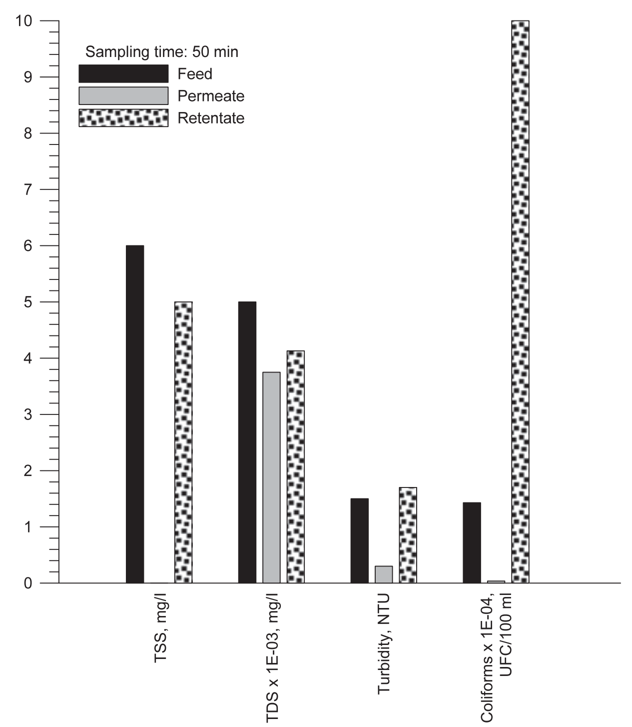

Fig. 3. Permeate and retentate analysis after 50 min run. A comparison with feed values (Table 1).

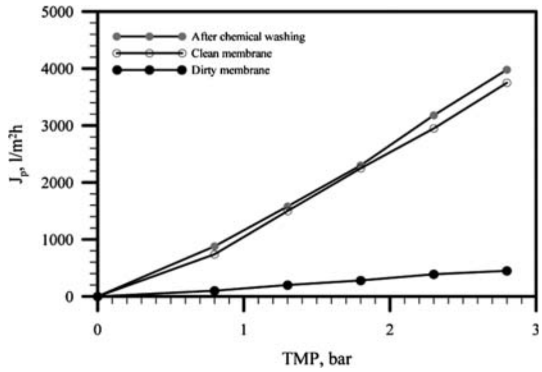

Fig. 4. A comparison between dirty membrane (after 6 h run), clean membrane (using distilled water), and clean membrane (using chemical solution) permeability.

3.2. Concentration experiments

As before specified, two different concentration experiments have been carried out: the former, named run 01, has been performed with wastewater directly processed with UF membrane; the latter, named run 02, has been managed with wastewater, which has been first prefiltered with a screen of 0.45 μm rating, then processed with UF membrane.

Table 3 reports experimental conditions for both runs.

3.3. Results obtained during run 01

Fig. 5 reports the comparison between permeability of dirty membrane—after 6 h run—clean membrane with distilled water and clean membrane after chemical washing. The permeability of dirty membrane is poor, because wastewater flux "as it is" shows high fouling capabilities due to the presence of several suspended solids, which have not been removed before.

Once again, as before detected in Fig. 4 during flux decay experiment, cleaning procedure with distilled water is very effective, because, after this treatment, membrane has shown permeability values very close to the ones that is possible to obtain by means of cleaning procedure with chemical solutions.

