

Research article

Treatment of natural dye wastewaters by electrochemical oxidation: Comparative analysis of anode materials and optimization for reuse

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ABSTRACT

This study evaluates the feasibility of electrochemical oxidation for treating real wastewater from the eco-dyeing of linen fabric with a natural dye extracted from *Punica granatum* (pomegranate peel), aiming to enable its reuse in new dyeing processes. The untreated wastewater exhibited a yellowish colour, chemical oxygen demand (COD) of 500 mg L⁻¹, low biodegradability (0.24), and moderate acute ecotoxicity toward *Daphnia magna* (2.3 toxic units), confirming the need for advanced treatment. Comparative electrooxidation experiments were conducted using boron-doped diamond (BDD) and different titanium-based mixed metal oxide anodes. The BDD anode exhibited the highest pollutant removal efficiency, achieving over 90 % COD reduction and significant mineralization. The optimal operating conditions were identified at 50 A m⁻² and an applied charge of 0.72 kC (≈4 h treatment), yielding a colorless effluent with COD below 400 mg L⁻¹ and an energy consumption of 5.4 kWh m⁻³. Under these conditions, the biodegradability index increased to 0.35, and ecotoxicity decreased to 2.0 toxic units. Prolonging the treatment time enhanced COD removal and mineralization. However, it also led to higher energy demand and a rise in toxicity. The effluent treated for 4 h, reused as a 50:50 blend with freshwater, produced fabrics with negligible color difference ($\Delta E < 1$) and excellent fastness, meeting industrial standards and, thus, representing the most balanced compromise between pollutant removal, energy efficiency, and dyeing performance.

1. Introduction

The growing concern over water pollution has intensified the search for more sustainable industrial practices, particularly within the textile sector, one of the main contributors to global wastewater discharge. Textile wastewater, particularly from dyeing operations, is rich in dyes and auxiliary chemicals that pose serious environmental and health risks due to their toxicity, color intensity, and persistence (Dutta et al., 2021a, 2024). Conventional dyeing processes rely on synthetic dyes, which contain recalcitrant aromatic structures resistant to biodegradation and capable of bioaccumulation in aquatic systems (Dutta et al., 2021b, 2022).

Natural dyes derived from plant, animal, or mineral sources have re-

emerged as eco-friendly alternatives due to their non-toxic and biodegradable properties (Che and Yang, 2022; Pizzicato et al., 2023). However, their industrial application remains challenging because natural dyes often show low affinity for fibers, limited color fastness, and shade reproducibility issues. To overcome these limitations, the use of auxiliary agents, such as salts, surfactants, and biopolymers like chitosan, is a common practice to enhance dye fixation and color uniformity (Dey et al., 2025). These additives, while improving dyeing performance, may reduce the biodegradability of the resulting wastewater and contribute significantly to its organic load (Handayani et al., 2018; Saraiva et al., 2025). Hence, even though the dye molecules themselves are biodegradable, the overall wastewater composition may still exhibit low biodegradability, requiring adequate treatment before discharge or

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reuse (Mia et al., 2021; Saraiva et al., 2025).

Increasing water scarcity has driven the textile industry to adopt innovative water management strategies, with wastewater reuse emerging as a key approach to achieving circularity and resource efficiency. However, only a limited number of treatment technologies can reach the quality standards required for reuse in new dyeing processes (Pinto et al., 2022a). Among them, electrochemical oxidation (EO) has demonstrated great potential due to its strong oxidative power, operational simplicity, and capability to degrade complex organic compounds into mineralized end-products (Orts et al., 2019; Pinto et al., 2022a, 2022b). Boron-doped diamond (BDD) is considered one of the most effective anode materials for EO because it generates highly reactive, physisorbed hydroxyl radicals that promote non-selective oxidation and near-complete mineralization of organic pollutants (Fu et al., 2023; Souli et al., 2025). Alongside BDD, titanium-based mixed metal oxide (Ti/MMO) electrodes are regarded as highly suitable for EO applications, owing to the remarkable durability of the titanium support and the enhanced catalytic performance resulting from the incorporation of the metal oxides onto the electrode surface (Afonso et al., 2024; Qiao and Xiong, 2021). In the presence of chloride ions, these electrodes favor indirect oxidation through active chlorine species (Cuesta-Mota et al., 2024).

Although several studies have reported the EO treatment of textile wastewaters and even their reuse, most focus on synthetic dyes or simplified model solutions under controlled laboratory conditions. Thus, considering the growing environmental relevance of natural dyeing and the lack of studies on the treatment and reuse of its real wastewaters, the present study aimed to bridge this gap by evaluating the EO feasibility as a sustainable treatment approach. Reuse in textile dyeing represents a particular challenge, as the process must meet stringent industrial standards of color reproducibility and color fastness (Pinto et al., 2022a). Demonstrating that electrochemically treated natural dyeing wastewater can meet these criteria establishes both the technical feasibility and innovative character of this work.

A real wastewater obtained from the eco-dyeing of linen fabric with a natural dye extracted from *Punica granatum* (pomegranate peel) was evaluated. Owing to the high tannin content in pomegranate peels, this extract is widely used to dye natural fibers like cotton and linen (Ahire et al., 2024; da Costa et al., 2025). As the dyeing process comprised the utilization of auxiliary chemicals containing, among others, chloride, chitosan, and carbonate, the last two known to scavenge hydroxyl radicals (Li et al., 2018; Mascia et al., 2007), BDD and different Ti/MMO electrodes were evaluated, aiming to determine the most suitable electrode type considering the wastewater composition. The ecotoxicity

towards *Daphnia magna* of the natural dyeing wastewater, before and after EO treatment, was evaluated, filling another gap literature, as the ecotoxicity of wastewaters from eco-dyeing processes is poorly described. The subsequent reuse of the treated wastewater was validated through its use in new eco-dyeings, which were evaluated in terms of color difference and colour fastness.

2. Materials and methods

2.1. Eco-dyeing wastewater

The wastewater samples were obtained from the eco-dyeing process of a linen fabric with a natural dye extracted from the fruit rind of *Punica granatum*. This eco-dyeing comprised four stages, according to the schematic presented in Fig. 1. The assays were carried out on the weight of fiber (owf), in sealed stainless-steel minireactors, using a Mathis Labomat type BFA-12 equipment. The fabric:bath ratio was 1:17, using 14.7 g of linen fabric and 250 mL of aqueous bath in each minireactor.

The function and the main constituents of the products used in the different stages (information retrieved from the Safety Data Sheets of the products) are presented as supplementary material (Table S1). At the end of each eco-dyeing stage, the respective wastewater was collected from the minireactors for subsequent treatment of the mixture by EO. The wastewater characterization is presented in Table 1. The

Table 1
Characterization of the wastewater samples from linen dyeing with the natural dye AquinAT Mallow.

Parameter	Mean Value (± SD)
Color (visual)	Yellow
Absorbance at 367.5 nm	0.284 ± 0.007
pH	10.5 ± 0.2
Electrical conductivity/mS cm ⁻¹	0.65 ± 0.06
Turbidity/NTU	31 ± 6
Chemical oxygen demand/mg L ⁻¹	(50 ± 3) × 10
Biochemical oxygen demand/mg L ⁻¹	(12 ± 1) × 10
Biodegradability index	0.24
Dissolved organic carbon/mg L ⁻¹	134 ± 7
Dissolved inorganic carbon/mg L ⁻¹	21 ± 4
Total dissolved nitrogen/mg L ⁻¹	24 ± 3
Chloride/mg L ⁻¹	71 ± 7
Sulfate/mg L ⁻¹	5.5 ± 0.5
Total suspended solids/mg L ⁻¹	60.5 ± 0.2
Total dissolved solids/mg L ⁻¹	535 ± 7
EC ₅₀ (48 h)/%	44

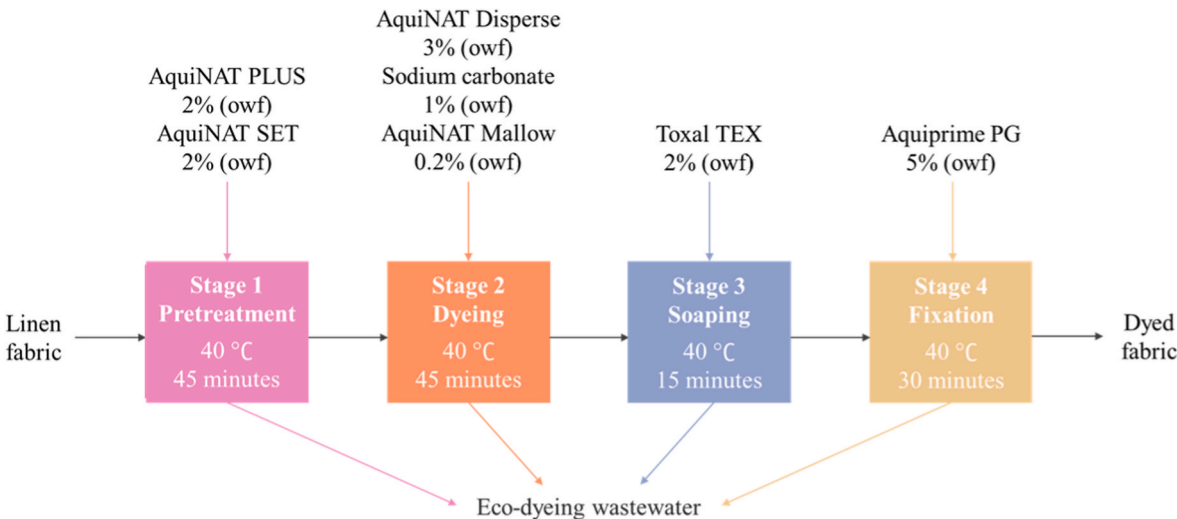


Fig. 1. Schematic diagram of the eco-dyeing process (adapted from Saraiva et al. (2025)).

concentration of the residual dye in the wastewater, (27 ± 1) mg L^{-1} , was estimated from absorbance measurements at the wavelength of the dye maximum absorbance (367,5 nm).

2.2. Electrochemical treatment

The EO experiments were conducted in batch mode with stirring (300 rpm), using an undivided cylindrical cell containing 250 mL of wastewater. The anode and the cathode, with an immersed area of 10 cm^2 and a 0.5 cm gap between them, were placed in parallel and centered in the electrochemical cell. A Multimatrix XA 3033 (0–30 V, 0–3 A) was used as the power supply. A stainless-steel plate was used as the cathode. For the anode, different materials were evaluated: BDD, purchased from NeoCoat SA, and several Ti/MMO materials, including Ti/RuO₂-TiO₂ and Ti/IrO₂-Ta₂O₅, supplied by Insoluble Anode Technology BV, and Ti/IrO₂-RuO₂, provided by UTron Technology Co., Ltd. As the Ti/MMO electrode performance is highly dependent on the material of the electrocatalytic layer coating the Ti substrate (Souli et al., 2025), the diversity of Ti/MMO compositions enabled the optimization of treatment conditions. Titanium-based RuO₂ and IrO₂ electrodes are well-known for their favorable oxygen evolution reaction potential and effectiveness in wastewater treatment (Zarei et al., 2023; Afonso et al. 2025). According to the literature, the incorporation of Ta₂O₅ improves passivation resistance and operational durability, extending the useable life of the electrode under harsh treatment conditions (Herrada et al., 2018; Guo et al., 2023).

Fig. 2 presents a schematic diagram of the experimental setup.

The EO experiments aimed at evaluating the anode material performance were conducted for 8 h at an applied current density (j) of 300 A m^{-2} . After selecting the anodes with the best performance, the influence of j on the treatment efficiency was analyzed. Experiments were carried out at 50, 100, 300, and 500 A m^{-2} , and their duration was prolonged until the applied electric charge of 3.6 kC was reached, corresponding to treatment times of 20, 10, 3.3, and 2 h, respectively. A constant charge value was selected to enable a consistent comparison of current efficiency across different current densities. The reference value of 3.6 kC was determined from preliminary experiments performed at the lowest current density (50 A m^{-2}), during which the charge required to reduce the chemical oxygen demand (COD) of the wastewater below

100 mg L^{-1} was established. Under these conditions, the target COD was achieved after approximately 20 h of electrolysis, corresponding to an accumulated charge of 3.6 kC. This charge was therefore adopted for all subsequent tests to allow a meaningful comparison of treatment efficiency under equivalent electrochemical input. The sampling frequency in these experiments was adjusted according to the applied current density to ensure representative monitoring of the physico-chemical parameters throughout the treatment. At lower current densities (50 and 100 A m^{-2}), samples were collected at longer time intervals due to the slower oxidation kinetics, whereas at higher current densities (300 and 500 A m^{-2}), shorter sampling intervals were employed to accurately capture the faster degradation profiles. In addition to these charge-controlled experiments, complementary assays were performed at fixed treatment durations of 8 h for each current density (50, 100, 300, and 500 A m^{-2}), with sampling every 2 h. All the EO experiments were performed in triplicate under controlled and reproducible conditions.

Using the optimal EO experimental conditions, sequential runs were conducted to obtain the required volume of treated wastewater for reuse experiments, extending the treatment time until COD values of 50, 100, 200, and 400 mg L^{-1} were achieved.

2.3. Reuse experiments

The reuse strategy adopted in this study is summarized in Fig. 3. The treated wastewater was reused in new eco-dyeing processes, considering the main requirements for the reuse of treated wastewater in the textile industry presented in the AquaFit4Use project (Silva et al., 2018). The reuse experiments involved the dyeing process as described in Section 2.1, but using 100 % treated wastewater and a mixture of 50 % treated wastewater and 50 % fresh water instead of solely fresh water. The 50:50 blend ratio was selected as a practical and representative condition to evaluate the feasibility of partial water substitution while ensuring consistent dyeing quality. From an industrial standpoint, the maximum achievable reuse in textile dyeing processes is approximately 70 %, as around 30 % of the process water remains absorbed in the fabric or is lost through evaporation. Therefore, the 50:50 proportion represents a realistic midpoint that allows substantial freshwater savings without compromising reproducibility or color performance. This ratio is also consistent with reported industrial practices and literature data,

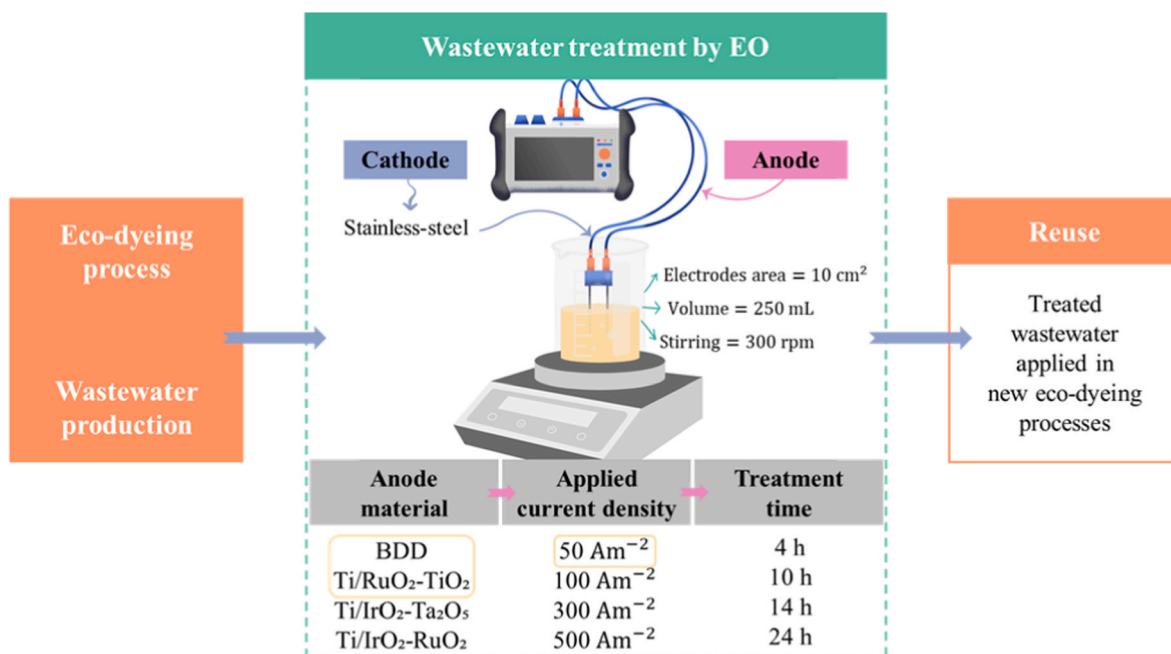


Fig. 2. Schematic diagram of the EO experimental setup.

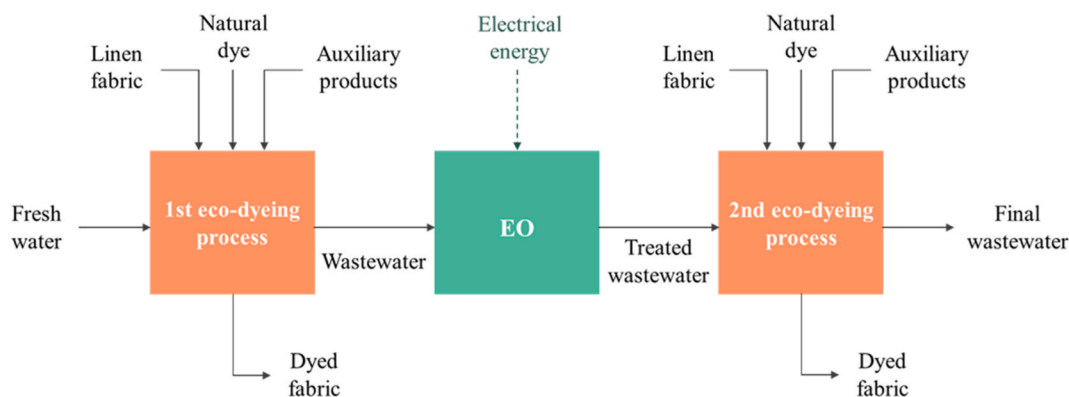


Fig. 3. Schematic representation of wastewater reuse strategy in eco-dyeing process (adapted from Pinto et al. (2022b)).

which indicate that blends containing 50–70 % recycled water provide the best compromise between water reuse efficiency and dyeing quality (Buscio et al., 2015; KLINGER Group, 2025; López-Grimau et al., 2011; Vajnhandl and Valh, 2014). Additional dyeing tests were performed using 100 % treated wastewater to explore the maximum potential for reuse. After each dyeing, the resulting fabrics were evaluated in terms of color difference (ΔE) and color fastness following standard ISO methods, as described in Section 2.4.

2.4. Analytical methods

The characterization of the wastewater samples, before and after EO treatment, was performed for the following parameters: pH, electrical conductivity (EC), turbidity, absorbance at 367.5 nm, COD, biochemical oxygen demand (BOD_5), dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), total dissolved nitrogen (TDN), ecotoxicity (EC_{50}) towards the model organism *Daphnia magna*, total suspended solids (TSS), total dissolved solids (TDS) and ions concentration.

EC was measured with a Mettler Toledo SevenEasy S30 conductivity meter, pH with a Mettler Toledo SevenEasy S20 pH meter, and turbidity with a Lovibond TB350 turbidity meter. UV–Visible absorption spectrophotometric analyses were performed at a Shimadzu UV-1800 spectrophotometer. COD determination followed the closed reflux titrimetric method, according to the Standard Methods (American Public Health Association, American Water Works Association, Water Environment Federation, 2017). BOD_5 was assessed through the respirometric approach, as detailed in previous studies (Fernandes et al., 2019). The biodegradability index was calculated based on the BOD_5/COD ratio. DOC, DIC, and TDN were determined using a Shimadzu TOC-VCPH analyzer combined with a TNM-1 unit, with all samples pre-filtered through 0.45 μm membrane filters. TSS and TDS were determined gravimetrically according to the Standard Methods (American Public Health Association, American Water Works Association, Water Environment Federation, 2017). Ion concentrations were measured by ion chromatography utilizing a Shimadzu 20A Prominence system, as previously described (Fernandes et al., 2019).

The ecotoxicity toward *Daphnia magna* was evaluated following the OECD guideline 202 (OECD, 2004), utilizing a commercial Daphtoxkit F microbiotest (DM230921). The concentration responsible for 50 % of immobilization (EC_{50}) was calculated through the standard data processing method Daphtoxkit F spreadsheet, by measuring the number of immobilized *D. magna* neonates exposed to different dilutions of the samples. The ecotoxicity results expressed as toxicity units (TU) were determined by Eq. (1) (Pablos et al., 2011).

$$TU = \frac{100}{\%EC_{50}} \quad (1)$$

The energy consumption (E , in $kWh\ m^{-3}$) and the specific energy consumption (E_{sp} , in $kWh\ kg_{COD}^{-1}$) for the electrochemical cell operation

were determined by Eqs. (2) and (3), respectively, where U is the average cell voltage, in V, resulting from the applied current intensity, I , in A, Δt is the duration of the treatment, in h, V is the volume of treated wastewater in L, and ΔCOD is the COD removed, in $g\ L^{-1}$, during Δt .

$$E = \frac{U \cdot I \cdot \Delta t}{V} \quad (2)$$

$$E_{sp} = \frac{U \cdot I \cdot \Delta t}{V \cdot \Delta COD} \quad (3)$$

To evaluate the dyeing performance, the CIELab coordinates (L^* , a^* , and b^*) and K/S values of the dyed samples were measured using a Minolta M-3600d spectrophotometer (Konica Minolta, USA) under D65 illuminant at 10° observer angle. K/S values were calculated from the diffuse reflectance R of the samples at the wavelength of maximum absorption ($\lambda_{max} = 400\ nm$). In addition, the color difference ΔE was determined from L^* , a^* , and b^* values according to the ISO 105-J03:2009 (ISO, 2009) standard. Color fastness of the fabrics dyed with the original dyeing bath and with the reused dyeing baths was evaluated following the procedure described in ISO Standards. Color fastness to rubbing (dry and wet) was tested using an AATCC Crockmeter equipment, according to the test method described by ISO 105-X12:2016 (ISO, 2016). Color fastness to washing was tested using a Linetest equipment, following the procedure provided by ISO 105-C06:2010 (ISO, 2010). Color fastness to water was determined according to ISO 105-E01:2013 (ISO, 2013a), and color fastness to perspiration was tested according to ISO 105-E04:2013 (ISO, 2013b).

3. Results and discussion

3.1. Evaluation of the performance of different anode materials

The characterization of the wastewater from the primary eco-dyeing process (Table 1) revealed concentrations of organic matter, expressed as COD and DOC, exceeding the limits established for wastewater discharge and reuse (Silva et al., 2018), thereby confirming the need for adequate treatment. These values can be attributed both to dye molecules that were not fixed onto the fibers and to the presence of auxiliary chemicals employed during the dyeing process. The yellowish coloration of the wastewater, together with the absorbance peak observed at the dye's characteristic wavelength (367.5 nm), confirms the presence of residual colorant, with an estimated concentration of approximately 27 $mg\ L^{-1}$. This observation is consistent with previous reports indicating that a substantial fraction of natural dyes used in textile dyeing processes fails to bind to the fibers and is instead released into the wastewater, contributing significantly to the overall pollutant load (Orts et al., 2019; Pizzicato et al., 2023). The wastewater biodegradability index ($BOD_5/COD = 0.24$) and acute ecotoxicity toward *Daphnia magna* (EC_{50} (48 h) = 44 %) confirmed that, although natural dyes are generally more

biodegradable and less toxic than synthetic dyes, due to their simpler molecular structures and natural origin, this assumption does not necessarily apply to real dyeing wastewaters. This is attributed to the presence of the auxiliary chemicals used to improve dye fixation, color leveling, and fastness (Mia et al., 2021; Saraiva et al., 2025). Thus, under realistic industrial conditions, wastewaters from natural dyeing processes can be nearly as recalcitrant as those from synthetic dyes, reinforcing the need for advanced oxidation technologies, such as EO, to achieve effective degradation and produce treated water suitable for reuse in new dyeing processes.

To evaluate the performance of different anode materials on the EO treatment of eco-dyeing wastewater, comparative experiments of 8-h duration, at an applied j of 300 A m^{-2} , were performed. Fig. 4 presents the COD decay with time and the specific energy consumption for the EO experiments performed with different anode materials. BDD anode was the most efficient anode material in removing organic load, followed by Ti/RuO₂-TiO₂. Similar results have been reported in the literature for the EO treatment of textile wastewaters containing synthetic dyes, which, according to the authors, are due to the greater ability of BDD to generate weakly adsorbed hydroxyl radicals (Afonso et al., 2024). Still, in the present study, the presence of chitosan and carbonate in the wastewater likely reduced BDD performance by scavenging hydroxyl radicals and limiting their availability for organic oxidation (Li et al., 2018; Mascia et al., 2007; Saraiva et al., 2025). This competitive mechanism can explain the lower COD removal observed for BDD compared to studies performed with synthetic dye wastewaters (Afonso et al., 2024).

The characterization of the treated samples from 8-h EO treatment with different anode materials is presented in the Supplementary material, Table S2. EO treatment with Ti/MMO anodes resulted in higher pH and DOC values compared to BDD, whereas DIC and TDN showed lower concentrations. These differences can be attributed to the distinct physicochemical and electrochemical characteristics of Ti/MMO electrodes. In contrast to BDD, which is a non-active anode where oxidation occurs mainly through weakly adsorbed hydroxyl radicals with high oxidation potential, Ti/MMO anodes are classified as active anodes, meaning that the oxidation process occurs through electrochemically generated higher oxide species or adsorbed chlorine intermediates on the electrode surface (Souli et al., 2025). The term “active” refers to this ability of Ti/MMO materials to participate directly in the oxidation mechanism, facilitating indirect oxidation reactions through the formation of active chlorine species in chloride-containing electrolytes (Cuesta-Mota et al., 2024). This mechanism promotes selective

oxidation of organic compounds, often stopping at partially oxidized intermediates rather than complete mineralization, which explains the higher residual COD and DOC observed after treatment. Moreover, the greater surface roughness and microscopic porosity of Ti/MMO coatings increase the electrochemically active surface area, enhancing charge transfer but also favoring adsorption of intermediate species, which can slow down full conversion to inorganic carbon. The lower DIC values obtained with Ti/MMO are consistent with this mechanism. The pH decrease in the experiments with BDD can be explained by the mineralization of the organic compounds, since H^+ is released from the involved reactions. The higher TDN removal when using Ti/MMO anodes is ascribed to the predominant oxidation of ammonium to nitrogen gas when metal oxide anodes are employed (Souli et al., 2025).

Ti/IrO₂-Ta₂O₅ and Ti/IrO₂-RuO₂ anodes were the worst performers in this comparative study, with negligible COD removals that led to extremely high E_{sp} values. Ti/RuO₂-TiO₂, although far from the organic load removal achieved with BDD, presented lower voltage values (data not shown) and, consequently, lower energy consumption, which balanced the E_{sp} value compared to BDD. The lower cell potentials attained by Ti/RuO₂-TiO₂, compared to BDD, are, according to the literature, due to facilitated direct electron transfer reactions on MMO anodes and also to higher overpotentials required for organics' oxidation on BDD (Rodríguez-Narváez et al., 2021). Considering the results attained and the high cost of BDD electrodes compared to Ti/MMO (Santacruz et al., 2024), the optimization of the EO process, in terms of current efficiency, was carried out for both BDD and Ti/RuO₂-TiO₂ anodes.

3.2. EO treatment optimization

Aiming at optimizing the EO current efficiency, experiments with BDD and Ti/RuO₂-TiO₂ anodes were carried out at current densities of 50, 100, 300, and 500 A m^{-2} . For each anode material, two experimental sets were performed: one in which the treatment time was adjusted to reach a total applied electric charge of 3.6 kC, and another in which the electrolysis was conducted for a fixed duration of 8 h. The constant-charge experiments ensured a uniform basis for comparing the effect of current density on organic load removal and energy demand, while the fixed-duration experiments allowed the evaluation of oxidation kinetics under comparable operational periods. Fig. 5 presents the COD decay as a function of the applied electric charge and, in the insets, as a function of treatment time, for these EO experiments conducted at different j .

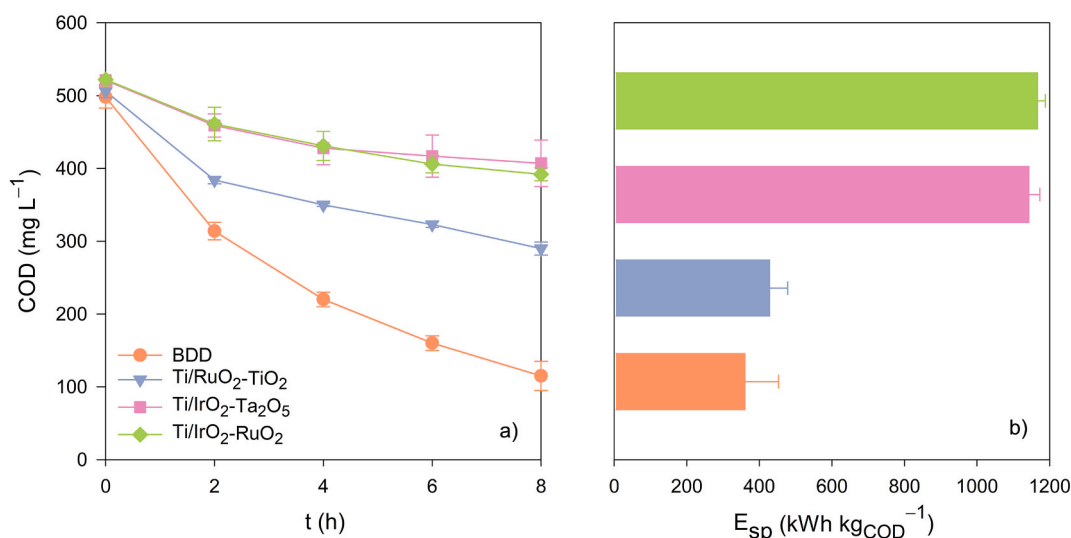


Fig. 4. (a) COD decay with time and (b) specific energy consumption for the EO experiments performed with different anode materials, at 300 A m^{-2} for 8 h.

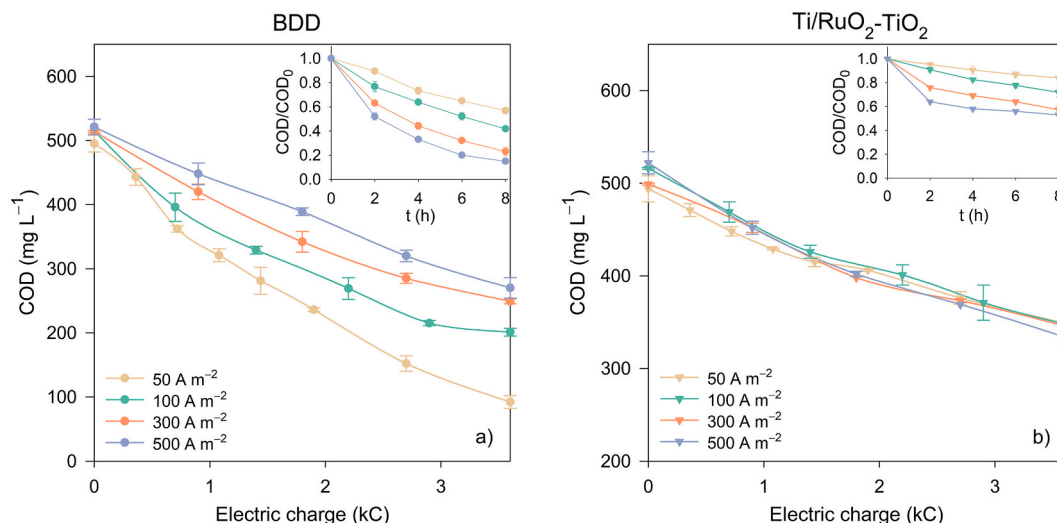


Fig. 5. COD decay with applied charge (absolute COD decay) and treatment time (normalized COD decay, inset) for the EO experiments performed at different current densities using (a) BDD and (b) Ti/RuO₂-TiO₂ anodes.

Focusing on the COD decay with applied charge, it can be observed that BDD presented higher current efficiency compared to the Ti/RuO₂-TiO₂ anode, for all the applied j . For BDD, the current efficiency increased with the decrease in j . According to the literature, this can be attributed to the enhancement of secondary reactions at higher j , such as oxygen evolution and the oxidation of inorganic ions (Panizza and Cerisola, 2009). At higher j , the COD removal rate is higher (Fig. 5 (a) inset), and the process becomes earlier diffusion-controlled, enhancing the occurrence of secondary reactions. For the EO experiments with Ti/RuO₂-TiO₂ anode, a distinct trend was observed, with similar COD decays for the different applied j , indicating similar current efficiencies. This different response of BDD and Ti/RuO₂-TiO₂ at increasing j is ascribed to the lesser reactive hydroxyl radicals produced at the Ti/MMO anode, as described above. When analyzing the COD decay with treatment time (insets), it can be observed that, for all j , BDD attains higher COD removal rates than Ti/RuO₂-TiO₂, although a regular increase in COD removal with j is observed for both anode materials.

Table 2 presents the characterization of the wastewater samples treated by EO at different applied current densities (applied electric charge = 3.6 kC), using BDD and Ti/RuO₂-TiO₂ anodes, as well as the specific energy consumption attained. Besides the lowest COD values, treatment with BDD also attained the lowest pH, turbidity, DOC, and E_{sp} values. For both anode materials, E_{sp} increased with j , probably due to the increase in the cell potential. The higher E_{sp} values observed for the Ti/MMO anode are explained by the lower COD removal, compared to BDD. These results further highlight the superior performance of the BDD anode in the treatment of wastewater from natural dyeing processes. At a fixed applied electric charge of 3.6 kC, the BDD electrode

consistently achieved greater COD removal, lower turbidity, and more effective mineralization (as reflected in lower DOC values) than the Ti/RuO₂-TiO₂ anode, across all tested j . Moreover, the BDD anode operated with lower specific energy consumption, particularly at lower current densities, reaffirming its higher oxidative efficiency. Conversely, Ti/RuO₂-TiO₂ required significantly more energy to achieve comparable COD reductions and still exhibited poorer overall performance.

The obtained results confirmed that the use of the BDD anode at low current density (50 A m⁻²) represents the most effective and energy-efficient condition for the electrooxidation of the eco-dyeing wastewater. While the Ti/RuO₂-TiO₂ anode constitutes a more economical alternative, its significantly lower efficiency and higher energy demand limit its suitability for high-quality wastewater generation intended for reuse. Therefore, the BDD anode at 50 A m⁻² current condition was chosen to further optimize the treatment in terms of its duration. For this purpose, experiments were run where the duration was prolonged until the COD achieved values below 50 (high-quality), 100, 200 (moderate quality), and 400 mg L⁻¹, considering the main requirements for the reuse of treated wastewater in the textile industry (Silva et al., 2018).

Table 3 presents the characterization of the treated wastewaters under these conditions, as well as the energy consumption for the electrochemical cell operation at the different treatment times evaluated.

Turbidity, COD, DOC, Cl⁻, and TSS decreased progressively with increasing treatment time, consistent with the enhancement of oxidation reactions at higher applied electric charges. In contrast, the biodegradability index and ecotoxicity did not follow the same trend. An increase in the biodegradability index was observed for all treatment durations

Table 2

Characterization of wastewater samples treated by EO at different applied current densities, using BDD and Ti/RuO₂-TiO₂ anodes, and specific energy consumption attained.

Parameter	BDD				Ti/RuO ₂ -TiO ₂			
	50 A m ⁻²	100 A m ⁻²	300 A m ⁻²	500 A m ⁻²	50 A m ⁻²	100 A m ⁻²	300 A m ⁻²	500 A m ⁻²
Color (visual)	Non-visible	Non-visible	Non-visible	Non-visible	Non-visible	Non-visible	Non-visible	Non-visible
Abs. 367.5 nm	–	–	–	–	–	–	–	–
pH	7.7 ± 0.1	7.4 ± 0.1	7.3 ± 0.1	7.82 ± 0.01	11.0 ± 0.4	11.12 ± 0.02	10.9 ± 0.1	7.8 ± 0.3
EC/mS cm ