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Title	Removal of non-ionic (TAS) and anionic (MBAS) surfactants from real laundry wastewater by means of a full-scale treatment system
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Abstract

Surfactants are considered emerging contaminants (ECs), that can represent a source of problems for environment and human health. This paper aims to quantify the effect of advanced biological (Thermophilic Aerobic Membrane Reactor - TAMR), physical (Nanofiltration - NF) and chemical-physical (Active Carbon - AC) treatments on non-ionic surfactants (TAS) and anionic surfactants (MBAS) removal from a real laundry wastewater (WW). The experimentations included daily monitoring of a full-scale plant for more than three months. The results showed that the TAMR process has been able to withstand high stress conditions (sudden load peaks) and resist to a high concentration of surfactants, allowing it to perform an effective pre-treatment activity. Both in the case of the removal of TAS and of MBAS, the combination of processes made it possible to obtain higher removal yields. Evaluating the operating costs, the results suggested that TAMR+NF has been the optimal combination of processes for the removal of TAS and MBAS. The TAMR+NF+AC sequence allowed almost complete removal of TAS (> 95%) and high removal of MBAS (> 76%) but the costs per unit of mass removed were high.

Keywords	surfactants; thermophilic; biological; nanofiltration; activated carbon; laundry wastewater

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Pavia, August 26th, 2019

Dear Editor-in-Chief,

we would like to submit the manuscript titled "REMOVAL OF NON-IONIC (TAS) AND ANIONIC (MBAS) SURFACTANTS FROM REAL LAUNDRY WASTEWATER BY MEANS OF A FULL-SCALE TREATMENT SYSTEM" for possible publication in *Process Safety and Environmental Protection*.

Surfactants are considered emerging contaminants, that can represent a source of problems for environment and human health. Numerous negative effects that surfactants had on wildlife and, in high concentrations, on humans have been presented. Despite the legislation, a high quantity of surfactants continues to be produced and therefore discharged as wastewater into the environment, for example in laundries wastewater.

This experimentation included daily monitoring of a full-scale industrial wastewater treatment plant for more than three months evaluating the effect of different processes on TAS (non-ionic) and MBAS (anionic) removal, the most common surfactants in the world. A real laundry wastewater has been characterized and three types of treatments have been tested: advanced thermophilic biological (TAMR), physical (NF) and chemical-physical (AC). The yields of six possible combinations have been evaluated (TAMR, NF, AC, TAMR+NF, NF+AC, TAMR+NF+AC).

The results showed that the TAMR process has been able to withstand high stress conditions (sudden load peaks) and resist to a high concentration of surfactants, allowing it to perform an effective pre-treatment activity. Both in the case of the removal of TAS and of MBAS, the combination of processes made it possible to obtain higher removal yields. Evaluating the operating costs, the results suggested that TAMR+NF has been the optimal combination of processes for the removal of TAS and MBAS. The TAMR+NF+AC sequence allowed almost complete removal of TAS (> 95%) and high removal of MBAS (> 76%) but the costs per unit of mass removed were high.

This paper is original and unpublished.

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Title: Removal of non-ionic (TAS) and anionic (MBAS) surfactants from real laundry wastewater by means of a full-scale treatment system

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Abstract:

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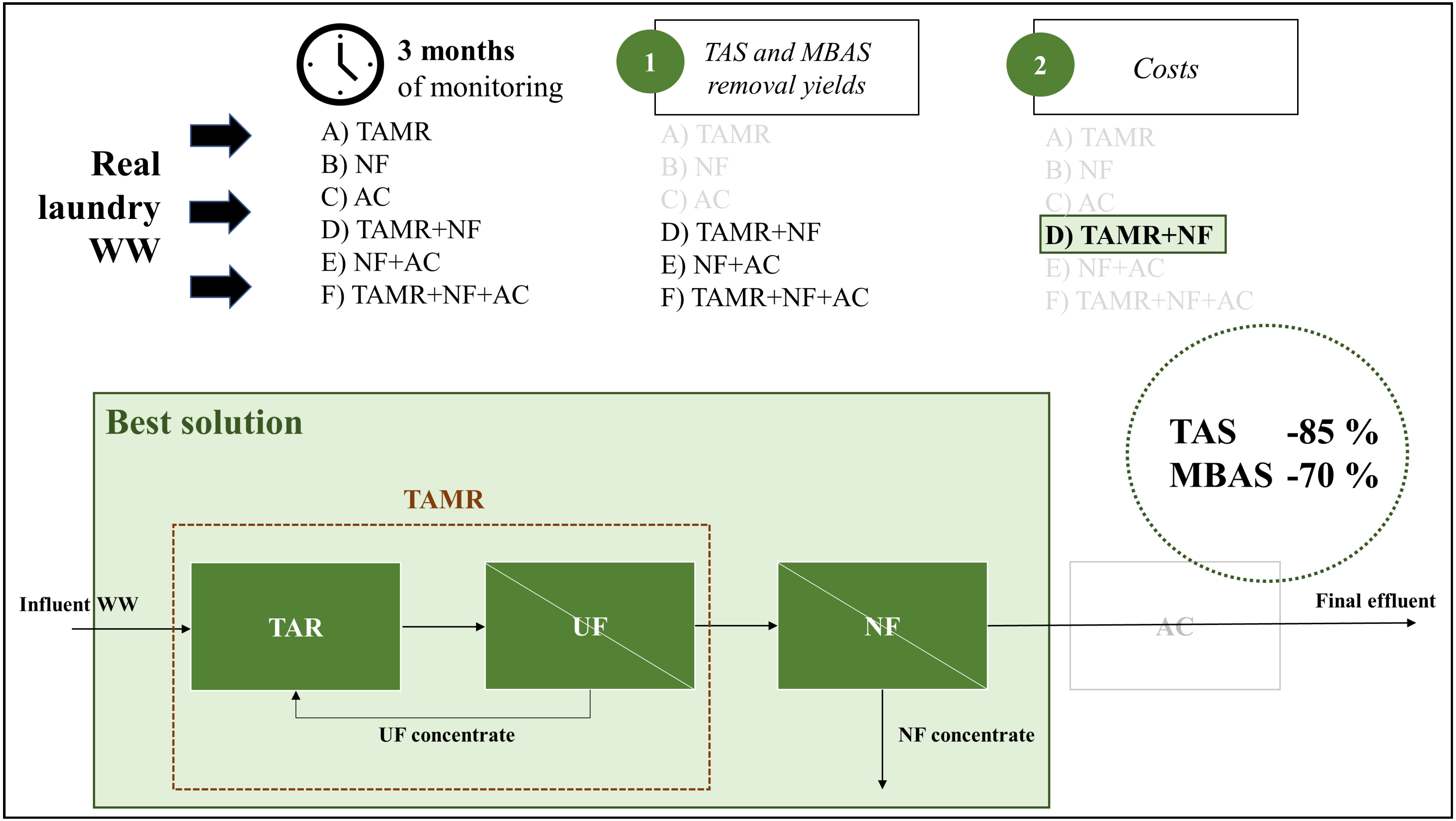
Keywords: *surfactants, thermophilic, biological, nanofiltration, activated carbon, laundry wastewater*

- A real laundry wastewater has been treated for the removal of surfactants
- A full-scale plant has been monitored for more than three months
- The yields of six possible combinations of processes have been evaluated
- TAMR has been able to perform an effective pre-treatment activity
- Costs analysis suggested that TAMR+NF was the optimal solution


Real laundry WW
3 months of monitoring
A) TAMR
B) NF
C) AC
D) TAMR+NF
E) NF+AC
F) TAMR+NF+AC
1
TAS and MBAS removal yields
A) TAMR
B) NF
C) AC
D) TAMR+NF
E) NF+AC
F) TAMR+NF+AC
2
Costs
A) TAMR
B) NF
C) AC
D) TAMR+NF
E) NF+AC
F) TAMR+NF+AC
Best solution
TAMR
Influent WW
TAR
UF
NF
AC
UF concentrate
NF concentrate
Final effluent
TAS -85 %
MBAS -70 %

Removal of non-ionic (TAS) and anionic (MBAS) surfactants from real laundry wastewater by means of a full-scale treatment system

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Abstract

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Keywords *surfactants, thermophilic, biological, nanofiltration, activated carbon, laundry wastewater*

1. Introduction

Surfactants are considered emerging contaminants (ECs) in wastewater (WW) [1]. Generally, WW can contain a wide range of pollutants such as dyes [2], heavy metals [3], pharmaceutical products such as antibiotics [4] and surfactants [5]. The main characteristic of surfactants is that they do not need to be necessarily persistent in the environment to cause negative effects, since they are introduced into the environment continuously [6].

Moreover, surfactants in WW can also represent a source of problems for human health. In fact, numerous negative effects that surfactants had on wildlife and, in high concentrations, on humans have been presented [7,8]. For instance, Linear Alkylbenzene Sulphonate (LAS) effects, as irritation to skin and problem to respiratory system, have been reported [9]. Moreover, in 2017, another surfactant (i.e. Perfluorooctanoic Acid - PFOA), used in the industrial sector as chemicals in industrial processing, has been inserted by the IARC in the list of possible carcinogenic compounds - class 2B [10] and it can also be considered potentially toxic agents for human reproduction [10,11]. Other examples of the negative effects on human health due to the presence of surfactants in water are presented in Table 1.

Table 1: *Several examples of the negative effects of surfactants on human health. (double-column table)*

Surfactants		**Health problems**	**References**
Complete names	**Abbreviations**		
4-nonylphenol	NP	Classified as EDCs	[5,12]
4-tert-octylphenol	OP		
Sodium Dodecyl Benzene Sulfonate	SDBS	Absorbed through the skin, it damages the liver and causes narrowing and other chronic symptoms. It is also teratogenic and carcinogenic	[13]
Linear Alkylbenzene Sulphonate	LAS	Irritation to skin and problem to respiratory system. It can generate carcinogenic and toxic by-products when is degraded	[9,14]
Sodium dodecyl sulfate	SDS	Inhibit ATPase activity of P-glycoprotein, damage membrane structures and initiate oxidative stress response	[15]
Perfluorooctanoic Acid	PFOA	Possible carcinogenic compounds and potentially toxic agents for human reproduction	[10,11]
Perfluoroalkyl Carboxylates	PFCAs	Developmental and hormonal effects, immunotoxicity, and promotion of tumour growth in rodents through their role as PPARα agonists	[16]

Surfactant molecules have a particular structure and contain both hydrophobic and hydrophilic moieties. This structure gives to surfactants properties that can be used in cleaning and textiles industries, consumer goods such as bath soaps and dishwashing detergents, in laundry [17]. There are four different groups of surfactants. The difference is due to the type of charge of the hydrophilic group. Anionic (MBAS) and cationic surfactants contain respectively a negative and positive charged hydrophilic group. Instead, non-ionic surfactants (TAS) have a non-ionised hydrophilic group. Surfactants are classified amphoteric if the charge on the hydrophilic head changes as a function of the pH [18]. The production of the different types of surfactants is not equal. Around 60% of the world's surfactant production is represented by MBAS while TAS accounted for 30% while cationic and amphoteric surfactants represents only the 10% [7].

In recent years the use of surfactants in consumer products such as detergents has been regulated in order to protect the environment and the water quality. Stricter standards, such as *Regulation No. 648/2004* of the EU, have been issued [19]. Despite this, a high quantity of surfactants continues to be produced and therefore discharged as WW into the environment. In fact, still approximately 15 million metric tons of surfactant are produced in the world every year [1]. Therefore, the potential challenge of the removal methods could be the presence of high concentration and various types of surfactants in WW [7]. As reported by Jardak et al. (2016) [5], different types of surfactants present different biodegradability and toxicity behaviour in the environment.

WW produced by industrial laundries present in their composition different levels of suspended solids, turbidity, COD, but the main problem is represented by the surfactants (one of the main constituents that assist in the removal of dirt from the fabric) [20]. MBAS and TAS represent the first and the second (by volume) most relevant groups of surfactants used in cleaning products, respectively [21,22]; in fact, TAS and MBAS together stand for more than 90% of total surfactants produced in the world and discharged in the WW [7].

Among the treatments for laundry WW, physico-chemical and chemical are reported as effective in surfactants removal [23]. However, their major disadvantage is the high operational costs (e.g. oxidants cost), especially when the surfactants concentration is very high. Therefore, biological processes could present a significant alternative due to the lower cost but some disadvantages need to be considered: long reaction time, foam formation and death of the biomass at high concentration of surfactants and high sewage sludge production, that need to be disposed of [5,7,24].

In order to cope this disadvantage, in this experimentation, an advanced thermophilic aerobic biological reactor (Thermophilic Aerobic Membrane Reactor - TAMR) has been tested. Thermophilic treatments show low sludge production (0.05–0.3 $kg_{TSS}\ kg_{CODremoved}^{-1}$) compared to mesophilic conditions helping to reduce the total management costs of a WW treatment plant because sludge management costs represent about 50% [25,26]. Moreover, they present high removal kinetics of biodegradable substrates, about 3–10 times higher than those measured in mesophilic conditions [27]. The presence of ultrafiltration (UF) membrane combined with thermophilic aerobic reactor (TAR) allows to keep all the biomass and reduce the volume of the reactor. As reported by Chang et al. (2001) [28], surfactants (TAS and MBAS) are not influenced by the presence of this membrane because they can pass through the pores of UF, due to their smaller molecular size.

This experimentation included daily monitoring of a full-scale industrial WW treatment plant for more than three months evaluating the effect of different processes on TAS and MBAS removal, the most common surfactants in the world. A real laundry wastewater has been characterized and three types of treatments have been tested: advanced biological thermophilic (TAMR), physical (NF) and chemical-physical (AC). This research aims to test TAMR as a pre-treatment, in order to reduce the peaks of TAS and MBAS concentration exploiting his stability, adopting NF and adsorption on AC as polishing treatment. Therefore, the yields of six possible combinations have been evaluated (TAMR, NF, AC, TAMR+NF, NF+AC, TAMR+NF+AC). Finally, an analysis of the costs of the combined processes in relation to the TAS and MBAS removal yields was discussed.

2. Materials and Methods

2.1. Full-scale wastewater treatment system

The full-scale treatment system was located in province of Bergamo (Italy). The plant allowed to treat 245 m^3 d^{-1} of laundry WW. As reported in Figure 1, it was composed by three different stages of treatments in series: a biological (TAMR), a physical (NF) and a chemical-physical (AC) phase. The samples have been taken before and after each process in order to assess not only the overall performance on the TAS and MBAS but also that of the individual phase of treatment. The full-scale plant has been monitored daily for more than 3 months in order to examine the concentration of TAS and MBAS. More than 1000 samples have been analysed.

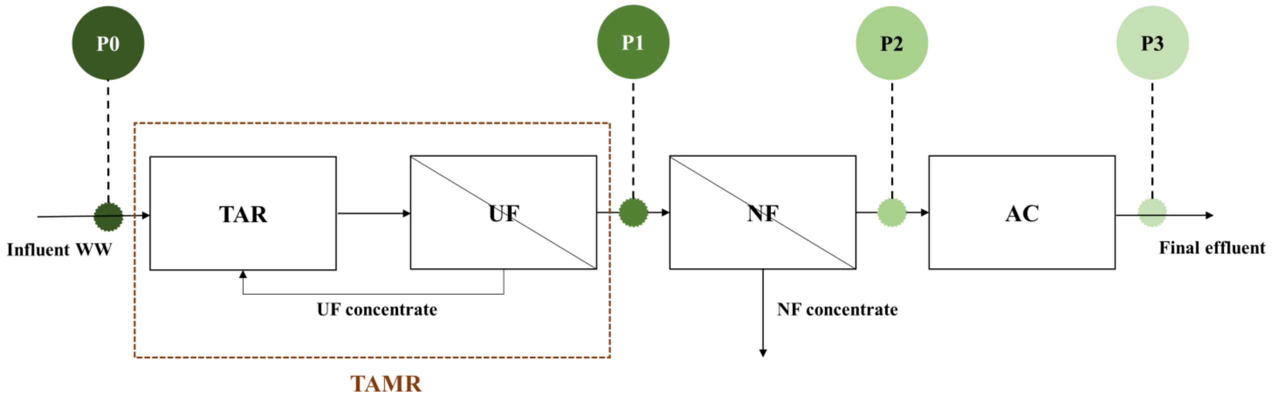

Figure 1: *Scheme of the full-scale WW treatment system and sampling points. TAR: Thermophilic Aerobic Reactor; UF: Ultrafiltration; TAMR: Thermophilic Aerobic Membrane Reactor; NF: Nanofiltration; AC: Active carbon. (double-column figure)*

2.1.1. Characteristics of Thermophilic Aerobic Membrane Reactor (TAMR)

The TAMR was composed by a TAR coupled with UF membranes, as reported in Figure 1. The values of the operating parameters in TAMR system are shown in Table 2.