Table 3
Experimental conditions for both run 01 and run 02

Parameter	Value
Membrane rating	50 nm (≈1,400 kDa)
Temperature	Room value (25°C)
Speed filtration	7 m/s
Concentration experiment length	6 h
TMP during permeability test	0.8, 1.3, 1.8, 2.3, 2.8 bar
TMP permeate sample	2.8 bar

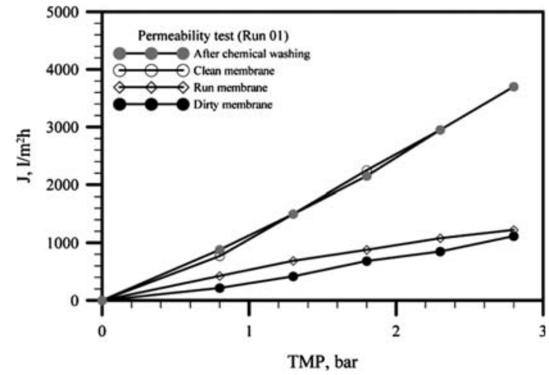

Fig. 5. Membrane permeability during run 01.

Fig. 6 shows a comparison between characterization of UF membrane process permeate and retentate flow against the feed flow, that is the run 01 wastewater sample. Low retention of conductivity (about 4%) means that UF membrane is not able to separate salts and dissolved ions.

Remarkable iron (about 80%) and lower (15%) manganese retention are both due to adsorption process on membrane surface gel cake: make sure membrane, without concentration polarization, is not able

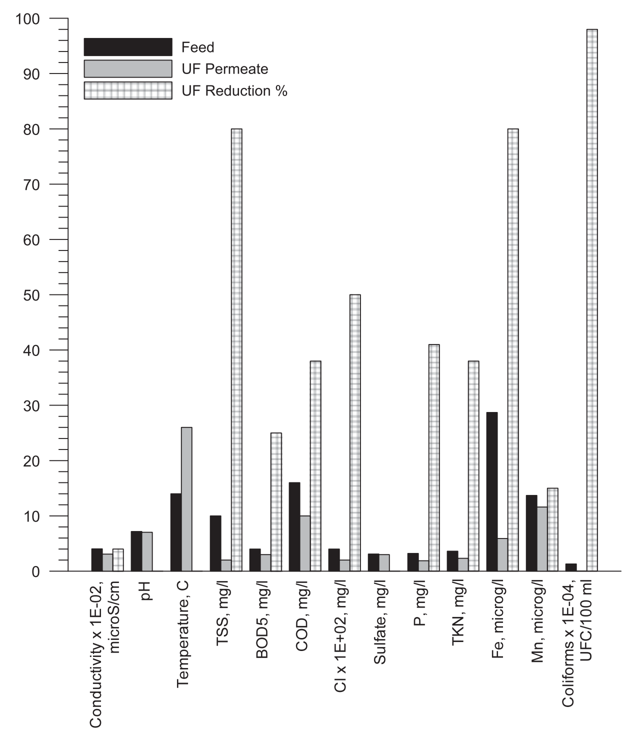

Fig. 6. Analysis of feed and permeate flux in UF process: run 01.

to retain none of them. Anyway concentration values of iron and manganese in feed flow are very low, so a pretreatment aimed to their removal is not required.

Total phosphorus has decreased of about 40% and so TKN: the performance of nitrogen, mainly constituted of organic nature, is due to the high abatement micro-organism value, which is possible to perform in permeate flux. As shown in Fig. 6, UF membrane is able to retain the half of free chloride, as a residue specie of chlorination step in Ponte Rosarolo Plant. COD and BOD_5 have been decreased about 25 and 38%, respectively: this behavior confirms micro-organism concentration reduction, due to the disposal of oxidable substances in gel cake polarization layer.

The most satisfying result has been obtained in TSS abatement, which has performed about 80% reduction. This trend demonstrates that UF membrane is very suitable as pretreatment to a more advance filtration process like RO, to obtain a wastewater which can be employed in industrial water reuse.

UF membrane process has the great advantage to separate all the species, which can cause premature RO membrane fouling.

