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# Holistic managements of textile wastewater through circular, greener and eco-innovative treatment systems developed by minimal to zero liquid discharge

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ABSTRACT

New pragmatic and viable solutions to reduce or prevent discharge and to protect reserves are currently among the top-prioritised research for cleaner, circular, and resource-efficient use of industrial waters. So, the development of eco-sustainable water management is essential for green industrial development that will meet versatile and eco-sensitive regulatory standards, especially in water-intensive industries. Textile wastewater was reclaimed in semi to fully closed loops for minimal to zero liquid discharge. Concentrate-mixed wastewater was steadily treated in a hybrid membrane oxidation reactor at 60–80 % synergistic performances with remarkable UF fluxes of 96.4–820 L/m$^2$h without any sludge discharge. Effluent was purified with 90–100 % removals and 20–80 L/m$^2$h in nanofiltration and reverse osmosis. Due to Fenton-specific operation, more handling by ion exchange and neutralisation required to harvest membrane reuse waters and reactor discharge effluents with guaranteed Fe and pH. All-in-one system simulations indicated that high quality reuse waters are produced by 99.9 % efficiency and 98 and 100 % savings in iron and acid but 20–51 % more oxidant through concentrate recycling and regenerant reuse. It was also revealed that reactor effluents can be released to the sea or conventional biological treatment or can be eco-sustainably exploited for in-situ chemical and ex-situ bio-induced recovery of vivianite. This research demonstrates that how textile wastewater can be managed holistically by liquid discharge approaches from 50 % minimal to 99.9 % zero just in two-step, *i.e.* pretreatment and pre-concentration, with consumable minimisation and valuable waste recovery through the eco-innovative systems which are developed as circular, greener, and sludge-free compatible with sustainable development goals.

## 1. Introduction

As of climate crisis, now a commonplace fact that industrial development being executed in a way that suppresses eco-sustainable management of water, resource, and waste threats the ecosystem continuity. New approaches preventing water pollution, recycling wastes, and reducing reserve supply need to be created to compensate deficits and inefficiencies in providing circularity of pollutants in wastewaters. Thus, today's cure priorities should be set on water scarcity and put into practice onto green innovations with environment and energy efficient water recovery, resource usages, waste reductions and recycling, and minimal and even, if can make, zero emissions to all the natural environments at optimised costs [24,78]. Initially, to move on to a green phase where fresh waters are eco-protectively managed, it is crucial to develop more diligent solutions to be willingly accepted by society and target industries by meeting the costs with lower energy or offering less source and cost. Considering industry 4.0, the most prised prospect for the green management of industrial wastewaters would be upon the resolutions provided on reuses of industrial waters by the closed-circuit recoveries. This strategy would not only lead to recover reusable water out of process water, especially in water-intensive industries, but also to reduce water costs by prudently conserving current reserves [67,25].

Currently, most conventional wastewater technologies may not be

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**Nomenclature**

| | |
|---|---|
| *A* | apparent water permeability coefficient (L/m$^2$hbar) |
| *A*/*B* | water/solute permselectivity (1/bar) |
| AOP | advanced oxidation process |
| [AOP/UF] | advanced oxidation process and ultrafiltration hybrid reactor |
| AOX | adsorbable organic halogens |
| *B* | apparent solute permeability coefficient (L/m$^2$h) |
| CE | circular economy |
| COD | chemical oxygen demand |
| DO | dissolved oxygen |
| {Fenton/UF} | Fenton and submerged UF hybrid reactor |
| HFO | hydrous ferric oxide |
| IE | ion exchange |
| IWSA | Istanbul water and sewerage administration |
| KWSA | Kocaeli water and sewerage administration |
| MBR | membrane bioreactor |
| MF | microfiltration |
| MLD | minimal liquid discharge (50–80 % water recovery) |
| MOR | membrane oxidation reactor |
| {MOR} − [NF/RO] | MOR-integrated single or dual NF and RO systems |
| {MOR} − [NF$_{tight}$] | MOR-integrated tight NF system |
| {MOR} − [RO] | MOR-integrated RO system |
| {MOR} − [NF$_{loose}$ + RO]: | MOR-integrated loose NF and RO system |
| NF | nanofiltration |
| [NF/RO] | single or dual NF and RO processes |
| [NF$_{loose}$ + RO] | loose NF and RO dual membrane system |
| NF$_{tight}$ | tight NF |
| NTU | nephelometric turbidity unit |
| NZLD | near-zero liquid discharge (80–95 % water recovery) |
| PES | polyethersulfone |
| PVDF | polyvinylidene fluoride |
| *R* | rejection (%) |
| RO | reverse osmosis |
| SS | suspended solids (mg/L) |
| TDS | total dissolved solids (mg/L) |
| TN | total nitrogen (mg/L) |
| TOC | total organic carbon (mg/L) |
| TSS | total suspended solids (mg/L) |
| UF | ultrafiltration |
| UVA or UVA$_{365}$ | ultraviolet A (at 365 nm) |
| {UVA-Fenton/UF} | UVA-Fenton and submerged UF hybrid reactor |
| UVC or UVC$_{254}$ | ultraviolet C (at 254 nm) |
| {UVA-Fenton/UF} | UVC-Fenton and submerged UF hybrid reactor |
| ZLD | zero liquid discharge (>95 % water recovery) |

sufficient to meet all aspects of final effluent limitations. For instance, typical chemical and biological treatment techniques leave much not to be desired as they do not adequately treat wastewater with producing low-volume non-toxic residues. Further, many existing treatment technologies require the use of harmful chemicals and considerable energy for operations such as pumping and aeration, and their sludges and gaseous wastes are usually problematic [6,44]. Given complex natures and distinct qualities of industrial wastewaters, it is unlikely that a single method is best for available technology and regulatory obligations [59]. Here, developing hybridised or integrated processes, combining advantages and minimising drawbacks will offer the cost and time-efficient improvements. In this way, the treatments of industrial wastewaters would be more effective and less costly than alone processes of interest [65,56]. Hereby, it is worth mentioning that a such-way innovation, which can also allow the expansion of green and clean technologies, would be so welcome that it would help to achieve green industrial development, harmoniously with the Porter hypothesis [44,11].

As one of the main target industries, the textile sector uses large amounts of water in manufacturing, generates various types of contaminants such as acid, dye, salt, phosphate, nitrogen, phenol, and metals. Many organics in textile wastewater having complex and recalcitrant nature are resistant to degradation and pose significant risks through accelerating oxidative stress and genotoxicity to all living organisms. Due to the increasing costs of industrial water, the sector is leading efforts to produce reusable effluents for resource conservation and to comply with stricter regulations [26,41,64,14,5,43]. This regular continuity suggests that conventional treatment solutions will evolve to become more reliable and sophisticated with the holistic management transitions from minimal to zero pollution faster than in other industries. Strategies to reduce or eliminate the discharge of liquid pollutants through all-inclusive managements from minimal liquid discharge (MLD) to zero (ZLD) will be absolute milestones for substantial and progressive developments towards eco-sustainability. They will not only contribute to the preservation of ecosystem integrity but will also strengthen the pillars of circular economy (CE) model which stands out as modern sustainable development tool promoted by the European Union [24,46,10,48].

In view of the above, the development of cleaner and more sustainable water management practices that meet the versatile and stringent regulatory requirements by improving treatment efficiencies is an eminently popular research topic to achieve a green progress vision, especially for the textile industry. Enhanced hybrid reactors integrating advanced oxidation processes (AOPs) with a range of applicable membranes, offer a talented pathway in this focus through higher purification, less material and energy. Photocatalytic membrane integration is the one of the most common configurations and incorporates a photoactivated layer to reduce membrane fouling while increasing permeate flux. With photocatalysts suspended in the feed or immobilised on the membrane surface, the technique multiplies the benefits of improved contact between catalysts and pollutants by means of increased loading in suspension and repeated exposure of the photocatalyst surface to a light source. It is also noted that spent photocatalyst can be effectively separated and proven to be highly effective in long-term operation for reuse [65,56,59]. Membrane oxidation processes, which can be combined not only with (photo)catalytic oxidation but also with other AOPs, ensure improved effluent quality with simultaneous operations of uniform processes and are characterised by a smaller space, self-cleaning and fewer consumables, driven by two popular AOPs –Fenton and photo-Fenton– that are among the best [41,56,66,20,43,19].

Fenton and photo-Fenton oxidation has been successfully applied to textile wastewaters with moderate performance slightly above 50 % organic and 70 % colour [14]. A wide variety of Fenton combined membranes have been worked to treat paint and textile wastewaters by membrane bioreactor (MBR), and microfiltration (MF), ultrafiltration (UF) and nanofiltration (NF) [26,60,33]. For a paint wastewater, the combined systems were found to be 65 % more effective than the individual processes [26]. In the recent past, numerous integrated systems have been designed for the treatment and reuse of textile wastewater, and scientific innovations have emerged on the systems configured as integrated ozonation and photocatalysis MBR [60], and Fenton-assisted sequencing batch reactor [63], photo-Fenton based MF + AOP + NF and MF + NF + AOP [33], MF + NF and later AOP to NF concentrate [43], and sequential advanced oxidation and adsorption [76]. Despite these advances, a sole focus on water recovery and reuse and a failure to consider resource reduction, sludge disposal, energy, and cost issues as complementary enough to identify breakthrough solutions emerge as

**Table 1**
Physico-chemical characteristics of textile wastewater.

| Parameter | Symbol | Unit | Value |
|---|---|---|---|
| pH | pH | – | 6.22 ± 0.15 |
| Temperature | $T$ | °C | 25.0 ± 0.5 |
| Turbidity | – | NTU | 5.49 ± 2.73 |
| Electrical conductivity | $E_C$ | $\mu$S/cm | 1079 ± 95 |
| Total dissolved solids | TDS | mg/L | 565 ± 59 |
| Dissolved oxygen | DO | mg/L | 6.72 ± 0.98 |
| Chemical oxygen demand | COD | mg/L | 804 ± 189 |
| Total organic carbon | TOC | mg/L | 289 ± 79 |
| Suspended solids | SS | mg/L | 7.96 ± 2.58 |
| Adsorbable organic halogens | AOX | mg/L | 0.682 ± 0.020 |
| Phenol | $C_6H_5OH$ | mg/L | 8.57 ± 1.61 |
| Ammonia | $NH_4^+$ | mg/L | 1.88 ± 0.57 |
| Nitrate | $NO_3^-$ | mg/L | 6.83 ± 5.27 |
| Sulphate | $SO_4^{2-}$ | mg/L | 69.7 ± 39.5 |
| Chloride | $Cl^-$ | mg/L | 26.0 ± 1.18 |
| Ferrous iron | $Fe^{2+}$ | mg/L | 0.42 ± 0.47 |
| Ferric iron | $Fe^{3+}$ | mg/L | 1.49 ± 1.70 |
| Total iron | Total Fe | mg/L | 1.91 ± 2.17 |
| Total nitrogen | TN | mg/L | 8.89 ± 4.57 |
| Total hardness | – | mg $CaCO_3$/L | 35.8 ± 38.1 |
| Colour | $\lambda$ | Abs@436 nm | 0.150 ± 0.055 |
| | | Abs@525 nm | 0.153 ± 0.061 |
| | | Abs@620 nm | 0.172 ± 0.081 |

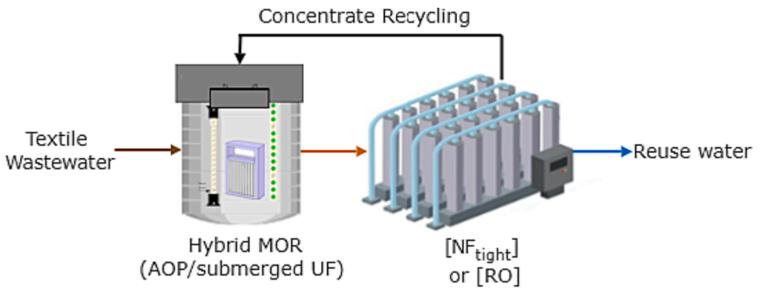
key ambiguities in hoping for green innovation and CE [49,25,78,10]. Many pragmatic and feasible solutions to current industrial problems are still so controversial for technology developers and producers [41] that removing uncertainties would be of the utmost urgency for rapid green development, not only in textile sector but in all other high-consuming industries.

This research was conducted to reveal the main arguments for water and resource recovery from textile wastewater with a technologically easy to apply and eco-soundly management of lower energy needs. The main framework depended on the determination of the experimental steady state operability of systems configured with eligible process sequences within water loops completely closed by the ZLD. Appropriately, the reclamations of textile wastewater using MOR, NF and/or RO ([NF/RO]) to produce a reusable water without the generation of concentrate effluent and residual iron sludge were examined to develop eco-innovative systems that allow superior water effectiveness than similar research works. Later, to eliminate the Fenton's specific iron and pH problems in the discharges and reuse waters, ion exchange (IE) and pH neutralisation were conveniently involved in real-scale designs having simulated based on experimental {MOR} – [NF/RO] performances along with software-created IE performances. From MLD approach to ZLD, all just in two-step processing, *viz.* pre-treatment and pre-concentration, which reduces systematic energy demand, holistic managements of textile wastewater were conducted with the performance-based practicable configurations developed as circular, greener, and sludge-free and knowledge on the current novel technological solutions or practices. To the best of our knowledge, this is the first all-inclusive pivotal study for readily applicable MLD and ZLD management strategies that would have a central role in promoting greener and eco-sustainable uses of industrial waters.

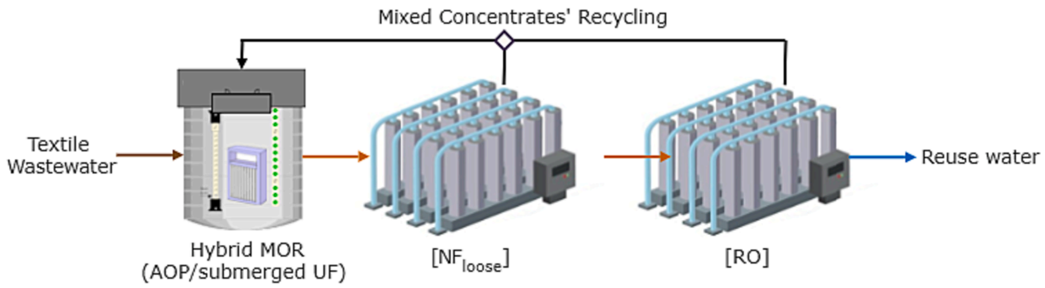
## 2. Materials and methods

### 2.1. Wastewater, membranes, and chemicals

A real wastewater from wash baths of a cotton dyeing and manufacturing factory in Sakarya (Türkiye) was used (Table 1). Three different commercial membranes were explored for reverse osmosis (RO), three for loose nanofiltration ($NF_{loose}$), three for tight nanofiltration ($NF_{tight}$), and two for ultrafiltration (UF) (Table S1). The Fenton or photo-Fenton reactions employed by using ferrous sulfate heptahydrate ($FeSO_4 \cdot 7H_2O$, 99.5 %) and hydrogen peroxide ($H_2O_2$, 35 % w/v). The pH of MOR influent was adjusted to the operating values by either sulfuric acid ($H_2SO_4$, 95–98 %) or sodium hydroxide (NaOH, 99.5 %). Hydrochloric acid (HCl, 32 %) along with NaOH was used as membrane cleaner. All analytical reagents were supplied by Merck and prepared using deionised water produced by Millipore Milli-Q.



(a) {MOR}-[$NF_{tight}$] and {MOR}-[RO] systems



(b) {MOR}-[$NF_{loose}$+RO] system

**Fig. 1.** Systematic layouts of hybrid MOR ([AOP/UF]) treatments, complemented with [$NF_{tight}$] or [RO] single (a) or [$NF_{loose}$ + RO] dual membrane filtration (b).

**Table 2**
Technical specifications and scaling-up data and information of lab-pilot MOR, and optimal operating conditions of Fenton and photo-Fenton supported UF.

| Parameters | Remark | Lab-pilot scale MOR | | |
|---|---|---|---|---|
| | | Fenton | UVA-Fenton | UVC-Fenton |
| – *Technical specifications* | | | | |
| · Lamp number | unit | – | 10 | 5 |
| · Irradiation power | W/lamp (W) | – | 20 (200) | 40 (200) |
| · Submerged module geometry | – | rectangular | | |
| · Membrane surface area, $A$ | $cm^2$ | 298 (8.5 cm × 35.0 cm) | | |
| · Geometric shape | – | cylindrical | | |
| · Volume | L | 20.0 | | |
| · Dimensions, $h/d/w$[a] | cm | 22.1/61.0/2.1 | | |
| – *Scale-up data*[b] | | | | |
| · Membrane area ratio | $cm^2/cm^2$ | 7.56 | | |
| · Reactor volume ratio | L/L | 6.67 | | |
| · Proportional deviation | % | 11.9 | | |
| · Cross-sectional area ratio | $cm^2/cm^2$ | 2.76[c] | | |
| · Operated lamp number | unit | – | 3[d] | 2[d] |
| – *Optimal operating conditions*[b] | | | | |
| · Process time, $t$ | min | 60 | 60 | 41 |
| · Temperature, $T$ | °C | 26.5 | 40 | 40 |
| · pH | – | 3.73 | 4.0 | 4.44 |
| · $H_2O_2$/TOC | g/g | 6.00 | 7.75 | 9.88 |
| · $H_2O_2/Fe^{2+}$ | g/g | 15.00 | 10.75 | 7.27 |
| · Aeration rate, $\nu_A$ | L/min | 5.0[c] | 2.8[c] | 8.0[c] |
| · Vacuum rate, $\nu_W$[e] | rpm (mL/min) | 80.0 (67.0) | 71.0 (60.0) | 64.7 (55.0) |
| · UF membrane type | – | UH050 | UV150 | UH050 |
| · Intensity of light | $mW/cm^2$ (W) | – | 0.4[d] (60)[d] | 0.6[d] (80)[d] |

[a] $h/d/w$ is the height/diameter/wall thickness of the MOR made of borosilicate glass.

[b] by hybridisation of UF and oxidation in the same reactor, the proportional deviation in scaling is at an acceptable level of 11.9% from the membrane and volume ratios of bench-scale and lab-pilot MORs. Hence, scaling-up of the lab-pilot MOR were fulfilled in line with the intensity of light and aeration rate to adapt the same conditions on the optimum performance of bench-scale MOR applied for textile wastewater [4].

[c] 2.76 ratio was obtained by dividing the cross-sectional area of the large reactor by that of the small to provide the equivalent aeration per unit cross-sectional area of the reactor. So, the aeration rate values for the Fenton and photo-Fenton operations in the large MOR were determined by means of multiplying this scaling ratio with the values optimized for benchtop reactor [4].

[d] Optimized intensities of light in the benchtop UVA-UVC operations were 0.4–0.6 $mW/cm^2$ with 3 lamps [4]. Lightening at the same conditions in the large operations was provided using 3 and 2 lamps equal to total irradiation powers of 60 and 80 W, respectively, considering the **Fig. S2**.

[e] vacuum rates of rpm are the values applied in the small reactor [4]. Those for large reactor equipped with a high-capacity peristaltic pump to filtrate reactor liquor at the same vacuum effect were applied with experimentally measured average water withdrawal flows providing that clean membrane water fluxes were the same at submerged filtrations of distilled water.

## 2.2. Applications of advanced treatment systems

Fig. 1 shows the major arrangements of integrated operations for purifying textile washing wastewater, including water recovery and concentrate recycling. The designs were based on UF immersed into the MOR hybridised by Fenton or photo-Fenton ($UVA_{365}$ and $UVC_{254}$) oxidations. The first pretreatment step involved treating the mixture of wastewater and concentrate with MOR. This was followed by a pre-concentration step, which included recovering water with either single ([$NF_{tight}$] or [RO]) or dual filtrations ([$NF_{loose}$ + RO]), and recycling the single or dual concentrates back to MOR. The concentrate-mixed wastewater treatment in MOR was conducted separately for the optimal Fenton or photo-Fenton conditions as determined in our previous study [4] and the reactor liquor was filtered by UF after treatment. The NF or RO concentrates were fed directly back into the MOR inflow to blend with raw wastewater in single operation to ensure prolonged oxidation of organics. In dual operations, both concentrates were mixed evenly with each other before being fed back into the MOR treatment. The systemic operations were executed semi-continuously by recycling concentrate outflows manually to the continuous MOR treatments after water recovery by NF and/or RO.

## 2.3. Experimental set-ups and operational procedures

### 2.3.1. Lab-pilot membrane oxidation reactor (MOR)

The membrane oxidation reactor was made of borosilicate glass and stainless-steel components, with a lab-pilot scale capacity of 20 L (Fig. S1). During the photo-Fenton, reactor was operated using two separate light sources: $UVC_{254}$ from inside and $UVA_{365}$ from outside. The lamps used were either 40 W-$UVC_{254}$ lamps protected by quartz glass or 20 W-$UVA_{365}$ lamps without protection. The total power of the lamps was 200 W, and they were positioned symmetrically along the inner and outer cross-sections, respectively. The variations of light intensities vs. lamp numbers used were depicted in Fig. S2. The UF module was submerged inside MOR and had a rectangular, flat-sheet membrane with an effective surface area of 300 $cm^2$ (8.6 cm × 35 cm). It was made of Delrin® (DuPont, USA) and operated using a peristaltic pump to withdraw permeate. Water fluxes were calculated from weight changes by taking the filtrate to a glass beaker on a precision balance. The automation unit monitored and recorded data on air feed, wastewater pH, and temperature. Using benchtop MOR treatment data from Aydiner et al. [4], a lab-pilot reactor was scaled up with a deviation of 11.9 %. The aeration rate, vacuum rate, and light intensity were adjusted based on the benchtop conditions and lab-pilot specifications outlined in Table 2.

The hybrid MOR treatments were performed twice a day with each operation treating 20L of inflow. The UF of the reactor liquor oxidised by AOP was conducted to produce 5L of filtrate in each operation. All AOPs were executed under conditions where influent organics were decomposed to below 100 mg TOC/L and in-reactor total iron was kept constant at 3.0 ± 0.3 g/L. With two consecutive operations of which each achieved this level targeted in the reactor liquid, a 10*L* of UF filtrate was obtained and then it was subjected to the single [$NF_{tight}$] and [RO] processes as well as the dual [$NF_{loose}$ + RO] filtrations. Side-stream membrane concentrates were disposed of by following the approaches outlined in Fig. 1**.** 2L of concentrate produced at 80 % water recovery was mixed with 3L of wastewater per concentrate recycle back to MOR, and experiments were conducted for four consecutive tours by three concentrate recycles (Tables S2 and S3). At the end of the operating day, the fouled membranes were chemically cleaned by means of backwashing with 1 L of 1 % HCl, 1 % NaOH and distilled water at equal intervals for a total of 30 min.

### 2.3.2. Bench and lab-pilot [NF]/[RO] set-ups

Following the MOR treatment of textile wastewater, the bench and lab-pilot scale [NF/RO] filtrations as the side-stream filtrations of MOR effluents were performed individually. The benchtop experiments were used to determine suitable membranes with a dead-end pressurised filtration system (Fig. S3(a)). The set-up consisted of a stainless steel cylindrical stirred cell placed on a magnetic stirrer with a 300 mL feed volume and 14.6 $cm^2$ effective membrane area (Sterlitech® HP4750, USA). The experiments were carried out under the conditions of 26.5 ± 0.5 °C for Fenton-MOR and 40.0 ± 0.5 °C for photo-Fenton MOR, and 12 bar in NF and 40 bar in RO at a constant stirring speed of 300 rpm. After bench studies, the pilot scale experiments of single [$NF_{tight}$] and [RO] and dual [$NF_{loose}$ + RO] were carried out using a cross-flow filtration set-up to recover water from UF effluent and produce concentrates (Fig. S3(b)).

The cross-flow installation had a 100-bar pump, 12L feed volume

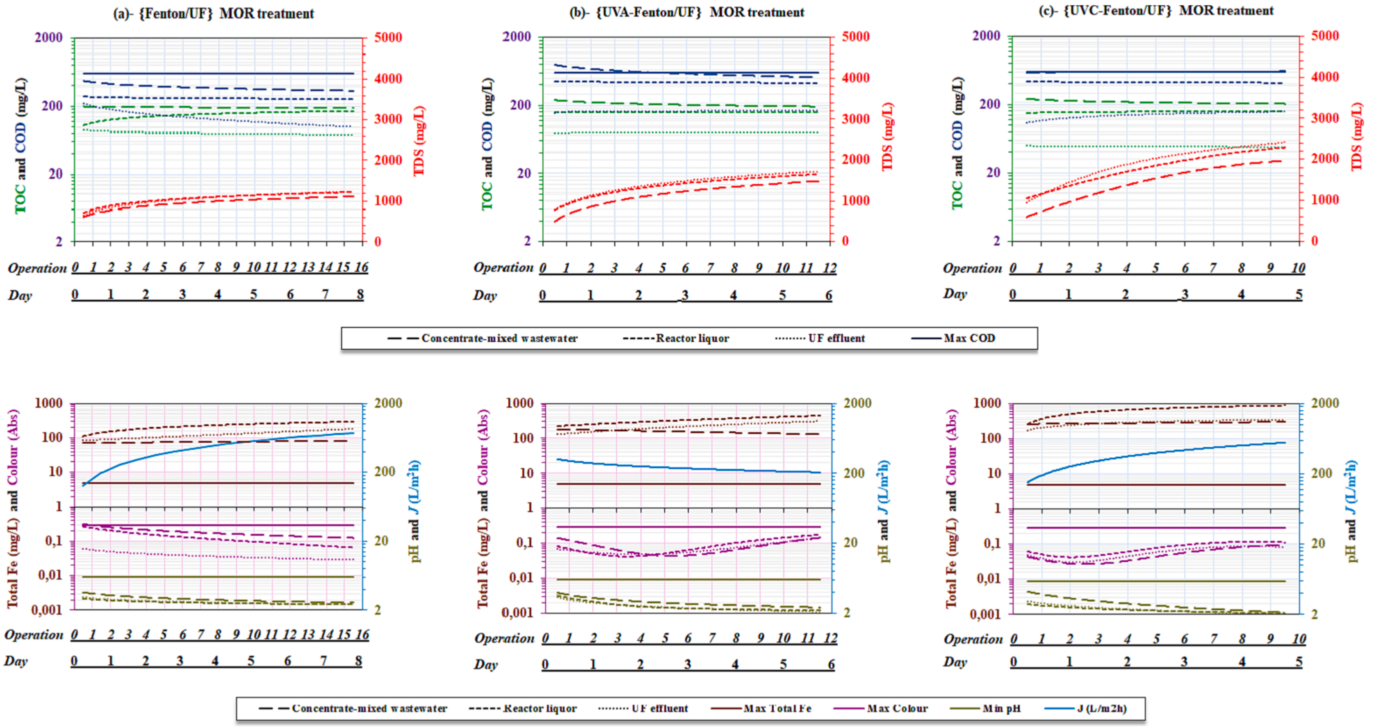


**Fig. 2.** Operational and daily temporal changes of TOC, COD and TDS concentrations and pH, iron, and colour in concentrate-mixed wastewater (– – – –), reactor liquor (- - - - -), and UF effluent (········) in {Fenton/UF} **(a)**, {UVA-Fenton/UF} **(b)**, {UVC-Fenton/UF} MORs **(c)** along with water fluxes of UF membranes in blue and permissible discharge limits shown by solid lines having same assigned colours.

with water cooling jacket and 140 cm$^2$ active surface area within a flat sheet membrane module (SEPA CF II GE Osmonics®, USA). In concentration mode, where the flow was returned to the water bath cooled feed, the cross-flow rate and *trans*-pressure were manually set to 0.93 m/s (10 L/h) and the desired operating values, respectively. The operations were carried out for 10L of UF effluent at the trans-pressures of 12, 20 and 40 bar for [NF$_{loose}$], [NF$_{tight}$], and [RO], respectively, preserving the optimum temperatures of the MOR effluents. As with the benchtop study, the filtrate was collected in a beaker on an electronic balance for weight measurements, and the water flux was calculated with the data transferred to the computer via an RS-232 interface. Details of the analytical procedures, performance calculations and flux – rejection trade-off are given in the Supplementary Material.

## 3. Results

### 3.1. Hybrid MOR performances and discharges

Fig. 2 shows daily treatment performance variations of hybrid MOR. The competency of two-in-one hybridisation of AOP and membrane in remediation of textile wastewater not only possessed removal of persistent organics, but more the abolishment of concentrates under closed-circuit recycling strategy. Although cyclic treatment of concentrates has been thought as a challenge leading to increasing organic and inorganic loads of mixed liquor [27,35], this was not the case of iron and TOC-limited stable MOR operations. Organics disruptions occurred by 57 ± 5–64 ± 6 % Fenton, 61 ± 4–72 ± 5 % UVA-Fenton and 77 ± 3–78 ± 4 % UVC-Fenton MOR with 21 ± 12–26 ± 15 %, 19 ± 5–14 ± 5 % and 11 ± 6–15 ± 6 % UF synergies for TOC and COD, respectively. The same COD but almost 10 % more TOC was removed than the intermittent benchtop treatment of wastewater not mixed with concentrates [4]. This comparison indicated that exposing organics that were degraded to smaller intermediates to oxidation for a prolonged duration more effectively mineralise them into end products. The amount of iron in the permeate was reduced by 42 ± 8 % Fenton, 55 ± 12 % UVC-Fenton (UH050) and 34 ± 6 % UVA-Fenton (UV150) by retaining ferric oxides with UF. In fact, AOP has no an explicit impact on the removal of inorganic iron colour. Due to 97–100 % discolouration of organically sourced colour in each hybrid MOR treatment of textile wastewater optimised by Aydiner et al. [4] (*data not shown*) as also reported in Fenton-integrated UF of textile dyes by Buthiyappan et al. [9], colour was stemming from dissolved and colloidal iron species in aliquot samples filtered by submerged UF. That's why, the removal efficacies of colour signifying the iron-based colour seemed not to be adequate despite the controlled operations of 3.0 ± 0.3 g total Fe/L. The best efficacy was reached by the Fenton-MOR of 78 ± 6 %. Despite better removals of organics than in the Fenton, almost no colour removal was observed in both photo-Fentons operated at higher iron additions, meaning that some species of iron produced much more colour in effluents depending on their higher concentrations. TDS concentrations increased by concentrate recycle and reached steady state, but effective UF removals were not achieved as expected.

Submerged UF was run with the average fluxes of 738.9, 388.2 and 586.3 L/m$^2$h in Fenton, UVA- and UVC-Fenton, respectively; 5 to 10-fold an immersed UF flux of 70 L/m$^2$h for natural waters in China [12], 2 to 4-fold both the pure water flux of 170 L/m$^2$h in TiO$_2$-photocatalysis and UF [34], and 197 L/m$^2$h in UF following ZnO-photocatalytic oxidation of textile wastewater [22]. Though the predominant fouling mechanism has been determined to be dependent on the cake layer formation on membrane surface by Aydiner et al. [4], temporal flux enhancements cannot be explained with enlargements of the membrane pores narrowed and clogged by iron particles and organic aggregates solely after sequentially HCl and NaOH cleaning (Fig. S4). If this were specific to hybrid MOR, either UF membranes should have shown a reduction in purification efficacy [18], or the flux should have become stable or decreased after a certain time [80]. Yet, the effect of pore expansions on the flux decline appeared to be insignificant, even if it happens. The increase in flux against the time indicates the existence of a membrane-specific situation observed just for UH050 polyethersulfone (PES) membrane, not for UV150 polyvinylidene fluoride membrane. Abdi

**Table 3**
Quality analyses results of Fenton or photo-Fenton enhanced MOR effluents and industrial discharge limitations.

| Parameter | Unit | {MOR} effluents | | | Discharge limits | | |
|---|---|---|---|---|---|---|---|
| | | Fenton | UVA-Fenton | UVC-Fenton | KWSA [32][a] | IWSA [28][a] | |
| | | | | | | (1) | (2) |
| pH | – | 2.33 ± 0.27 | 2.19 ± 0.09 | 2.11 ± 0.07 | 6–10 | 6–12 | 6–12 |
| $T$ | °C | 26.5 | 40.0 | 40.0 | 40 | 50 | 50 |
| $E_C$ | $\mu$S/cm | 2593 ± 263 | 3337 ± 265 | 4593 ± 272 | –[b] | | |
| TDS | mg/L | 1321 ± 139 | 1728 ± 171 | 2383 ± 123 | – | | |
| COD | mg/L | 185 ± 17 | 165 ± 37 | 162 ± 24 | 800 | 1000 | 600 |
| TOC | mg/L | 92 ± 14 | 83 ± 13 | 56 ± 16 | – | | |
| SS | mg/L | 24 ± 3 | 32 ± 5 | 45 ± 9 | 350 | 500 | 350 |
| $C_6H_5OH$ | mg/L | 2.9 ± 0.5 | 3.4 ± 0.7 | 4.1 ± 0.8 | 20 | 10 | 10 |
| $SO_4^{2-}$ | mg/L | 603 ± 145 | 368 ± 85 | 387 ± 101 | 1700 | 1700 | 1700 |
| $Cl^-$ | mg/L | <10 | <10 | <10 | – | 15,000 | – |
| Total Fe | mg/L | 161 ± 58 | 275 ± 39 | 379 ± 153 | 5.0 | – | – |
| Total N | mg/L | 7.4 ± 1.6 | 8.3 ± 1.5 | 6.5 ± 1.7 | 100 | – | – |
| $\lambda_{436\text{-}620}$ | Abs | 0.031 ± 0.018 | 0.082 ± 0.047 | 0.080 ± 0.034 | 450[c] | – | – |

[a] in the IWSA Regulation, there are two different discharge limits for industrial effluents to be discharged into the wastewater infrastructure facilities whose sewage systems result in either "*a complete treatment (1)*" or "*pre-treatment + deep sea discharge (2)*". The KWSA regulation applicable to Kocaeli is that which is equivalent or comparable to the IWSA (2) in force of Istanbul Metropolitan Municipality.

[b] not specified.

[c] colour unit defined as Pt-Co for the limit value corresponds to 0.290 absorbance at 456 nm when using a reference colour solution at 450Pt-Co [50].

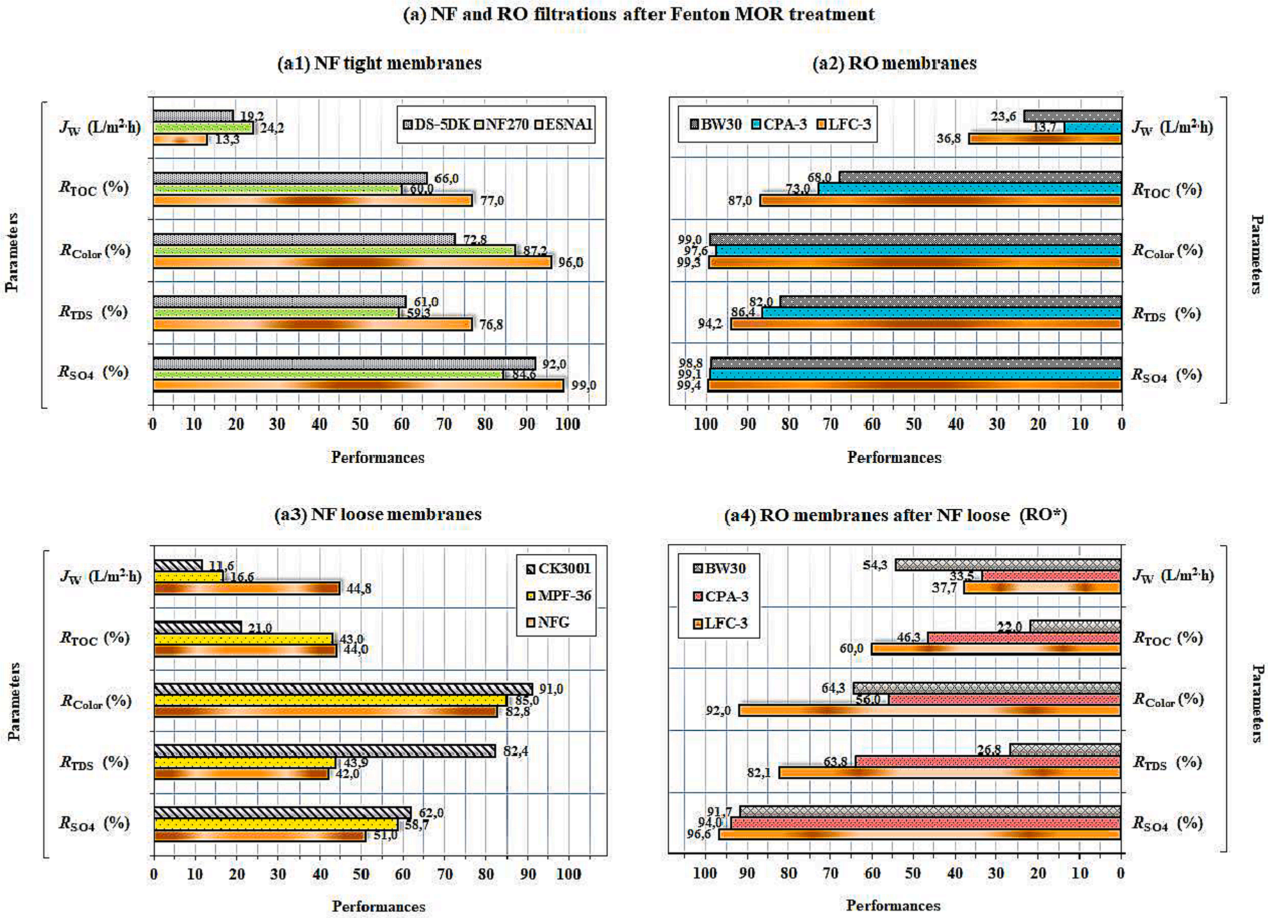
et al. [1] fabricated super-hydrophilic PES–UF membranes with excellent antifouling features by combining hydrous ferric oxide (HFO) particles and UV irradiation. They reported that pure PES membrane flux of 183.40 L/m$^2$h has been increased to 943.23 and 864.62 L/m$^2$h by 5 % HFO particles in the matrix and coating 500 mg HFO/L on the surface of the HFO containing membrane, respectively. UV irradiation of 5 % HFO containing + 0.5 % HFO coated membranes for 10 min showed a high-water flux of 433.18 L/m$^2$h with no decrease in rejection of bovine serum albumin. They also specified that novel membrane produced was capable of rejections of 97.5, 97.3 and 96.6 % for hexane, *iso*-octane, and *para*-xylene, respectively. Accordingly, it can be concluded that the iron oxides produced during oxidation accumulated on the surface and inside the PES-UF membrane, allowed the flux to increase to a stable high value by increasing the membrane hydrophilicity without reducing the organic removal by acting as a secondary selective layer.

The quality adequacy of the [AOP/UF] effluents was evaluated according to the discharge standards established either for the sewage infrastructures of Istanbul, which end with a full treatment, or for the marine receiving environments of the cities of Istanbul and Kocaeli after pre-treatment step (Table 3). Hybrid MOR effluents of have a high degree of compliance for both discharge alternatives, especially for temperature, COD, SS, colour, phenol, $SO_4^{2-}$, $Cl^-$ and total nitrogen. However, prior to discharges, pH should be adjusted to the standard range and the amount of iron should be reduced as a precaution. It is therefore sufficient to neutralise the pH to a reasonable value above 6 with a base such as KOH or NaOH. After pH adjustment, MOR effluent can be discharged directly into municipal infrastructure, which terminates at the conventional wastewater treatment plant in the city of Istanbul, as there is no iron standard for effluent discharge. Before direct discharge to the deep sea, which is not standardised in Istanbul with a limit of 5 mg/L in Kocaeli, an additional process that reduces the high concentrations of 125 and 555 mg Fe/L in MOR effluent would be order of an apple-pie for the sustainability coasts. It should also be noted that the effluents cannot be discharged directly into the receiving environments because of higher organic matter and total suspended solids content than the associated emission levels of the best available technologies according to the EU directive 2010/75 [57].

### 3.2. Determination of suitable [NF/RO] membranes

Suitable NF and RO membranes were found for post Fenton-supported MOR treatment, and the Fenton eligible membranes' performances were also elicited for photo-Fenton assisted applications (Fig. 3). In [NF$_{tight}$] separations of {Fenton/UF} effluents by ESNA1, DS5DK and NF270, ESNA1 membrane gave the lowest flux of 13.3 L/m$^2$h, in contrast to the preferable TOC, colour, TDS, and $SO_4^{2-}$ removals. In [RO] experiments of BW30, CPA3, and LFC3, the highest flux was obtained for LFC3 (36.8 L/m$^2$h), which showed the best quality rejections. With slightly lower colour and $SO_4^{2-}$ but lower TDS rejection, NFG was selected as the best loose NF membrane due to a comparable organic removal at 44.8 L/m$^2$h. In RO filtration of NFG effluents using the same membranes as single [RO], organics, colour and $SO_4^{2-}$ rejections were at the best-in-levels for LFC3 membrane just as for single [RO], but partially acceptable flux of 37.7 L/m$^2$h. The most effective Fenton-based benchtop operation emerged with {MOR} − [RO] allowing to 87.0 % TOC, 99.3 % colour, 94.2 % TDS, 99.4 % $SO_4^{2-}$ and 36.8 L/m$^2$h. For the photo-Fenton (Fig. 3**(b)**), [RO] was one step ahead for water recovery from MOR effluents compared to the [NF$_{tight}$] and [NF$_{loose}$ + RO]. UVA-Fenton supported {MOR} − [RO] appeared as more advantageous than Fenton by 95.3 % TOC, 99.4 % colour, 95.4 % TDS, 99.3 % $SO_4^{2-}$ and 109.4 L/m$^2$h. [NF$_{loose}$ + RO] rejections are also notable close to those of [RO] with 89.6 % TOC, 97.3 % colour, 94.6 % TDS, and 95.3 % $SO_4^{2-}$. In terms of water recovery and contaminant removal, systemic performances showed that the parameters that Fenton and photo-Fentons in hybrid MOR can significantly affect the subsequent [NF/RO] can be related particularly to organics and iron content. Since $H_2O_2$ determines the overall organic efficiency [4] and iron can reduce the organic degradation when increased [26], [NF/RO] membranes can be more effectively combined with hybrid MOR by in-detailed analyses and performance-related tracking of organic and iron species by fine-tuning $H_2O_2$ and iron optimised.

Characteristically, a trade-off between flux rates and rejection capabilities in NF and RO membranes prevails (Fig. 4 **(a) and (b)**). In nanopores, solutes are transported by the diffusion, convection, and electromigration over the selective layer of the membrane and are rejected mainly by size (steric) and charge exclusion (dielectric and Donnan) mechanisms [53]. However, the membrane pore size and surface charge and the presence of gel, cake and polarisation can drastically differentiate permeability and selectivity by also influencing solute transport [77,3]. Apparent water/solute permselectivity ($A/B$) for TOC, colour (dominantly $Fe^{2+}$ and $Fe^{3+}$), TDS, and $SO_4^{2-}$ reduced exponentially by the apparent solute permeance ($B$) especially for the Fenton because of greater penetrations of organics and inorganics into the membrane, *i.e.* lower rejection rates, except only for UVA-Fenton supported tight NF. Both UVC-Fenton NFs and UVA-Fenton loose NF showed similar permselectivity propensities with a unique correlation



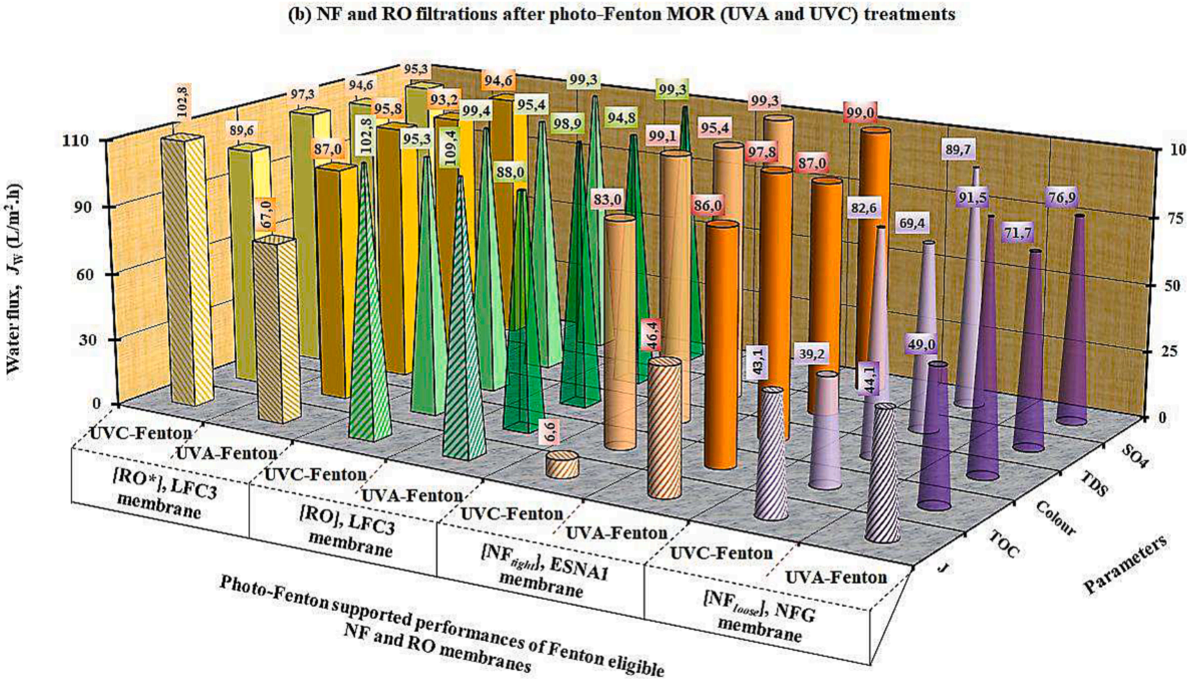
(a) NF and RO filtrations after Fenton MOR treatment

(a1) NF tight membranes

(a2) RO membranes

(a3) NF loose membranes

(a4) RO membranes after NF loose (RO*)



(b) NF and RO filtrations after photo-Fenton MOR (UVA and UVC) treatments

**Fig. 3.** Water flux and rejection performances of different NF and RO membranes ((1): [NF$_{tight}$], (2): [RO], (3): [NF$_{loose}$], and (4): [RO*] ([RO] after [NF$_{loose}$]) after Fenton MOR (a), and comparative NF and RO efficiencies after photo-Fenton MOR (b) by Fenton eligible membranes determined as NFG in [NF$_{loose}$], ESNA1 in [NF$_{tight}$], and LFC3 in both [RO] and [RO*].

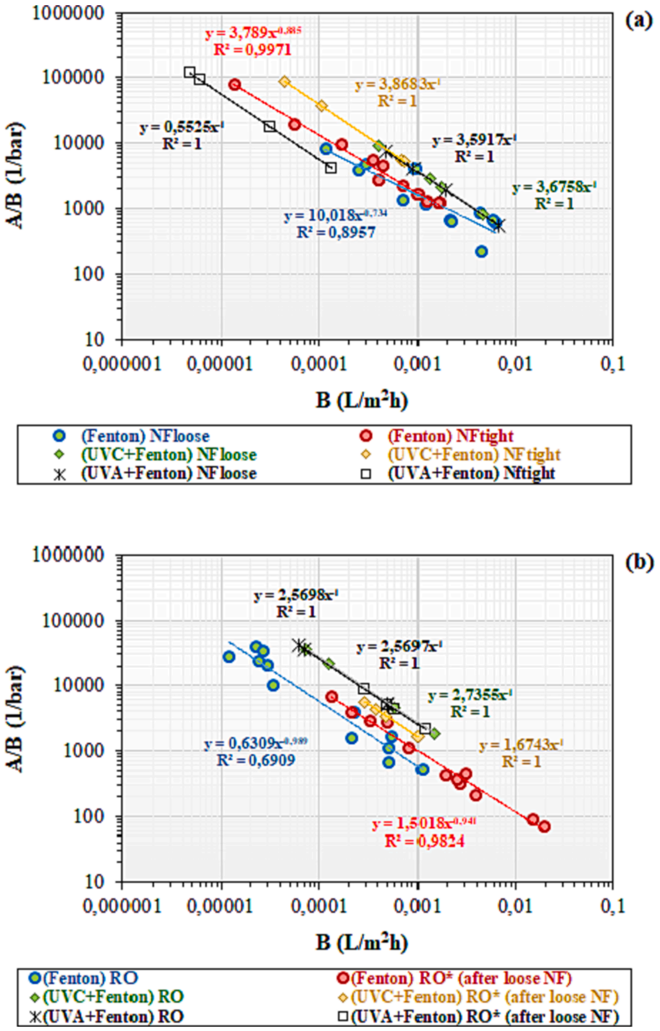


**Fig. 4.** Flux and rejection trade-off observed for TOC, colour, TDS, and $SO_4^{2-}$ in Fenton and photo-Fenton integrated NF/RO systems (a) loose and tight NF membranes, (b) single and loose NF-behind RO membranes.

by cake and concentration polarisation, despite possible diverse pathways of membrane-solute interactions. Such a tendency was also observed for UVA/UVC-Fenton RO and UVA-Fenton RO after loose NF. As the effluent pHs remained about or below 3 in batch experiments (*data not shown*), the isoelectric points of membranes can be assumed to be around zero or at positive [16]. According to tight NFs, loose NFs had comparable removals of iron species, but lower removals of $SO_4^{2-}$ with the largest Stokes radius, indicating lower steric rejections but a greater contribution of the secondary layer to the selectivity. All membranes lead to the formation of a dense iron-containing cake layer by repelling iron ions and oxides even at electrostatic attractions between positively charged membrane and negatively charged solutes [68]. Better organic and inorganic removals point to stronger Donnan and dielectric repulsions of positive-charged solutes in tight NFs than in loose ones to meet electroneutrality [53]. Higher permeate fluxes in the RO membranes behind loose NF according to the single NF and RO membranes did not result in better water/solute permselectivity despite relatively less cake fouling and polarisation. Apparently, a selective fouling layer on the membrane surface significantly improved the removal of co– and counter-ions by the RO membranes when compared by their loose NF subsequent runs. This means that lower RO permselectivity is due to greater electrostatic repulsion of contaminants not only by the membrane but also by the secondary layer which enhances steric repulsion, whereas higher selectivity is more likely to be due to greater charge exclusion by the membrane itself, leading to more severe fouling on the membrane surface [8].

### *3.3. Performance comparisons of {MOR} – [NF/RO] systems*

In order to make water recovery possible and feasible in the devised wastewater management systems, supplementary treatment efficiencies were explored in the single [$NF_{tight}$] and [RO] and combined dual [$NF_{loose}$ + RO] membrane filtrations of effluents from Fenton and photo-Fenton MOR hybrid treatments. All [NF/RO] operations were studied at three concentrate cycles into the influent wastewater to achieve a permanent water flux, signifying sustained efficacy under varying operating conditions. Each specific process, *i.e.*, AOP, submerged UF, hybrid MOR, NF, and RO, and the overall MOR integrated NF/RO treatment systems ({MOR} – [NF/RO]) performed similar-level efficiencies (Fig. 5 **(a), (b), and (c))**. Once making a thorough analysis regarding colour, organic and inorganic matter removal effectiveness was inferred that neither Fenton nor photo-Fenton MORs yielded as high as having desirable for reuse. UF partly improved effluent colour by retaining more ferric particles than ionic or colloidal iron species in the hybrid reactor. Under the operating conditions where the total effluent was at 25 % MOR volume, UF could be exploited with high water fluxes changing between 96.4 and 820 L/m$^2$h, being especially greater in the Fenton and UVC-Fenton MORs, most probably due to the secondary layer containing iron oxides which made the membrane more hydrophilic [1]. It can be undertaken that membrane filtration is one of the most prominent methods to remove COD, colour, and salinity from wastewaters [59]. The integration of NF and RO membranes behind hybrid MOR resulted in quite high performances exceeding 90 % even reaching 100 % in some of quality parameters.

At 80 % water recovery, side-stream NF and RO processes was operated with the water fluxes changing between 23.6–34.7 in [$NF_{tight}$], 20.0–69.9 L/m$^2$h in [RO], and 20.1–80.0 L/m$^2$h in [RO] after [$NF_{loose}$]. Total performances close to these levels but lower were reported by Lebron et al. [33] and Moreira et al. [43] for textile industry wastewater treated with the serial MF, AOP and NF integrated systems. Lebron et al. [33] employed NF after MF + AOP treatment with 19 L/m$^2$h at 12 bar by 10 % recovery, compared to this work at the same pressure but 8-fold high recovery. Moreira et al. [43] also reported 13 L/m$^2$h water flux of at 60 % recovery for the same system. Wang et al. [72] found 59.4 L/m$^2$h RO flux after UV/$O_3$ oxidation at 60 bar and 60 % recovery, as higher than 34.9 L/m$^2$h after Fenton in pilot-scale advanced treatments of effluents treated with coagulation, sedimentation, and aerobic-anaerobic. Flux benchmark deduced that {MOR} – [NF/RO] systems are operable effectively by substantial fluxes even at a high-water recovery rate of 80 % to be made water recovery more economical, compared to the oxidation-included similar treatment systems suffering from the membrane fouling and flux decline [13]. The most significant factor contributing to higher water fluxes and rejection rates compared to literature studies is based on the fact that the purification was carried out using a hybrid reactor specifically designed for the high-performance purpose, for which two patent applications have been filed [4]. None of the studies in the literature except our publication on paper wastewater treatment constantly implemented (photo)Fenton hybrid MOR to mixture from the outset, with concentrate and raw wastewater being mixed, as was the applied case here [19]. Moreover, integration of hybrid oxidation treatments under optimal conditions of MOR pre-treatment step with various pressurised membrane has been operational determinant of systematic performances, accompanied by the use of the most suitable membranes for water recovery and contaminant retention in the pre-concentration step [NF/RO]. On the other hand, although a cleaning process for the NF and RO membranes and the possible strategies for potential fouling problems were not considered in this study, the long-term adverse effects of membrane fouling can be reduced and their safe operation ensured by elaborately acidic, basic, and Fenton cleaning procedures [45,73,36].

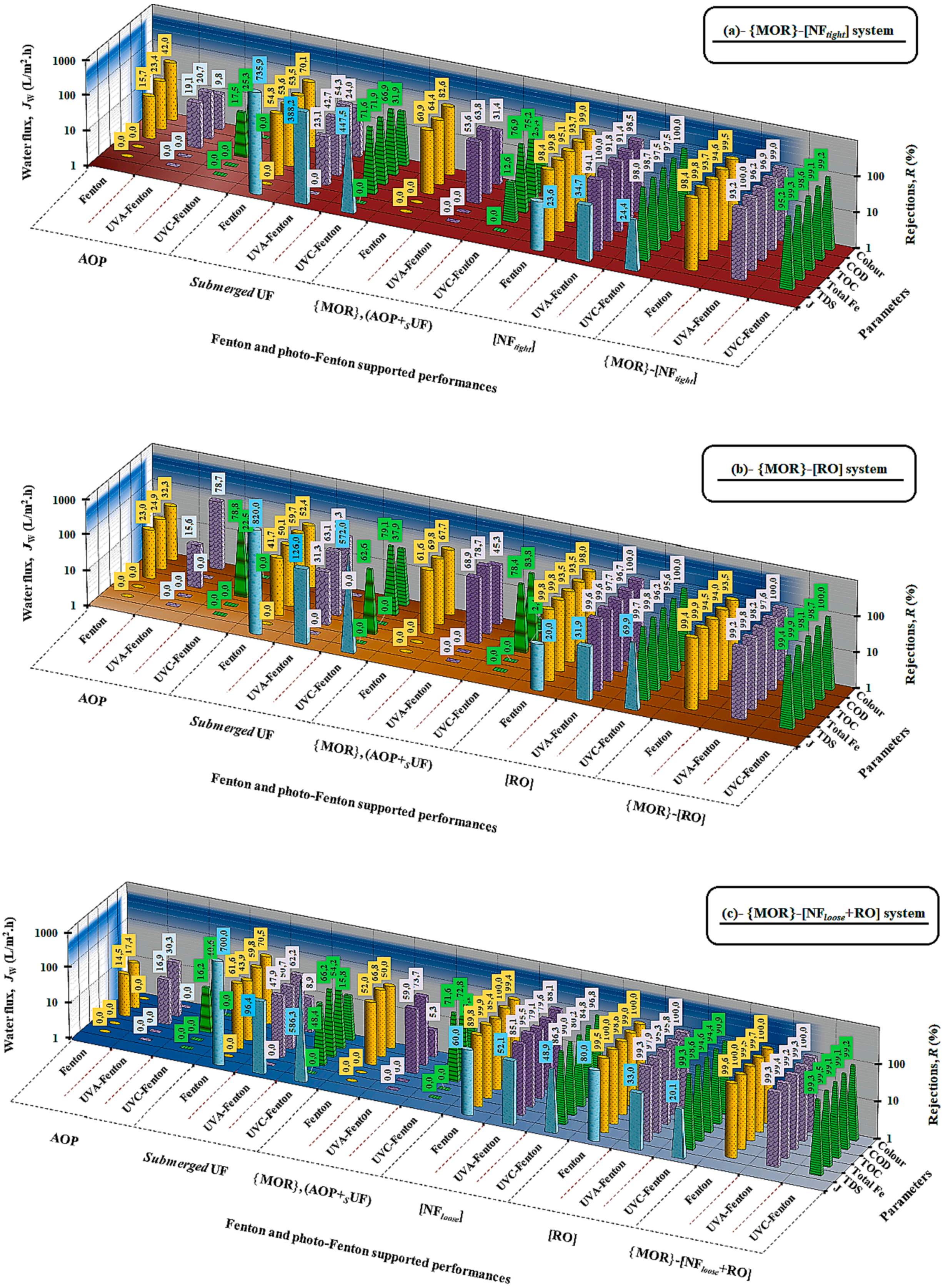


**Fig. 5.** Technical performances of individual and overall in the Fenton and UVA/UVC-Fenton MOR integrated NF/RO systems ((a) {MOR}-[NF$_{tight}$], (b) {MOR}-[RO], and (c) {MOR}-[NF$_{loose}$ + RO]) (Fenton, UVA-Fenton and UVC-Fenton supported removal of colour, COD, TOC, iron and TDS are characterised by yellow cylinder, purple quadrangular and green pyramid, respectively; but water flux is coloured blue for all). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

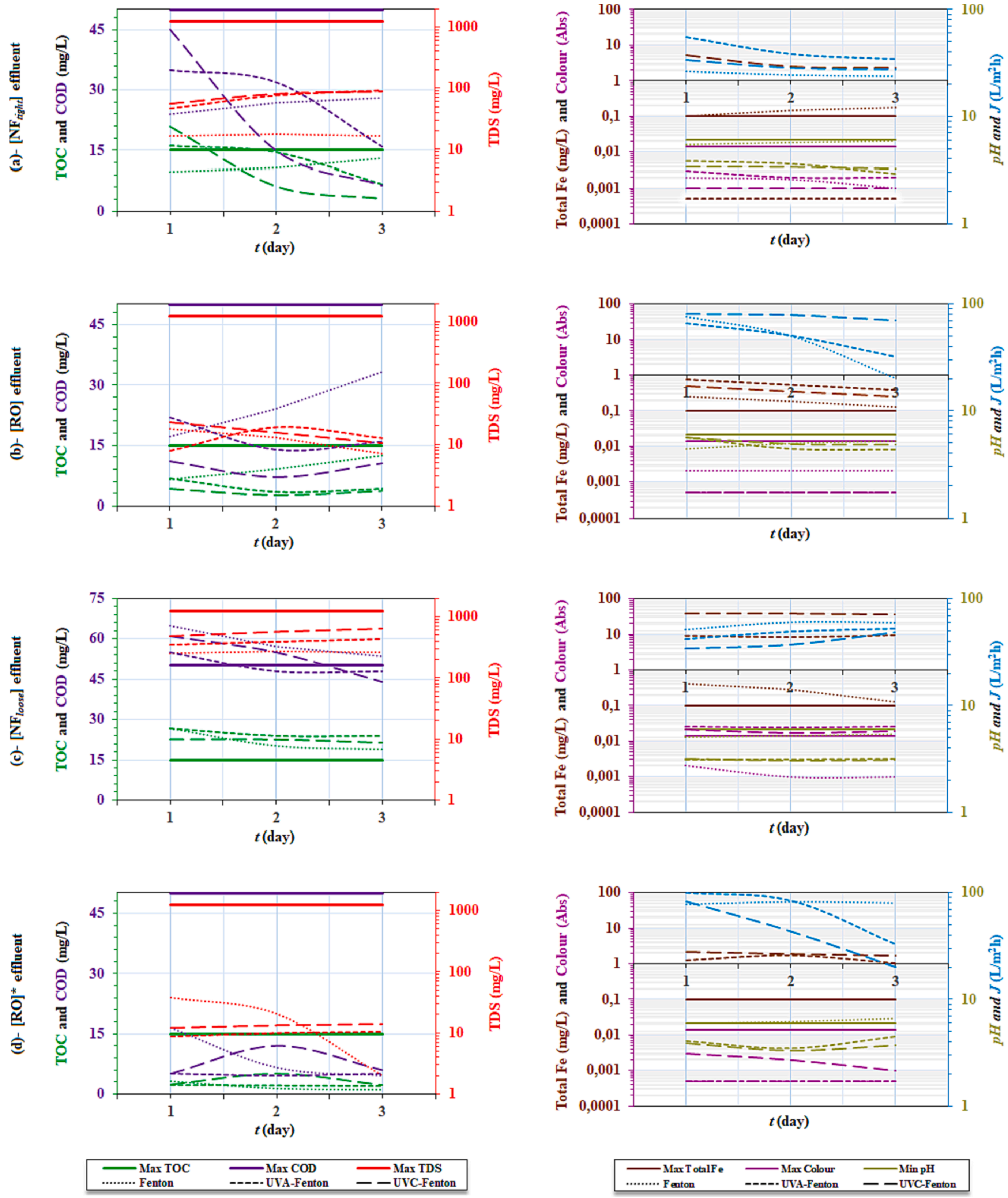


**Fig. 6.** Daily temporal changes of TOC, COD and TDS concentrations, NF and RO water fluxes, pH, iron, and colour in reuse waters produced by NF and RO filtrations of UF effluents from Fenton (⋯⋯), UVA-Fenton (- - - - -) and UVC-Fenton (- - - -) supported MOR (**(a)** [NF$_{tight}$], **(b)** [RO], **(c)** [NF$_{loose}$], and **(d)** [RO*] ([RO] after [NF$_{loose}$]), and high-quality reuse water limits are shown with solid lines having same assigned colours).

**Table 4**
Qualifications of reuse waters produced by integrated {MOR} − [NF/RO] systems.

| Parameter | Unit | Integrated {MOR} − [NF/RO] systems | | | | | | | | | Reuse water quality | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | {MOR}-[NF$_{tight}$] | | | {MOR}-[RO] | | | {MOR}-[NF$_{loose}$ + RO] | | | | |
| | | Fenton | UVA-Fenton | UVC-Fenton | Fenton | UVA-Fenton | UVC-Fenton | Fenton | UVA-Fenton | UVC-Fenton | High$^{a,b,c}$ | Moderate$^{b}$ |
| pH | – | 5.87 | 2.92 | 3.28 | 5.17 | 4.37 | 4.79 | 6.57 | 4.44 | 3.74 | 6.0–8.0$^{a,c}$<br>6.5–7.5$^{b}$ | 7.0–8.0 |
| $T$ | °C | 26.5 | 40.0 | 40.0 | 26.5 | 40.0 | 40.0 | 26.5 | 40.0 | 40.0 | –$^{d}$ | – |
| $E_C$ | μS/cm | 38 | 179 | 185 | 15.0 | 27.0 | 22.0 | 10.0 | 21.1 | 26.6 | <2500$^{c}$ | – |
| TDS | mg/L | 17 | 91 | 88 | 7.1 | 12.7 | 10.3 | 5.0 | 10.6 | 14 | <1250$^{c}$ | – |
| COD | mg/L | 28.0 | 16.0 | <10 | 33.3 | 16.0 | 10.5 | <10 | <10 | <10 | 20 − 50$^{b}$<br><25$^{c}$ | <200 |
| TOC | mg/L | 13.0 | 6.7 | 3.1 | 12.5 | 4.2 | 3.6 | 1.1 | 1.9 | 2.2 | ~7 − 18$^{e}$ | – |
| Turbidity | NTU | 0.39 | 0.22 | 0.29 | 0.51 | 0.22 | 0.34 | 0.27 | 0.44 | 0 | <1.0$^{c}$ | – |
| SS | mg/L | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | – |
| $NH_4^+$ | mg/L | 0.32 | 0.17 | 0.25 | 0.50 | 0.52 | 0.36 | 0.57 | 0.69 | 0.60 | – | – |
| $\lambda_{436\text{-}620}$ | Abs | 0.001 | 0.002 | 0.001 | 0.002 | 0 | 0 | 0 | 0 | 0.001 | *Non-visible$^{a,b}$<br>0$^{c}$ | *Non-visible |
| $NO_3^-$ | mg/L | 1.56 | 1.95 | 1.39 | 0.30 | 0.40 | 0.30 | 0.20 | 0.40 | 0.38 | – | – |
| $SO_4^{2-}$ | mg/L | 6.2 | 0 | 0 | 40.3 | 40.9 | 31.8 | 0 | 0 | 0 | – | – |
| $Cl^-$ | mg/L | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <500$^{b}$ | 500–2000 |
| Total Fe | mg/L | 0.18 | 0 | 2.3 | 0.13 | 0.38 | 0.25 | 0.1 | 1.01 | 1.66 | <0.1$^{a,b}$ | <0.1 |
| Total N | mg/L | 1.88 | 2.12 | 1.64 | 0.80 | 0.92 | 0.66 | 0.77 | 1.09 | 0.98 | – | – |
| Total Hardness | mg $CaCO_3$/L | 11.89 | 33.60 | 11.89 | 11.90 | 11.80 | 0.56 | 0 | 2.09 | 2.50 | <60 − 80$^{a}$<br><100$^{b}$<br><10$^{c}$ | <100 |
| $C_6H_5OH$ | mg/L | 0.01 | 0.01 | 0.01 | 0 | 0.01 | 0 | 0.02 | 0.06 | 0.03 | – | – |

[a,b,c] are the qualifications specified by [a] Confederation of British Wool Textiles [67] and for [b] high quality recycled effluent at textile finishing processes and moderate quality washing-off water [61,67], and [c] high quality direct reuse water [7].
[d] not specified.
[e] a limit value determined considering together the maximum limit value of 50 mg COD/L and the TOC/COD ratio of the raw textile wastewater.
*is the suggested specification for water with no visible colour absorbance as 0.02–0.04 at 450 nm, 0.02–0.05 at 500 nm, 0.01–0.03 at 550 nm and 0.01–0.02 at 600 nm that can be attributed to a reference $\lambda_{450\text{-}600}$ value of 0.014–0.033 as the geometric average of the specified values [67].

### 3.4. *Qualifications of reuse waters*

Together with reuse concentrations, the daily parametric changes in NF and RO effluents during overall treatments via the MOR integrated systems were presented in Fig. 6 separately for the [NF$_{tight}$], [RO], [NF$_{loose}$], and [RO] after [NF$_{loose}$]. The quality of reuse waters produced by {MOR} − [NF/RO] systems was also given in Table 4, with high and moderate quality waters reusable at levels corresponding to 10–20 % and 50–70 % of total water demand [61,7,67]. All long-term operations resulted in a significant improvement in final effluent quality because TOC, COD, and TDS levels in the permeate were reduced to below maximum allowable reuse limits. Even if the MOR effluent appears to be recoverable with slightly less than 50 mg COD/L by loose NF, moderate-quality recovery might be possible using [NF$_{loose}$] effluents due to high TOC of 20–30 mg/L. Despite meeting the limit colour at all final effluents, the iron content could not be completely reduced below 0.1 mg Fe/L, so further handling would be required. For this, IE or adsorption can be exploited as one of the leading techniques with 95–99 % high efficiency [30]. Yet, recent studies have also clearly demonstrated the technological competency of IE on the iron recovery from Fenton effluents [40,54].

Despite pH increases in some final effluents, target pHs could not be achieved except only for {UVA-Fenton/UF} − [NF$_{loose}$ + RO]. Single loose NF after MOR seemed lacking the compatible iron and colour. To adjust volume of high-quality water up to 20 % in RO, [NF$_{loose}$] can be combined for more polishing either with IE of iron or adsorption of iron and organics whereby 70 % moderate-quality water can also be provided by fit-to-purpose recovery instead of 100 % high-quality water supply in the final-step RO. Since it provided higher flux than tight NF and offered greater performance when combined with RO. It can be thus expressed that utilisation of the loose NF before RO would improve field-scale practices reducing the performance losses in RO membrane. {MOR} − [NF/RO] could undoubtedly harvest good-quality waters excluding pH and iron, and satisfactory operations are fulfilled using [NF$_{loose}$] and [RO]. It was resolved that developed eco-innovative systems reclaim concentrates and not produce residual sludge wastes, make it technically possible to produce moderate- and high-qualified waters for reuses.

## 4. Discussion

### 4.1. *Major findings and implications*

In prevalent apps for recoveries and reuse of industrial wastewater, advanced treatment techniques are exploited mostly after biological or chemical treatments [29]. Oxidation and membrane systems have already begun to be researched for the water recovery in line with eco-friendly disposals of concentrates as a crucial barrier of industrial sustainability [33,43]. Thus, here first, textile wastewater was treated through a hybridised operation of Fenton AOPs and UF to discharge legitimately with concentrate treatment and zero sludge discharge. Later, MOR effluents were more purified with [NF/RO] sequences for various reuse options. As notified by Moradihamedani [42], UF cannot respond to the absolute dye removals from waters and is typically usually appointed for pre-treatment of industrial wastewaters depending on its relative larger pore sizes. As applied here, assembling UF with the Fenton-situated AOPs conduced an assured utility up to 40 % synergistic effect to satisfactorily removals of dyes, large molecular organic matters, and even degradation intermediates, but not of colour associated to ferrous and ferric ions depending on the Fenton-specific circumstances. It also contributed to 10 % higher mineralisation of organics by retaining longer them within the reactor. Hybrid MORs showed substantial potential in treating textile wastewater with membrane concentrates. By returning concentrate to MOR and treatment here repeatedly, a good treatability with no concentrate and iron sludge out succeeded without any increment in reactor organic content at the fixed iron level.

**Table 5**
Comparative performance of water and material recovery from textile wastewater by liquid discharge application approaches.

| References | Wastewater | Operation scale | Technology application step in liquid discharge approaches | | | | | | Resource recovery | | Liquid discharge solution | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | I. Pre-treatment | | II. Pre-concentration | | III. Dewatering | IV. Precipitation | | | | | |
| | | | technology | efficiency | technology | efficiency | technology | technology | Water | Material | MLD | NZLD | ZLD |
| Vergili et al. [70] | Textile dye bath wastewater | Lab | UF, loose NF | 14.8–64.6 % COD 99.3–99.9 % colour | tight NF, RO | 99.6 % COD 100 % colour | MD | Incineration | 95–96 % | NaCl<br>$Na_2CO_3$ | | | ✓ |
| Sahinkaya et al. [58] | RO concentrate of biologically treated textile wastewater | Lab | UF integrated pellet reactor | 93 % total hardness | RO | 97.8 COD 90 % colour | – | – | 92–94 % | – | | ✓ | |
| Çelebi et al. [17] | RO retentate from conventional and advanced treated textile wastewater | Lab | Coagulation + Precipitation + MF | 45 % $Ca^{2+}$ 25 % $Mg^{2+}$ 30 % $SiO_2$ 87 % TOC | NF | 97 % $Ca^{2+}$ 83 % $Mg^{2+}$ 92 % $SiO_2$ 87 % TOC | – | – | 80 % | – | | ✓ | |
| Moreira et al. [43] | Textile wastewater | Lab | MF + NF | 92.2 % COD > 98.5 % colour | UV/$H_2O_2$, Fenton and photo-Fenton | <0.001 g/L indigo blue | – | – | 60–80 % | >99 % indigo blue | ✓ | ✓ | |
| Partal et al. [47] | RO retentate from conventional and advanced treated textile wastewater | Pilot | $O_3$ + NF + NF | 14 ± 7Pt-Co colour 85 ± 3.8 mg/L COD 4.6 ± 0.9 g/L TDS | RO + IE | <1Pt-Co colour < 10 mg/L COD 0.1 g/L TDS | – | – | 77 % | 66 % NaCl | ✓ | | |
| Wang et al. [72] | high-saline textile wastewater | Pilot | UV + $O_3$ | 85 % colour 43.2 % COD | RO | 52 ± 15 mg/L TDS | Evaluation | | 70 % | – | ✓ | | |
| This study | Textile washing wastewater | Lab-pilot | Fenton or photo-Fenton + UF | 66 ± 12 % TOC 70 ± 12 % COD | NF, RO, NF + RO; all with IE | 98.6 ± 0.5 % TOC 98.9 ± 0.2 % COD 99.6 ± 0.4 % colour 99.6 ± 0.3 % TDS | Optional | – | 50–99.9 % | $Fe^{2+}$ $Fe^{3+}$<br>$H_2SO_4$<br>$PO_4^{3-}$ | ✓ | ✓ | ✓ |

NF and RO are among the best available technologies for textile wastewaters [42]. Their integrations to the MOR-treated effluents accommodated able to polish as industrial reuse water in all three systems with high purification capacities viz., {MOR} – [$NF_{tight}$], –[RO], and – [$NF_{loose}$ + RO]. In order to highlight the value that this study adds to the literature, the novelties and specific advantages it offers have been comparatively presented in Table 5 against existing alternative treatment solutions. The concentrate-looped {MOR} – [NF/RO] applications have an excellent and exclusive competence for the elimination of dyes and derivative chemicals and inorganic reagents from textile wastewater. High-quality waters were successfully produced with the non-oxidation membrane systems which were operated with just-at-once reclamation or minimisation of concentrates [70,58,17]. However, only moderate, or low-quality waters could be produced by using the oxidation – membrane systems [43,73] except double NF included RO + IE system producing high-quality water [47]. Here, the systems treating concentrates cyclically in hybrid reactor accomplished recovery at good-quality necessitate inevitable adjustments of the lacks pH and iron by the Fenton's nature [43]. Integrity of pH neutralisation and Fe elimination affairs for field feasibilities would be thus so crucial that otherwise neither high-quality recovery nor discharges at desirable qualities would be provided. Contrary to the literature on many industrial wastewaters, the major drawbacks of NZLD and ZLD solutions, which are frequently practised in 3 or 4 steps [23,69,15,38,79,2,37], was avoided in this work by reducing to the just two application steps, pre-treatment and pre-concentration: limiting factors such as high energy requirement, high cost, more complex processing need and possible lack of stable operability.

On framework envisioned, the main advantages of systems applied, as freely of three Fenton varieties, can be summarised as quite great removal efficiencies of dyes and chemicals, too high UF water fluxes, in-situ disposal of membrane concentrates, direct wastewater treatment without a preliminary process, no generation of waste sludge, and readily practicability in centralised or decentralised industrial zones. The main challenges are compiled as more reclamation before discharge (pH control and iron reduce), chemical spends (oxidant, catalyst, acid, and base), and extra energy need when UV lighting is applied. Finally, gathering the findings and determinations pros and cons so far, eco-innovative solutions from minimal to zero liquid discharge could be uncovered obviously as in the following subsection.

### 4.2. *Eco-sustainable managements from MLD to ZLD*

Liquid discharges are included the recovery of water from 50 to 80 % (minimal), 80 to 95 % (near-zero) and over 95 % (zero) with resource recoveries and lower energy, if possible [52]. Many physical, biological, chemical and thermal technologies are used in MLD to ZLD systems, which consist of 2 (pre-treatment and pre-concentration) to 4 stages followed by dewatering and precipitation. MLD and NZLD are less costly and easier to operate with higher technology readiness levels than ZLD thanks to less complex processes and operations. All are compiled to recover water in semi- to fully-closed loops and reduce and/or recover concentrates in or out of the system devised by a CE model [46,10,48]. In systems here developed for all three approaches, the treatment of textile wastewater and the recovery of good-quality water were ensured only in the first two steps with the in-system disposal of concentrates. Accordingly, holistic greener and circular managements of textile wastewater have been suggested for the ZLD and MLD – NZLD applications in Figs. 7 and 8, respectively, based on performances experienced and recent technological innovations. The field-scale efficacies of systems for 1000 $m^3$/d capacity were simulated by using a process modelling and simulation software (Intelligen's SuperPro Designer® v9.0) in which IE design data were first captured using *Purolite* software (EcoLab Co.) and then adapted into the IE-integrated *SuperPro* designs.

In ZLD management, the effluent is fed in single or dual combined [NF/RO], once inflow from mixing wastewater and concentrate is

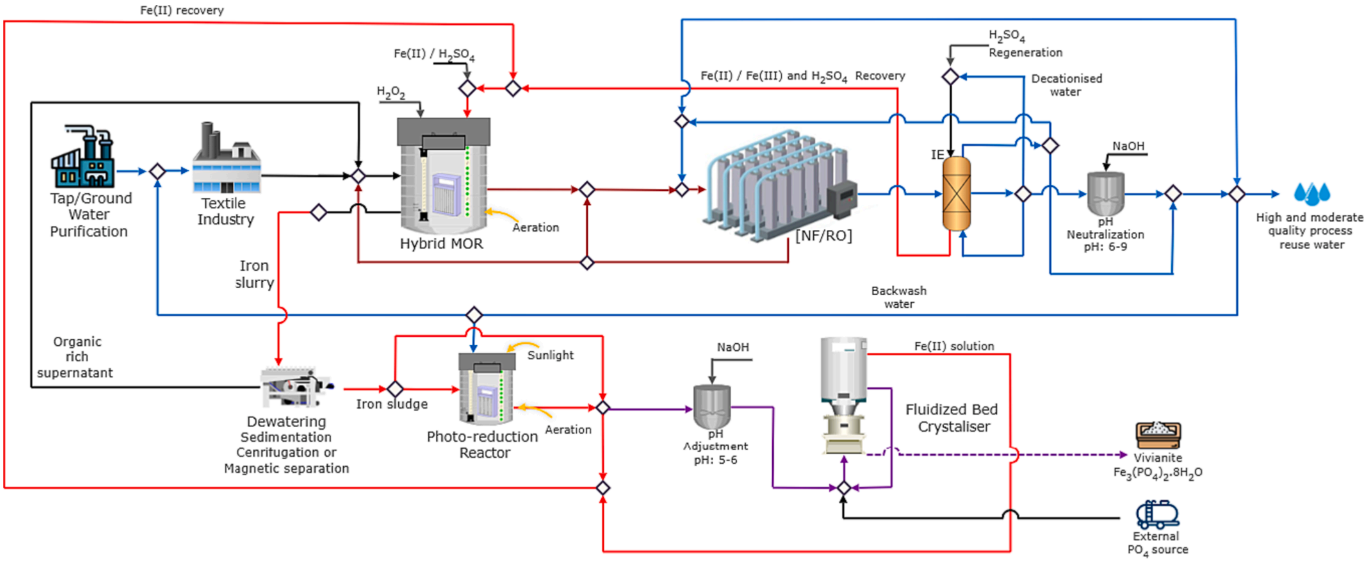


**Fig. 7.** Eco-innovative ZLD reclamation system of textile wastewater developed through in-situ membrane concentrate (brown) and IE regeneration water recycling (red) depending on hybrid MOR treatment (brown), [NF/RO] and [IE] effluents and reuse water production (blue), consumable ($FeSO_4$ and $H_2SO_4$) recovery and reuse (red); with in-situ chemically recovery of vivianite using external $PO_4^{3-}$ source (purple) proposed by Priambodo et al. [51].

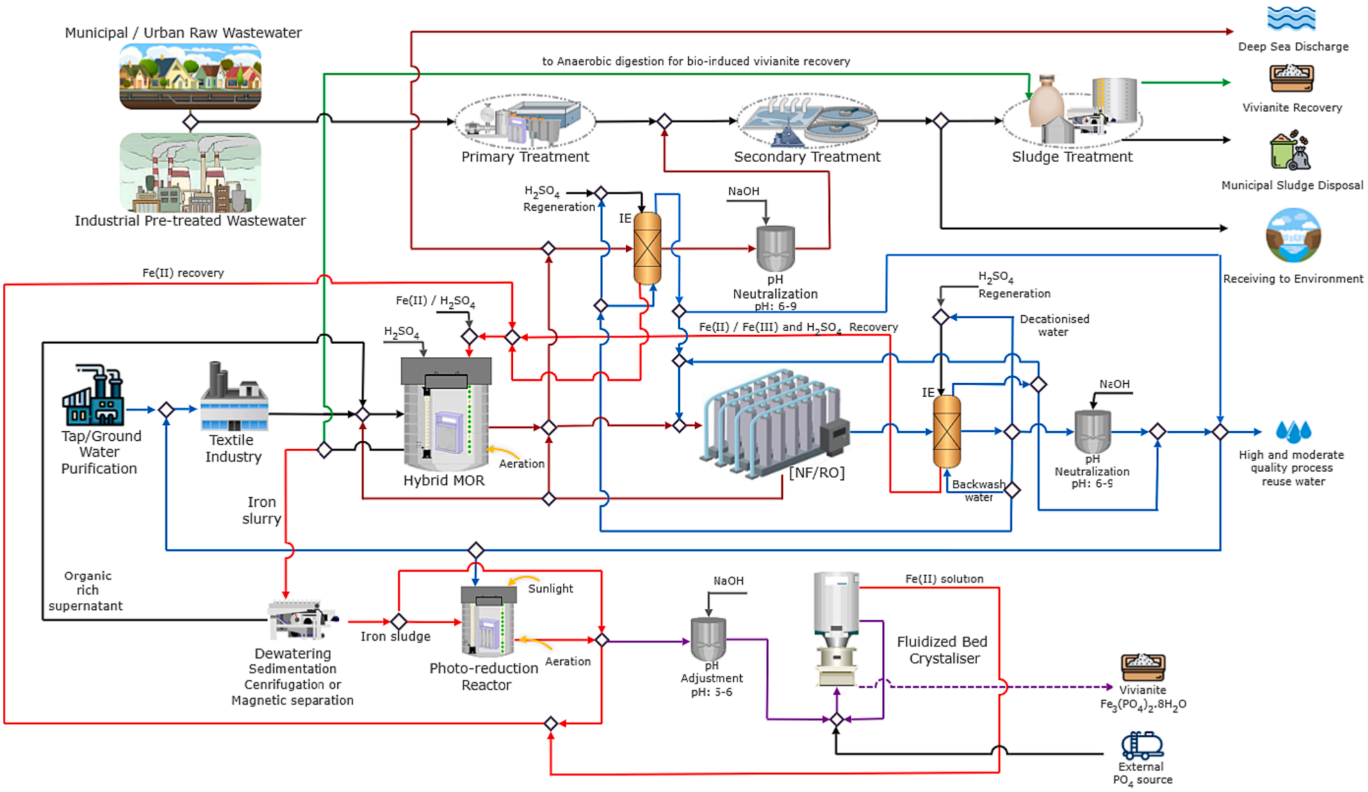


**Fig. 8.** Eco-innovative MLD/NZLD reclamation system of textile wastewater developed through in-situ membrane concentrate (brown) and [IE] regeneration water recycling (red) based on hybrid MOR treatment and discharging (brown), [NF/RO] and decationised [IE] effluents and reuse water production (blue), consumable ($FeSO_4 - H_2SO_4$) recovery and reuse (red); with in-situ chemically recovery of vivianite using external $PO_4^{3-}$ source (purple) proposed by Priambodo et al. [51] and ex-situ bio-induced recovery of vivianite via mixing with anaerobic digestion liquor (green) proposed by Wilfert et al. [75] and further developed by Wijdeveld et al. [74].

treated by MOR. After membrane operation, IE is substituted to decrease iron and backwash and regeneration waters are supplied from its operation. Backwash water, if clean, is mixed with reuse water [31], otherwise it is returned to [NF/RO]. $H_2SO_4$ and Fe-containing regeneration water is directed to meet Fenton's acid and iron needs. The pH neutralisation of final effluents after the membrane effluent was subjected to IE is completed to produce reuse waters. Meanwhile, acid and base can be utilised to clean the UF, NF and RO membranes, the resulting streams can be reused in the system to ensure no waste production, so iron and consumables from the cleaning could also be recovered. By this means, while the used acid and base streams in the MLD and NZLD can be directed to the MOR and discharge channel, respectively, both sent to the MOR in the ZLD. The ZLD strategy is practicable with 99.9 % reuse water and 98–100 % Fe − $H_2SO_4$ saving compared to the sole MOR treatment, but 48–51 % more $H_2O_2$. For full iron recovery, 0.4–0.7 % slurry in MOR can be used to produce $Fe^{2+}$ liquor by photo-reduction [71], with recovering vivianite using external $PO_4^{3-}$ in a fluidised bed crystalliser after pH adjustment to 5–6 [51]. In the case of high organic content, dewatering such as settling, centrifugation or magnetic separation is implemented to generate sludge of which in-reactor feed (0.5–5 $m^3$) reduces duration to a few hours [21]; and loading supernatant (0.2–2 $m^3$) to MOR ends zero sludge cycle [39].

In MLD and NZLD managements, all operations are the same as in ZLD with a main difference that MOR effluent is separated into two streams; one of which is for water and resource recoveries and the other is discharged. Before discharging, IE and pH adjustment is performed to ensure the limit values. Both strategies are also feasible with iron minimisation over 98 % but higher $H_2O_2$ demand increasing from 20 − 37 % in 50–80 % MLD to 37–48 % in 80–95 % NZLD than those in the single MOR treatments. IE-treated effluents can be transferred to biological process of centralised municipal treatment to safely discharge or released directly to the sea by deep discharge. Other choice, not requiring pH − Fe control and decreasing costs, is to discharge effluents to anaerobic digestion of centralised treatment to ex-situ bio-induced recover vivianite at 70–90 % even > 90 % P [75,74]. From an overall financial viewpoint, the implementation of the eco-innovative systems developed from MLD to ZLD will entail an extensive and detailed research, including cost-benefit analyses taking into account both operating and capital expenditures and environmental impacts in all aspects. Considering an overarching perspective specific to the city of Istanbul, the current industrial water tariff is USD 2.4 per $m^3$; 1.6 is the cost of using mains water and 0.8 is the cost of discharging into the city sewage canal. When operating costs meeting the discharge standards, *e.g.* 0.6 USD/$m^3$ for coagulation and advanced oxidation after biological treatment [62] and 1.0 USD/$m^3$ for biological treatment and NF (does not provide the target process water qualities in this study) [55], are taken into consideration, unit water cost for textile industry would be on average 3.2 ± 0.2 USD/$m^3$. Accordingly, it can be obviously stated accompanied by serious environmental benefits provided with recovering resources and reducing or eliminating liquid discharge that the techno-economic applicable eco-innovative solutions would serve to sustainable development goals below this value of water tariff for textile plants in centralised and decentralised regions not only in Türkiye but also all over the world.

## 5. Conclusions

In this study, eco-innovative systems, which capable of purifying textile wastewater and recover water, reducing or eliminating consumables, waste sludge and concentrate, and if possible, providing valuable waste recovery, were explored whether can be developed as to liquid discharges from minimal to zero. After long-term reclamations by (photo) Fenton-included UF of wastewater shuffled with concentrates, good-quality waters just needing to control pH and Fe were obtained by making to produce reuse water and not to discharge effluent, and sludge and concentrate possible using NF, RO, or NF + RO filtrations. Real-world simulations of {MOR} − [NF/RO] − [IE] systems designated that 50 %−MLD to 99.9 %−ZLD can be realised significant savings of 98 % catalyst and 100 % acid but increasing of 20–51 % oxidant. It was also established, if desired, MOR effluent can be either released to the sea or a central treatment plant after Fe recovery and pH adjustment or be directly exploited as a fundamental source of valuable $Fe_3(PO_4)_2{\cdot}8H_2O$ recovery from the plant's anaerobic digestion liquors. The pH neutralisation-included {MOR} − [NF/RO] − [IE] systems developed by MLD-NZLD and ZLD approaches are practicable with less energy due to two-step realisation of pre-treatment (MOR) and pre-concentration (NF/RO and IE). So, they will be able to serve green industrial development by raising water − resource − energy savings in real onsite conditions, especially for ZLD. Further study on holistic viabilities of these kind systems is recommended to reveal the field performances in environment, energy, and economy viewpoints not only for textile wastewaters but further other industrial wastewaters such as paper, food, meat, etc.

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## CRediT authorship contribution statement

**Coskun Aydiner:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Esra Can Dogan:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Berna Kiril Mert:** Data curation, Investigation, Methodology, Writing – original draft. **Burcu Pala:** Data curation, Resources, Software, Validation, Visualization, Writing – original draft. **Tugba Nur Demirozlu:** Data curation, Investigation, Methodology, Validation, Visualization, Formal analysis. **Esin Balci:** Data curation, Investigation, Methodology, Validation, Visualization, Formal analysis. **Sevgi Topcu Yakin:** Data curation, Investigation, Methodology, Validation, Visualization, Formal analysis. **Cemre Tongel:** Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization. **Ali Oguzhan Narci:** Data curation, Investigation, Methodology, Validation, Visualization, Formal analysis.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Prof.Dr. Coskun Aydiner reports financial support was provided by The Scientific and Technological Research Council of Türkiye (TUBITAK). If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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## Appendix A. Supplementary data

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