Table 2: *The values of the operating parameters in the Thermophilic Aerobic Membrane Reactor (TAMR). TSS: Total suspended solids; VSS: Volatile suspended solids; SLR: Sludge loading rate; SRT: Sludge retention rate. (single-column table)*

Parameters	**Values**
TSS [kg_{TSS} m^{-3}]	150-190
SLR [kg_{COD} kg_{TSS}^{-1} d^{-1}]	0.030
VSS TSS^{-1} [-]	0.22
SRT [d]	125
Temperature [°C]	49 ± 1
pH [-]	6.5

O_2 [mg L^{-1}]	1.5

The TAR had a surface of 267 m^2 and a height of 5.3 m for a total useful volume of about 1,000 m^3. The structure was made of reinforced concrete, with walls 30 cm thick and the roof in prefabricated panels that guarantee odour containment and temperature maintenance. For aerobic biological processes, in the tank pure oxygen was injected directly into the static mixers, oversaturating the slurry that was recirculated in them. The fluid thus found itself in the condition of having finely dispersed, in its interior, microbubbles of oxygen in a greater quantity than in a traditional system.

The UF plant was composed of two lines operating in parallel, each consisting of three ceramic membrane channels: each channel contains 99 tubular ceramic membranes with 25 channels. The pore sizes of the installed UF membranes allowed the passage of molecules with a molecular weight lower than 300 kDa and dimensions smaller than 0.3 µm. The operating pressure was 3-5 bar.

2.1.2. Characteristics of the nanofiltration (NF) and activated carbon (AC)

The NF plant was composed of one line, consisting of polyamide thin-film composite: the commercial membranes were FILMTEC® NF270. The cut-off of the installed NF membranes allowed the passage of molecules with a molecular weight lower than 300 Da. The operating pressure was 20-30 bar. The contact angle was 28.77±2.43° [29].

As activated carbon, CARBOFLOC® SP82 has been used. This powdered AC was physically activated with steam in an inert atmosphere. Its physico-chemical characteristics are presented in Table 3.

Table 3: *Physico-chemical characteristics of the activated carbon used in the experimentation. (single-column table)*

Parameters	**Values**
Humidity [%]	1.5
Apparent density [kg m^{-3}]	500
Iodine number [mg g^{-1}]	1000
Ash content [%]	10
pH [-]	9

2.3. Methods

Considering the point of sampling (Figure 1), the removal yields of TAS and MBAS have been calculated using the formula shown in Eq.1:

$$Surfactants\ removal\ [\%] = \frac{C_{in}\left[mg\ L^{-1}\right] - C_{out}\left[mg\ L^{-1}\right]}{C_{in}\left[mg\ L^{-1}\right]} * 100 \qquad (1)$$

Where C_{IN} is the incoming concentration of surfactants incoming and C_{OUT} represents the outgoing concentration of surfactants. Six different configurations of treatments have been considered:

A) TAMR (Considering points P0 and P1)

B) NF (Considering points P1 and P2)

C) AC (Considering points P2 and P3)

D) TAMR + NF (Considering points P0 and P2)

E) NF + AC (Considering points P1 and P3)

F) TAMR + NF + AC (Considering points P0 and P3)

2.3.1. Non-ionic surfactant (TAS)

In order to determine the TAS, the titration with sodium tetrakis-(4-fluorophenyl)-borate dehydrate, has been preceded by sublation in ethyl acetate by blowing nitrogen into the aqueous matrix. The ethyl acetate, containing the surfactants, has been evaporated; subsequently the residue is dissolved in 20-30 ml of distilled water in which the TAS will be quantified by titration. As reported by Tsubouchi et al. (1985) [30], a two-phase titration method was used to determine the concentration of TAS. The experimental procedure was the same reported by Eng et al. (2010) [17] in their experiments.

5 ml of KOH solution 6 M and 5 mL of 1,2-dicholoroethane have been added into the 20-30 mL aqueous solution containing the sublated TAS. After that, 1-2 mL of Victoria Blue B indicator have been added into the mixed solution and shaken vigorously. A 0.5 mM sodium tetrakis-(4-fluorophenyl)-borate dihydrate solution was then added dropwise having simultaneous mixing at every addition. The titration ended when, the colour, in the organic phase, change from pink to purple.

The results are expressed considering the nonylphenol OE (Conversion factor: 366) as reference surfactant (Eq.2).

$$TAS\ concentration = \frac{V_T - V_B}{V_C} * 366 \qquad (2)$$

Where V_T is the titrant volume, V_B is the blank volume and V_C represents the volume subjected to sublation.

2.3.2. Anionic surfactant (MBAS)

Instead of the IRSA-CNR Method 5170, the Lange Kits LCK 332, based on the standard methods ISO 7875-1 (1996) and ISO 7875-2 (1984) [31-32] on the same principle as the previous one, were used to determine the anionic surfactants as MBAS (Methylene Blue Active Substances) that has been quantified as Na-dodecilbenzensulfate. Considering that chloride ions and cationic surfactants can interfere in the measurements: (i) it has been verified that the chlorides and cationic surfactant content in the samples does not interfere with the analysis, (ii) if necessary, the samples are appropriately diluted. The analyses have been always carried out in triplicate so as to record the average value as the final data.

2.3.3. Other parameters

The temperature and pH analyses, in the TAR reactor, have been carried out simultaneously with an pHmeter with glass electrode and thermometer (WTW-IDS, model SenTix® 940). COD, and $N\text{-}NH_4^+$ in the raw influent WW have been quantified according to the standard methods for water and WW [33].

2.3.4. Costs analysis

To assess the economic sustainability of the aforementioned processes, a costs analysis has been carried out. Firstly, the operating costs per unit of treated volume (OC_{TAMR}, OC_{NF}, OC_{AC}) have been taken into account. Only the principle costs items have been evaluated and used in the analysis. For the TAMR, energy costs (recirculation pumps, optimum temperature maintenance and UF operation), O_2 supply cost and sludge management cost (transport and disposal) have been taken into consideration while for NF only the energy costs for its operation have been considered. OC_{TAMR}, OC_{NF} have been calculated as reported in Eq.3-4:

$$OC_{TAMR}[\text{€ m}^{-3}] = EC_{TAMR} * E + (OXC_{TAMR} * OX + FSP * CSDT) * \ \mu COD_{TAMR} * COD_{IN} \qquad (3)$$

$$OC_{NF}[\text{€ m}^{-3}] = EC_{NF} * E \qquad (4)$$

With:

EC_{TAMR} [kWh m^{-3}]: energy consumption per unit of treated volume of the TAMR

EC_{NF} [kWh m^{-3}]: energy consumption per unit of treated volume of the NF

E [€ kWh^{-1}]: cost of electricity, assumed equal to 0.11 € kWh^{-1} [34]

OXC_{TAMR} [kgO_2 $kgCOD_{removed}^{-1}$]: pure oxygen consumption of TAMR

OX [€ kgO_2^{-1}]: cost of pure oxygen, assumed equal to 0.05 € kgO_2^{-1} [35]

FSP [kg_{TSS} $kg_{CODremoved}^{-1}$]: factor of sludge production, assumed equal to 0.1 kg_{TSS} $kg_{CODremoved}^{-1}$ in the range of 0.05–0.3 kg_{TSS} $kg_{CODremoved}^{-1}$ proposed by Collivignarelli et al. (2019) [26]

CSDT [€ kg_{TSS}^{-1}]: cost of sludge disposal and transport, assumed equal to 400 € kg_{TSS}^{-1} Considering an average value among those reported by Foladori et al. (2010) and Kalderis et al. (2010) [36,37]

μCOD_{TAMR} [-]: COD removal efficiency achieved by TAMR, assumed equal to 0.80 [38]

COD_{IN} [kg m^{-3}]: COD ingoing to the TAMR process

Instead, the operating cost per unit of mass of the adsorption on AC (OC_{AC}) has been calculated considering the amount of carbon used in the experimentation per unit of treated volume (M_{AC}, expressed in kg m^{-3}) and the cost of AC per unit of mass (CM_{AC}, assumed equal to 0.7-1.3 € kg_{AC}^{-1} converting the range 0.8-1.5 US$ kg_{AC}^{-1} obtained by Bhagat et al. (2018) and De Gisi et al. (2016) [39,40]), as reported in Eq.5.

$$OC_{AC}[\text{€ m}^{-3}] = M_{AC} * CM_{AC} \qquad (5)$$

A specific energy consumption indicator (*ECI*) is defined as the ratio between the energy consumption and another relevant parameter in the process (in this case the surfactants removal yield) [41]. For configurations of processes D (TAMR+NF), E (NF+AC) and F (TAMR+NF+AC), *ECI* has been calculated both for TAS and MBAS removal, according to the following equation (Eq.6):

$$ECI_x[\text{€ } kg_{removed}^{-1}] = \frac{OC_x * FR}{SF_{rem}} \qquad (6)$$

With:

OC_x [€ m^{-3}]: operating costs of the processes per unit of treated volume

FR [m^3 d^{-1}]: daily flow rate, constant at 245 m^3 d^{-1}

SF_{rem} [kg d^{-1}]: load of TAS or MBAS removed by the configuration of the processes

The analysis was subsequently performed using Excel MS and Origin.

3. Results and discussion

3.1. Characterization of the industrial wastewater

In table 4, the results obtained for the characterized parameters of the raw laundry WW influent in the full-scale treatment system and a comparison with some values found in the literature are presented. Data showed high COD, N-NH_4^+ and surfactants concentration. The ToS concentration was more than 700 mg L^{-1}. However, the chemical characteristics of WW produced by industrial laundry generally can present variations due to procedures adopted by each industry [42]. Indeed, by comparing the data obtained in this experimentation with some provided by the literature, a great variability can be observed.

The COD had a higher value than that obtained by Ciabattia et al. (2009), Bering et al. (2011) and Šostar-Turk et al. (2005) [43–45], but lower than our previous experiments [24,46]. The N-NH_4^+ was similar to that showed by Collivignarelli et al. (2017) [24] but higher than Collivignarelli et al. (2017), Bering et al. (2011) and Šostar-Turk et al. (2005) [44–46] studies. The surfactants concentration was one order of magnitude higher than that obtained by Ciabattia et al. (2009) and Šostar-Turk et al. (2005) [43,44], but lower than that obtained in Collivignarelli et al. (2017) [46] experimentation.

Table 4: *Range of values of the characterized parameters of the raw WW influent in the full-scale treatment system and comparison with some values found in the literature. n.a.: not available; ToS: Total surfactants. For interpretation of the localization of P0, the reader should refer to Figure 1. (double-column table)*

Parameters	Raw influent WW (sampling in P0)	Ciabattia et al. [43]	Collivignarelli et al. [24][a]	Collivignarelli et al. [46][a]	Šostar-Turk et al. [44][b]	Bering et al. [45][b]
COD [mg L^{-1}]	10,000-20,000	400-1000	23,980-51,450	18,000-104,000	280	1159
N-NH_4^+ [mg L^{-1}]	100-300	n.a.	67-306	0.5-70	2.5	1.6
ToS [mg L^{-1}]	25-750	n.a.	750-1461	n.a.	n.a.	n.a.
TAS [mg L^{-1}]	5-680	1-10	104-465	20-17,000	n.a.	43.7
MBAS [mg L^{-1}]	20-70	1-15	403-1357	95-13,000	10.1	32.9

[a]**:** These values has been obtained taking into considerations different WW used in this experimentation

[b]**:** A single value has been reported in this paper

3.2. Full-scale plant monitoring

The full-scale plant has been monitored daily for more than 3 months in order to examine the concentration of surfactants. The concentration of TAS and MBAS have been extremely variable during the monitoring period as typical of real laundry WW [46–48]. In particular, in the case of TAS there were 2 significant peaks (> 650 mg L^{-1}) after 18d and 96d from the beginning of the experimentation (Figure 2A). This aspect allowed to test the performances of the processes also in stress conditions. Instead, the inlet concentration of the MBAS remained generally at lower values (20-70 mg L^{-1}) (Figure 2B).

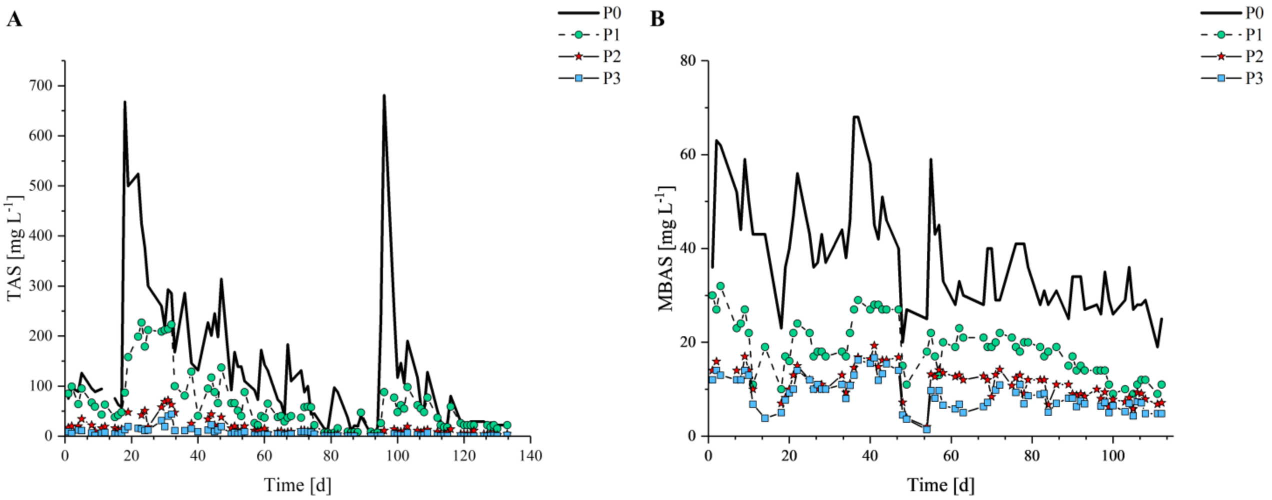

Figure 2: *Trend in the concentration of TAS* ***(A)*** *and MBAS* ***(B)*** *during monitoring of full-scale WW treatment system (Flow rate constant at 245 m^3 d^{-1}). For interpretation of the localization of P0, P1, P2 and P3, the reader should refer to Figure 1. (COLOR) (double-column figure)*

3.2.1. TAS and MBAS removal

The removal yields of TAS and MBAS substantially depended on the type of process adopted. In Figure 3 A,B, the removal yields of TAS and MBAS, with inlet and out concentration values, for single (TAMR, NF, AC) and combined treatments (TAMR+NF, NF+AC, TAMR+NF+AC) are reported.

Considering that one of the purposes of this study is to evaluate the performance of TAMR, NF and AC and their combinations in the treatment of real laundry WW, in Table 5 the Relative Standard Deviation (RSD) of the influent load of TAS and MBAS and the removal yields (μ) of the different combinations of processes are reported.

The NF demonstrated to be the best single treatment for TAS removal (74.1 ± 2.0%). TAMR allowed to obtain the 47.8 ± 5.1% of removal demonstrating the low biodegradability of these types of surfactants. In fact, the removal can be attributed to thermophilic biological degradation in TAR reactor and not to filtration on the following UF membrane. Considering that typical molecular weights of the surfactant ranged from 0.2-0.4 kDa, according to its molecular structure, and the molecular weight cut-off of the UF membrane used in this study was 30 kDa. Thus, the greater part of surfactants (TAS and MBAS) can pass through the pores of UF membranes [28]. Regarding couple treatments yields, TAMR+NF+AC permitted to remove almost totally the TAS (95.3 ± 0.8%) while NF+AC allowed to remove 91.4 ± 1.2%.

In this experiment, single treatments have generally proved to be not very effective in removing MBAS. In this case, the best removal yields were obtained with the TAMR biological process (49.5 ± 2.8 %). This result is confirmed by the literature, in fact the MBAS are generally more biodegradable due to their structure more easily attacked by microorganisms [49].

Data in Table 5 also suggested that AC appeared to be ineffective on MBAS removal. At first glance, this result could be related to the fact that high COD values in the WW generated a sort of competition for adsorption on AC. However, the significant removal of TAS in the same WW by means of AC suggested that COD did not interfere with the correct adsorption of surfactants. At the same time, it should be remembered that the TAMR (which preceded the AC process) showed in the previous experimentations a yield of COD removal equal to 80% [38]. Therefore, this behaviour would seem to have two different possible explanations: (i) adsorption on AC was not effective on removing all type of MBAS, or (ii) the short length of the molecules chain reduced the adsorption phenomenon.

Several studies have shown that adsorption on AC is a valid and effective process for the removal of MBAS [7,42]. However, commercial ACs exhibited relatively low adsorption capacity on some types of MBAS (e.g. PFOA) [50]. This would deserve a deeper understanding, which is out of the scope of the present paper.

Moreover, the explanation could be also attributed to the short length of the molecules. As explained by Du et al. (2015) [50], the long-chain MBAS are more preferential to be adsorbed on the AC than the short-chain ones in the competitive adsorption process. As the MBAS are generally more easily biodegradable [49] than the TAS, the biomass of the TAMR system (which preceded the adsorption on AC – see Figure 1) would be able to reduce the length of the MBAS molecular chain making the subsequent adsorption on AC more difficult. This would also explain the lower efficacy of NF alone on MBAS in the TAMR effluent.

The combined processes exhibited conflicting results. While the NF+AC gave totally unsatisfactory removal yields (52.5 ± 2.8 %), the combination TAMR+NF and TAMR+NF+AC showed much more encouraging results: 69.8 % and 76.7 % respectively. Therefore, also in this case, the ideal configuration was composed of the thermophilic biological process (TAMR) as pre-treatment and the physical treatments (NF) and chemical-physical (AC) as polishing treatments.

3.2.2. Resilience and resistance of the TAMR

One of the main aspects that can be highlighted is the ability of TAMR system to reduce the concentration peaks of surfactants present in the WW, especially the TAS (Figure 3). Regarding this, the TAMR proves to be endowed with a high degree of stability considering a load of surfactants in the input which is considerably variable. This behaviour can be noted in detail in Figure 4.

In fact, comparing surfactants concentrations in P0 and P1 (that represent the input and the out of the TAMR), the range of variation of concentrations is definitely lower in P1 than in P0, especially regarding the TAS. This aspect can be explained referring to the particular characteristics of resilience of the TAMR, as reported in our previous study [38], in which the thermophilic biomass was able to cope with peaks of pollutants and rapidly reactivate its optimal performance, after the stress conditions. Furthermore, despite the stress caused by the input surfactant peak, TAMR managed to guarantee removal rates around 50% (Table 5). This behaviour is very significant given that, as also demonstrated in this experimentation (Figure 2), the concentration range of surfactants in a real laundry WW is very variable [47].

Palmer and Hatley (2018) [7] evidenced also that high concentration of surfactants (around at 1,000 mg/L) depolarised the bacterial cell wall and therefore destroyed structure and function of the biomass. Despite the high ToS concentrations, in this experimentation, the results suggested that TAMR biomass was not inhibited and destroyed. This aspect can be related with the high resistance of thermophilic biomass that allowed TAMR to treat WW with very high concentration of surfactants [24]. Thus, TAMR proved to be able to perform good pre-treatment activity.

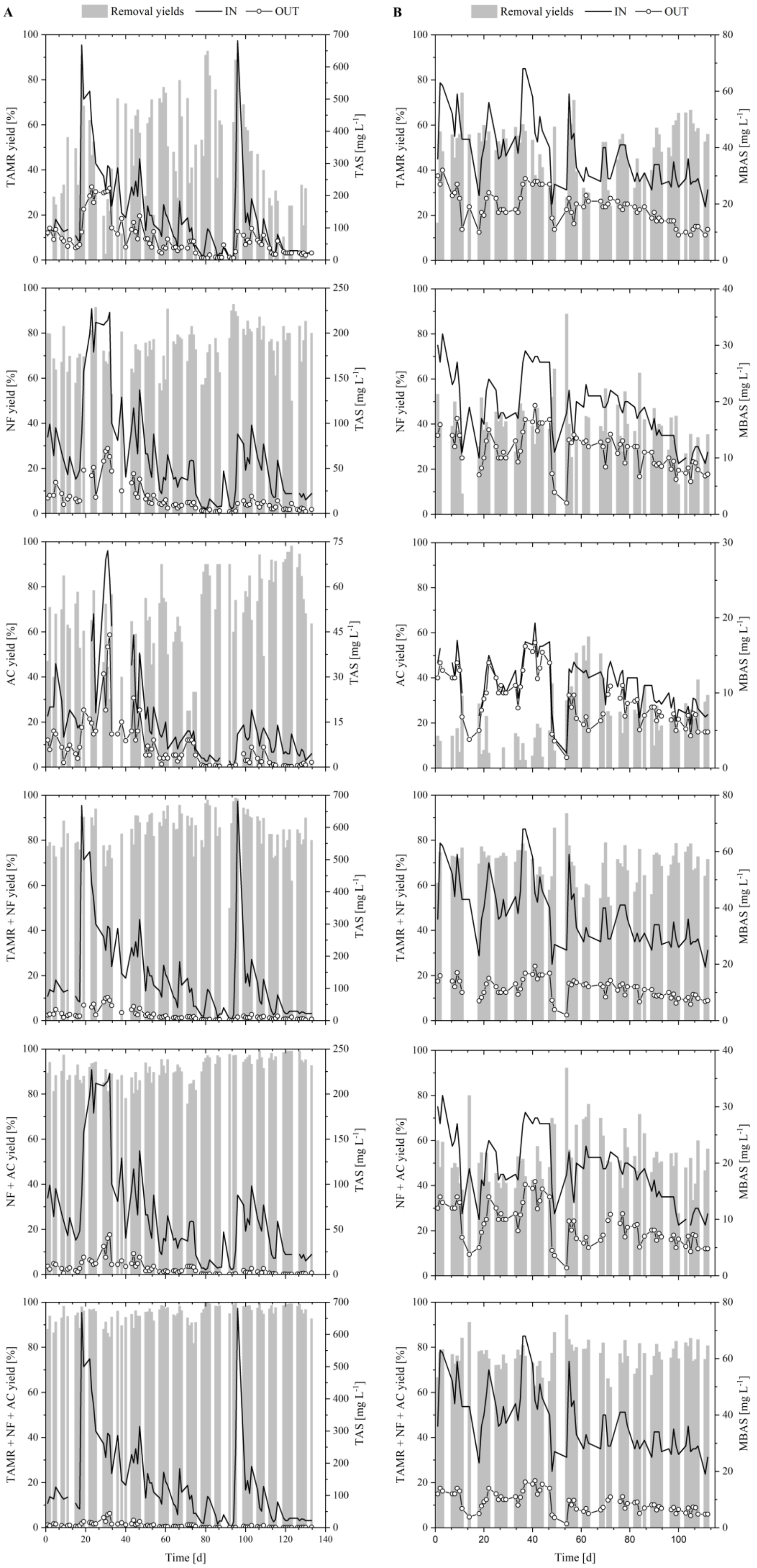
A
Removal yields
IN
OUT
TAMR yield [%]
TAS [mg L^{-1}]
NF yield [%]
AC yield [%]
TAMR + NF yield [%]
NF + AC yield [%]
TAMR + NF + AC yield [%]
Time [d]
B
MBAS [mg L^{-1}]

Figure 3: *Removal yields, inlet and out concentration values of (**A**) TAS and (**B**) MBAS for single (TAMR, NF, AC) and combined treatments (TAMR+NF, NF+AC, TAMR+NF+AC) during three months of experimentation (Flow rate constant at 245 m^3 d^{-1}). (double-column figure)*

Table 5: *Relative Standard Deviation (RSD) of the influent load of TAS and MBAS and removal yields (μ) of the different combinations of processes. (double-column table)*

Process	**TAS**		**MBAS**	
	RSD$_{IN}$ [%]	**μ [%]**	**RSD$_{IN}$ [%]**	**μ [%]**
TAMR	103	47.8 ± 5.1	30	49.5 ± 2.8
NF	90	74.1 ± 2.0	30	39.3 ± 2.8
AC	102	67.6 ± 4.3	30	22.9 ± 3.2
TAMR + NF	103	84.6 ± 1.8	30	69.8 ± 1.8
NF + AC	90	91.4 ± 1.2	30	52.5 ± 2.8
TAMR + NF + AC	103	95.3 ± 0.8	30	76.7 ± 1.4

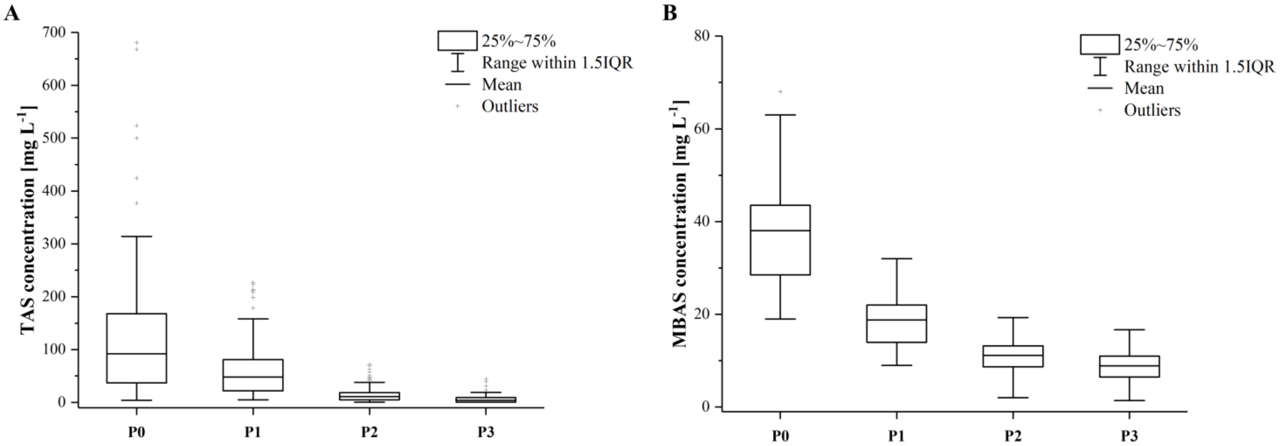

Figure 4: *Concentration values in P0, P1, P2 and P3 of (**A**) TAS and (**B**) MBAS during three months of experimentation (Flow rate constant at 245 m^3 d^{-1}). For interpretation of the localization of P0, P1, P2 and P3, the reader should refer to Figure 1. Boxplots represent the distance between the first and third quartiles while whiskers are set as the most extreme (lower and upper) data point not exceeding 1.5 times the quartile range from the median. Values outside such a range are outliers. (double-column figure)*

3.3. Costs analysis

The costs were assessed by monitoring the consumption of electricity, biological sludge transport and disposal, pure oxygen, and activated carbon used in the experimentation, and elaborated as described in Section 2.3.4. It should be noted that the analysis took into account only the main operational aspects and not the costs related to the investment which are out of the scope of the present paper. During the monitoring period, energy consumption per unit of treated volume of the TAMR and NF (*EC*) have been evaluated equal to 10 kWh m^{-3} and 45.5 kWh m^{-3}, respectively. The pure oxygen consumption of TAMR (OXC_{TAMR}) has been found equal to 1.1-1.2 kgO_2 $kgCOD_{removed}$. Therefore, the operating costs of the processes per unit of treated volume (*OC*) have been calculated as reported in Table 6. Regarding the TAMR treatment, the cost analysis showed that the main item of expenditure has been the electricity consumption (49%), followed by the supply of O_2 (30%) and the transport and disposal of the biological sludge produced (21%). Due to the very low production of biological sewage sludge by the TAMR, the economic weight of the disposal of biological sludge production was lower than that of other processes such as moving bed biofilm reactor (MMBR) that accounted for about 26% [45]. The results suggested that the *OC* of TAMR was greater than that of adsorption on AC but significantly lower than NF. These values are in accordance to those reported in literature. For instance, Samhaber and Nguyen (2014) [51] found that the operating costs of the industrial NF treatment processes per unit of treated volume ranged from 1 to 6 US$ m^{-3}. On the contrary, the results of the present experimentation are higher than those obtained, for instance, by Ciabattia et al. (2009) [43] with a complex system composed by a physico-chemical pre-treatment, sand filtration, ozonation, GAC filtration and UF (0.81 € m^{-3}). However, in this specific case, TAS and MBAS were present in concentrations two orders and one order of magnitude lower than the present experimentation, respectively.

***Table 6:** The operating costs of the processes per unit of treated volume (OC). (single-column table)*

Parameters	**Values**
OC_{TAMR} [€ m^{-3}]	2.24
OC_{NF} [€ m^{-3}]	5
OC_{AC} [€ m^{-3}]	1.5

Taking into account the fact that during the experimentation the best removal yields were obtained with the combination of different processes, the cost analysis was carried out analysing the most effective configurations: D (TAMR+NF), E (NF+AC) and F (TAMR+NF+AC).

In order to obtain the energy consumption indicator (*ECI*), *OC* values have been correlated with the surfactants removal yields, the most relevant parameter. Therefore, *ECIs* have been calculated for TAS and MBAS removal, for combinations of processes: D, E and F.

The energy consumption indicators (*ECI*, expressed in € $kg_{removed}^{-1}$) were studied in a logarithmic graph with the influential load of TAS and MBAS in order to understand a correlation with the influent load of surfactants (*SF*, expressed in kg d^{-1}). Therefore, the power law was applied (Eq. 7):

$$ECI\left[€\ kg_{removed}^{-1}\right] = a * SF^{b} \qquad (7)$$

Regarding the removal of TAS (Figure 5A), the results showed that the combinations TAMR+NF and NF+AC had similar costs of removal, both when the influent TAS load was high, and when it was low. Also, the yields in terms of removal were very similar (84.6% and 91.4%). Therefore, from the economic point of view, it can be stated that the combinations D and E have been completely comparable. The complete chain of treatments (TAMR+NF+AC) showed instead the higher cost, in relation to the quantity of TAS removed. This result was due to the fact that it was a very thorough treatment (over 95% removal). Therefore, the results of the costs analysis would suggest that the combination of TAMR+NF+AC processes could be adopted when the required percentage of TAS removal is close to 100%.

The results on the removal of the MBAS showed that the combination of NF+AC processes has been the most convenient only with low input loads (< 8 kg d^{-1}). Instead, when the influential load is greater (a typical situation with a real laundry WW), the economically optimal solution turned out to be the combination TAMR+NF. As in the case of TAS, the results suggested that the combination of TAMR+NF+AC has been the most effective but at the same time the most expensive and therefore this combination could be adopted only when the required percentage of surfactant removal is close to 100%.

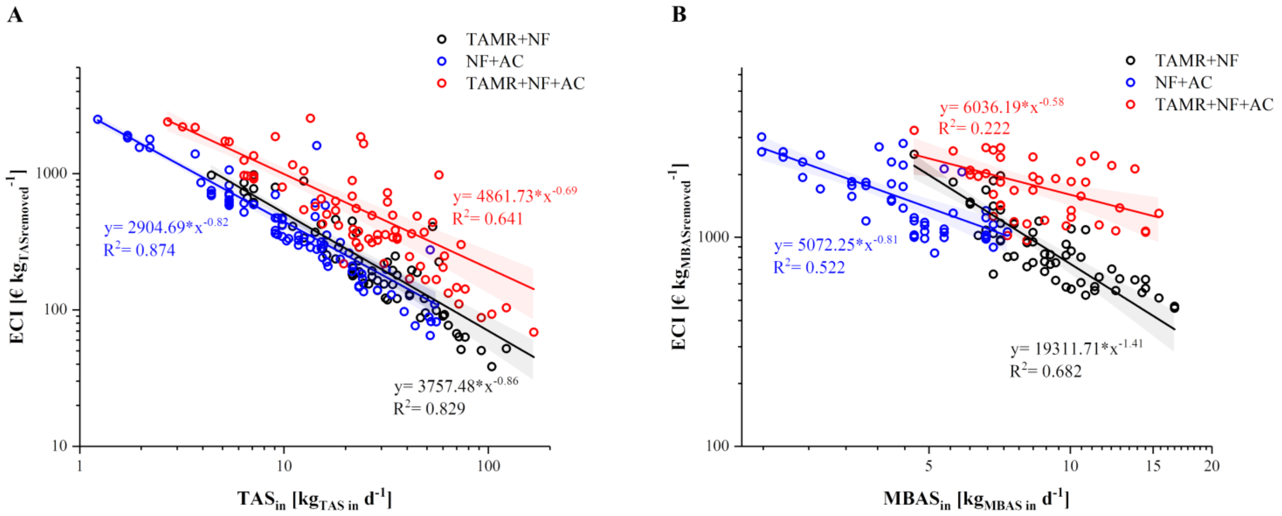

Figure 5: *Energy consumption indicator (ECI) for combined processes (TAMR+NF, NF+AC, TAMR+NF+AC) as a function of the influent load of (**A**) TAS and (**B**) MBAS. The coloured bands represent the 95% confidence interval. (For*

interpretation of the references to colour in this figure legend, the reader should refer to the Web version of this article). (COLOR) (double-columns figure)

4. Conclusions

For more than three months, a full-scale plant consisting of biological (TAMR), physical (NF) and chemical-physical (AC) processes in order to treat real laundry WW was monitored. Firstly, the monitoring period allowed to confirm that the concentration of surfactants in the laundry WW are very variable. Moreover, the removal yields of six different possible combinations of these processes against TAS and MBAS were evaluated.

Given the results, the optimal solution can be considered the combination TAMR+NF; TAS and MBAS removal yields have been 85% and 80%, respectively. In fact, as demonstrated, TAMR process was able to withstand high stress conditions due to sudden load peaks and resist to a high concentration of surfactants while keeping the removal yields of surfactants almost constant, allowing the TAMR to carry out an effective pre-treatment activity. The NF has guaranteed, if coupled with the thermophilic biological treatment, an excellent polishing treatment. Moreover, among all the combinations of treatments analysed, the combination TAMR+NF required lower costs per mass unit of surfactants removed.

The TAMR+NF+AC combination allowed almost complete removal of TAS (over 95%) and MBAS (> 89%) but the costs per unit of mass removed were high. Therefore, these outcomes suggested that the combination of TAMR+NF+AC processes could be adopted when the required percentage of surfactant removal is close to 100%.

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Conflict of interests

The authors have declared no conflict of interest.

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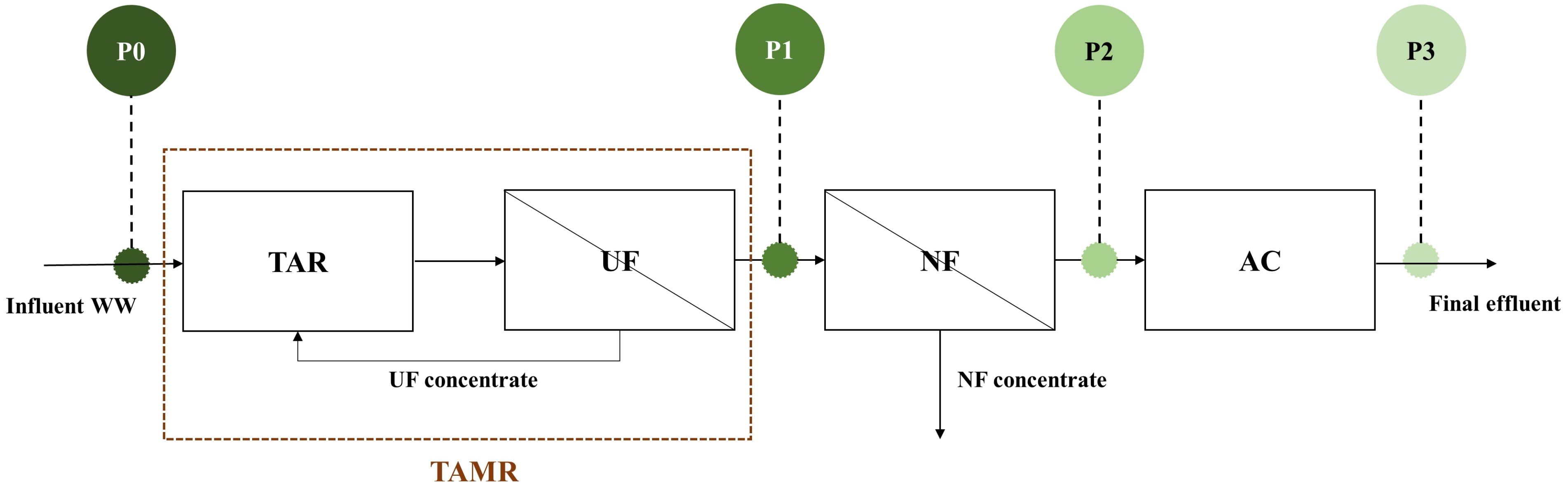
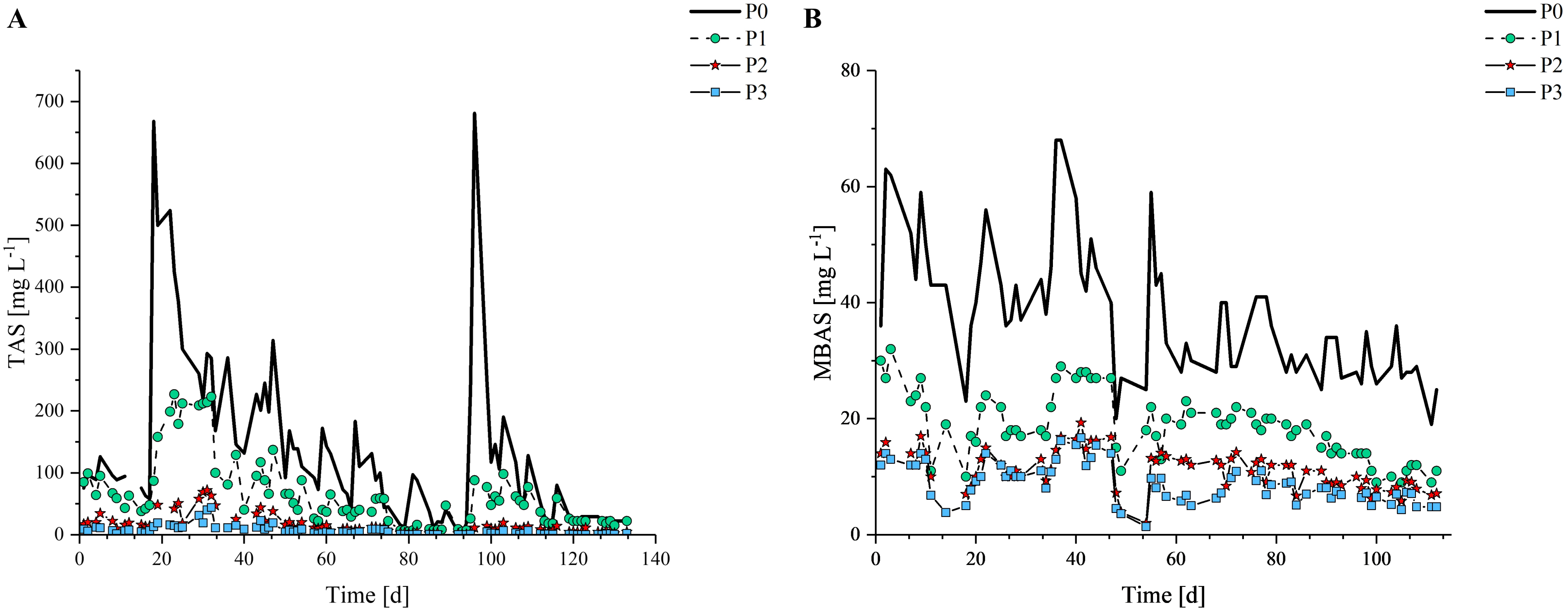
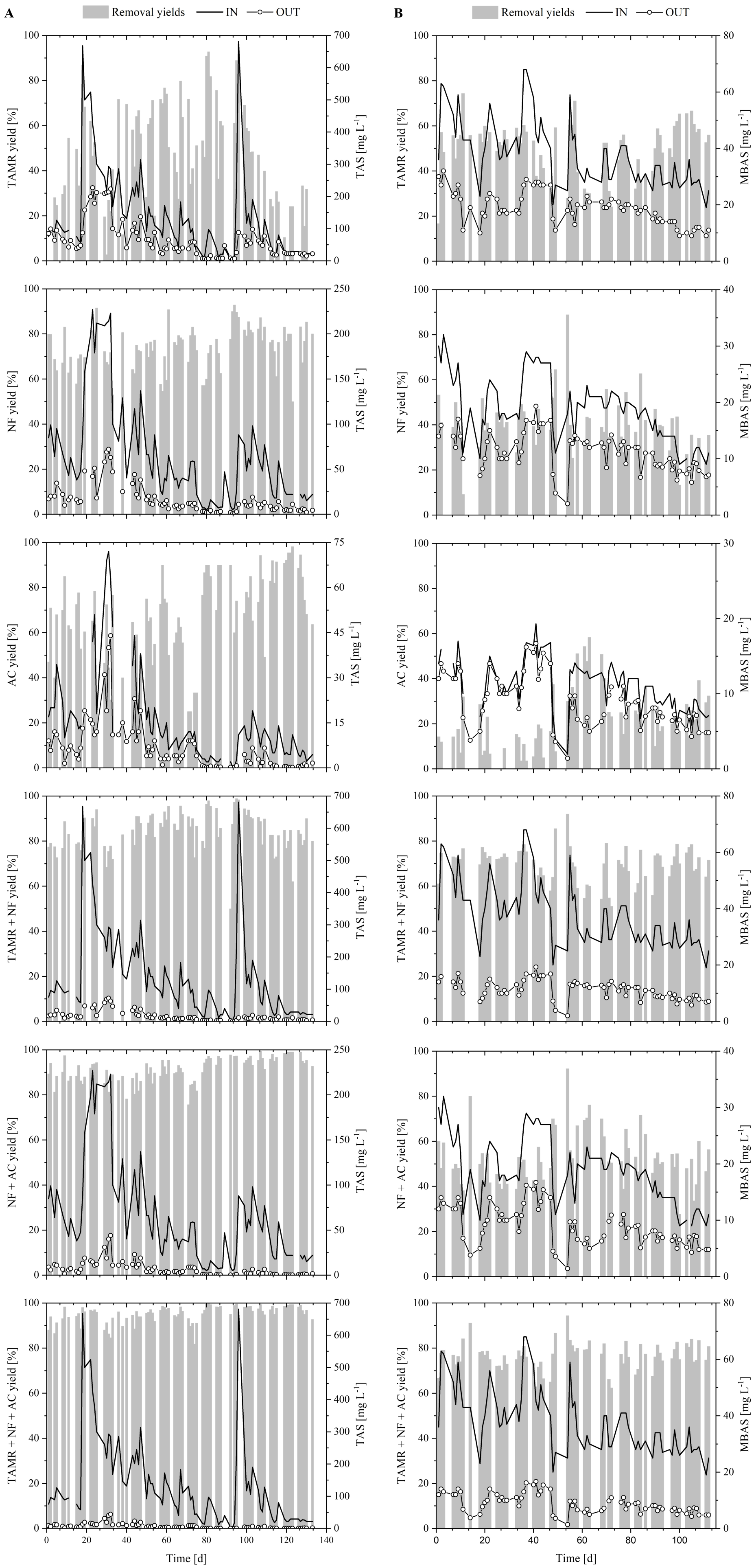
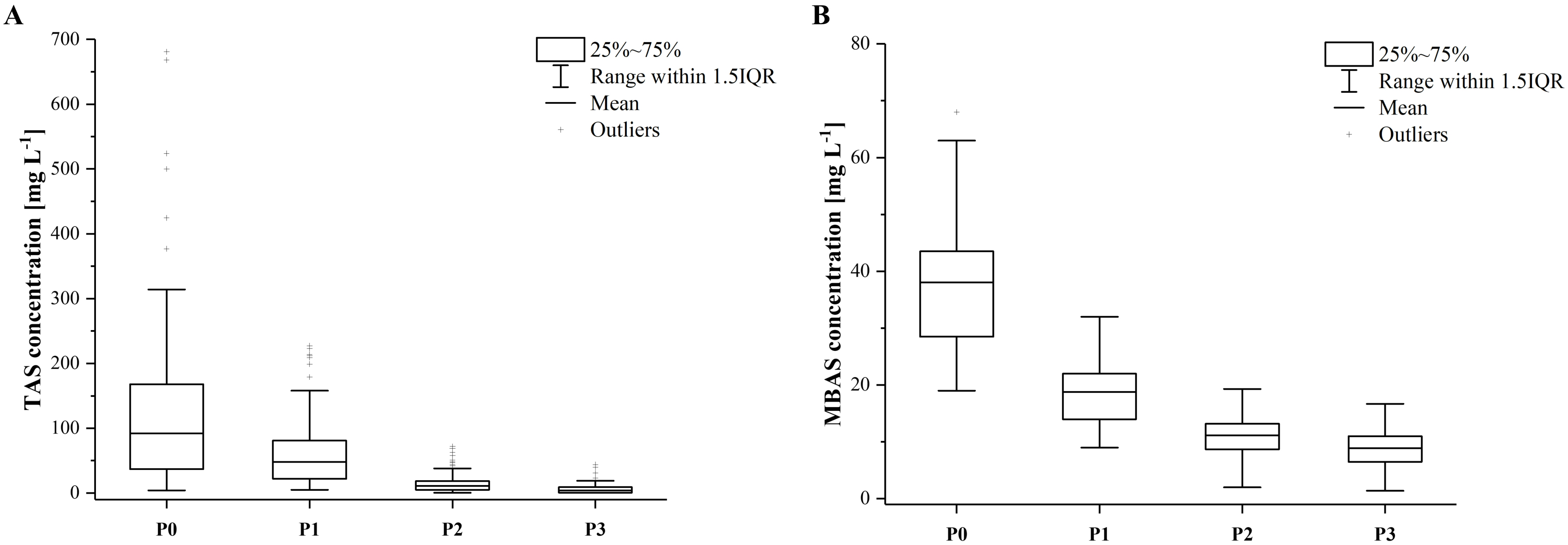
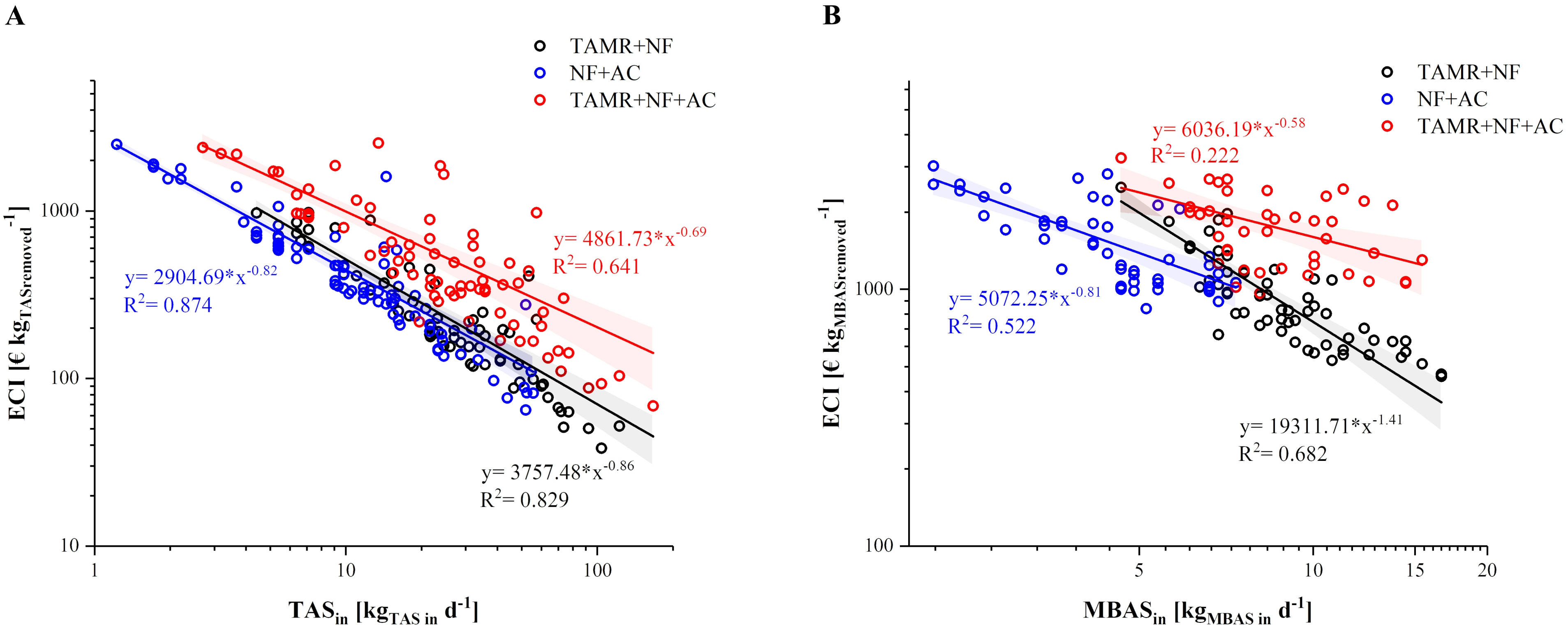

P0
P1
P2
P3
Influent WW
TAR
UF
NF
AC
Final effluent
UF concentrate
NF concentrate
TAMR


A
TAS [mg L^{-1}]
0
100
200
300
400
500
600
700
0
20
40
60
80
100
120
140
Time [d]
P0
P1
P2
P3
B
MBAS [mg L^{-1}]
0
20
40
60
80
0
20
40
60
80
100
Time [d]
P0
P1
P2
P3


A
B
Removal yields
IN
OUT
TAMR yield [%]
NF yield [%]
AC yield [%]
TAMR + NF yield [%]
NF + AC yield [%]
TAMR + NF + AC yield [%]
TAS [mg L^{-1}]
MBAS [mg L^{-1}]
Time [d]
0
20
40
60
80
100
120
140
200
300
400
500
600
700
50
150
250
15
30
45
60
75
10
25
35


A
TAS concentration [mg L^{-1}]
0
100
200
300
400
500
600
700
P0
P1
P2
P3
25%~75%
Range within 1.5IQR
Mean
Outliers
B
MBAS concentration [mg L^{-1}]
0
20
40
60
80
P0
P1
P2
P3
25%~75%
Range within 1.5IQR
Mean
Outliers


A
TAMR+NF
NF+AC
TAMR+NF+AC
y= 4861.73*x^-0.69
R^2= 0.641
y= 2904.69*x^-0.82
R^2= 0.874
y= 3757.48*x^-0.86
R^2= 0.829
ECI [€ kg_TASremoved^-1]
10
100
1000
1
10
100
TAS_in [kg_TAS in d^-1]
B
TAMR+NF
NF+AC
TAMR+NF+AC
y= 6036.19*x^-0.58
R^2= 0.222
y= 5072.25*x^-0.81
R^2= 0.522
y= 19311.71*x^-1.41
R^2= 0.682
ECI [€ kg_MBASremoved^-1]
100
1000
5
10
15
20
MBAS_in [kg_MBAS in d^-1]