Total coliforms undergo a reduction of 98%. It is a very satisfying result, even if Authors expect the 100% value: coliform dimension (1–3 μm) is higher than the membrane pores (50 nm) used in this study. However, it must consider the following reflections:

- Membranes in general show a high pore dimension distribution, so a little part of them can have a higher dimension than rating nominal value (membrane surface imperfections [9]).
- Membrane can be not totally intact: just one fiber corrupted may permit micro-organisms to flow in permeate flux.
- Water sampling should be done under complete sterile conditions, a state that is difficult to carry out.
- Back-flush step can be a further source of contamination: if performed with no chlorine solutions, permeate piping would not be completely sterilized. This condition permits micro-organisms to proliferate during each back-flush cleaning procedure, due to presence of nutrients inside the flow which passes through membrane.

In Table 4, a comparison between permeate flux analysis obtained in run 01 conditions and Italian existing provisions have been reported (D.Lgs 152/99, D.M. 185/2003). All the parameters investigated in the wastewater at the outlet of Ponte Rosarolo Plant are below the values stated in D.Lgs 152/99, which regulates drain water species concentration limits. This behavior confirms the good performance of Ponte Rosarolo Plant, considering furthermore that no nitrogen and phosphorus removal stages are provided.

However, total coliforms does not satisfy the value stated by D.M. 185/2003, that is the limit of 100 UFC/100 ml of *E. Coli* concentration, provided for wastewater assigned to the reuse in agricultural and industrial reuse. The value detected is about two orders higher than the limit value stated by D.M. 185/2003. This result confirm the assessment of Wintgens et al. [9]: UF membrane process cannot be considered a complete barrier to bacteria. So for wastewater reuse an advanced filtration process like RO is necessary.

Table 4
A comparison between permeate flux analysis in run 01 and values stated by provisions

Parameter	Permeate	D.Lgs 152/99	D.M. 185/2003
Conductivity × 1E−02, μS/cm	3.09		30
pH	7.03	5.5–9.5	6–9.5
TSS, mg/l	2	80	10
BOD_5, mg/l	3	40	20
COD, mg/l	10	160	100
Cl × 1E+02, mg/l	2	20	20
Sulfate, mg/l	3	1,000	500
P, mg/l	1.88	10	2
TKN, mg/l	2.32	15[a]	2–15[c]
Fe, μg/l	5.9	2000	2000
Mn, μg/l	11.6	2000	200
Coliforms × 1E−04, UFC/100 ml	0.03	0.5[b]	0.01[b]

[a]Ammonia nitrogen.
[b]This value is solely referred to *E. Coli*.
[c]Ammonia and total nitrogen, respectively.

3.4. Results obtained during run 02

Experimental conditions of run 02 are quite similar to run 01. The only difference is the pretreatment with a screen of 0.45 μm that occurs in run 02 case, to decrease the wastewater capability of fouling UF membrane.

Fig. 7 reports the comparison between dirty membrane permeability during experimental runs—detected after 6 h run—clean membrane with distillate water and clean membrane with chemical solution procedure. Trends reported are very close to the ones discussed in Fig. 5, confirming once again the effectiveness of cleaning membrane procedure with distillate water only.

Fig. 8 shows the comparison between permeability of run 01 and run 02, both during running operations. It can be clearly noticed the benefits are obtained in case of run 02 experimental conditions. Here, the pretreatment with the 0.45 μm rating screen allows to perform a higher permeate flux values than run 01, for each transmembrane pressure. This result permits to increase plant productivity, to extend membrane mean

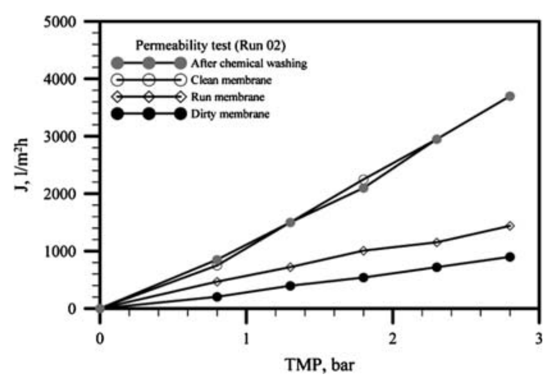

Fig. 7. Permeability trends detected in run 02.

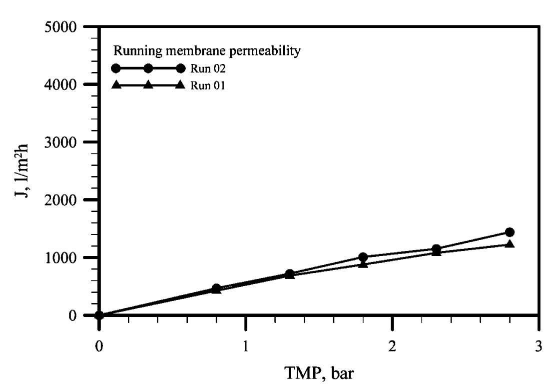

Fig. 8. Comparison between running permeability in run 01 and run 02.

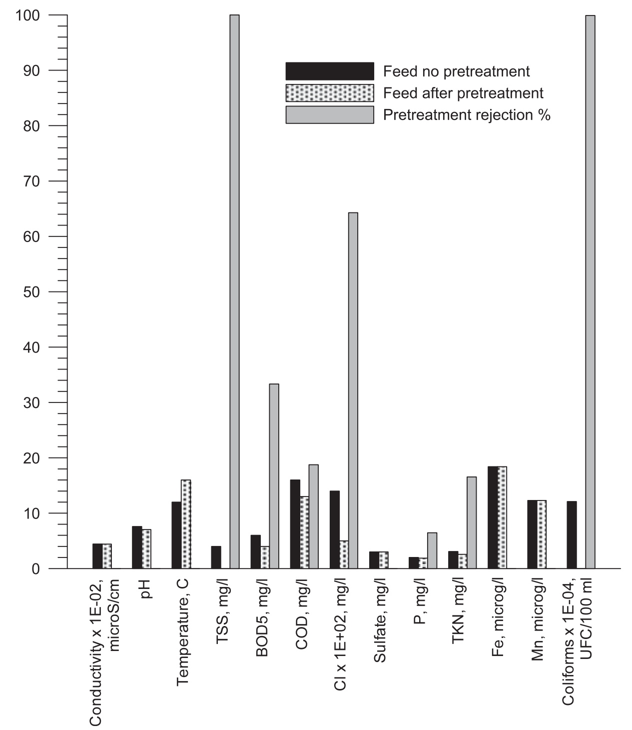

Fig. 9. Analysis of wastewater "as it is" and after pretreatment with the 0.45 μm rating screen.

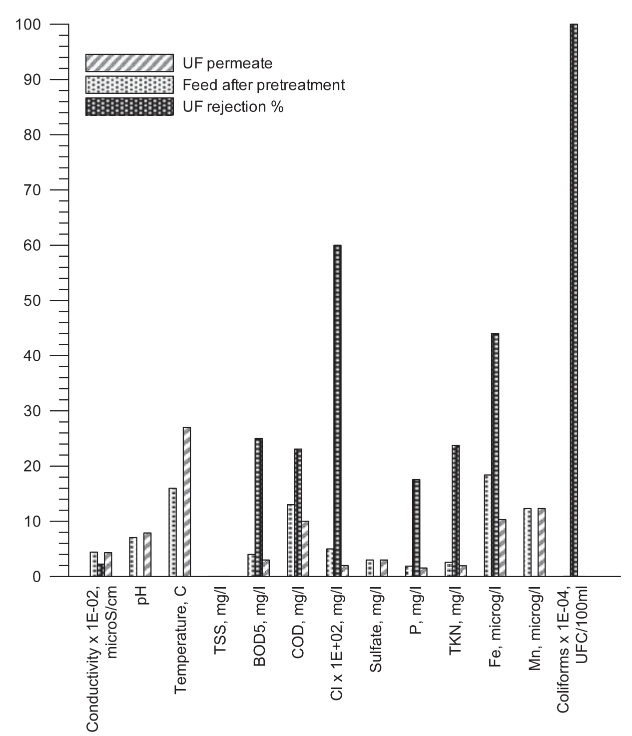

Fig. 10. Analysis of wastewater after pretreatment and of permeate flux after UF process.

Table 5
A comparison between UF permeate flux analysis in run 02 and values stated by provisions

Parameter	Feed	UF permeate	D.Lgs 152/99	D.M. 185/2003
Conductivity × 1E−02, μS/cm	4.44	4.32		30
pH	7.60	7.9	5.5–9.5	6–9.5
TSS, mg/l	4	0	80	10
BOD_5, mg/l	6	3	40	20
COD, mg/l	16	10	160	100
Cl × 1E+02, mg/l	14	2	20	20
Sulfate mg/l	3	3	1,000	500
P, mg/l	2.01	1.55	10	2
TKN, mg/l	3.08	1.96	15[a]	2–15[c]
Fe, μg/l	18.4	10.3	2000	2000
Mn, μg/l	12.3	12.3	2000	200
Coliforms × 1E−04, UFC/100 ml	12	≈0	0.5[b]	0.01[b]

[a]Ammonia nitrogen.
[b]This value is solely referred to *E. Coli*.
[c]Ammonia and total nitrogen, respectively.

life, which gets reduced in cleaning procedure, and finally to lower plant maintenance costs [11].

Fig. 9 reports the analysis of wastewater sampled for run 02 "as it is" and after pretreatment with the 0.45 μm rating screen.

The screen is very selective against coliforms, allowing reaching rejection values near to 100%. Satisfactory results regard TSS and free chlorine abatement, too.

Next UF filtration process (Fig. 10) has permitted to increase COD, BOD_5, and chlorine reduction. Furthermore, experimental data have demonstrated that UF membrane used for this study is very selective against iron and moderately selective against phosphorus and TKN.

Poor salt abatement is confirmed by low conductivity reduction (about 2%): more satisfactory results can be obtained with advanced filtration processes, like NF or, better, RO.

The few total coliforms, residue from the pretreatment with the 0.45 μm rating screen, have been further decreased by UF process.

In summary, it can be said that pretreatment shows a double benefit: on one hand, fouling membrane problems are put off, resulting in a higher permeate flux and lower use of expensive chemical solution for cleaning procedure; on the other hand, plant efficiency improves, due to the integrated process.

Table 5 shows that analysis value of UF permeate flux in run 02 experimental conditions meets the D. Lgs 152/99 Italian provisions for drain water species concentration limits, confirming once again the good performance of Ponte Rosarolo Plant.

However, some problems are still not solved in case of reuse in industrial and agricultural applications of wastewater; here, analysis values showed in Table 5 partially meet Italian provisions (D.M. 185/2003). There are two critical values. The first is the phosphorus concentration, whose value, nevertheless lower, is too close to the limit of 2 mg/l. The second is the *E. Coli* concentration; its value, due to the benefits of pretreatment, is near to 0 but not 0. So for wastewater reusing for agricultural and industrial application, it is necessary to arrange a disinfection stage, by means of chlorine or UV ray treatments, to be reasonably sure of zero bacteria concentration.

4. Conclusions

In this paper, wastewater sampled at Ponte Rosarolo Plant (Centre of Italy, AQ) has been treated with UF membrane process, in order to be reused for industrial and agricultural purposes. After verifying that flux decay curve follows its typical trend, two wastewater samples have been differently processed; in particular, wastewater sample treated with both prefiltered screen and UF membrane process have shown better behavior than the one processed with UF membrane process alone. The improvements detected concern not only on coliforms, whose rejection is near 100%, but also TSS, COD, BOD_5, iron, and chlorine reduction, too. However, results obtained do not permit to reuse permeate flux for industrial and agricultural purposes, as they do not meet Italian provisions (D.M. 183/2003), most of all in terms of coliforms content, whose value must be decreased until 0 limit.

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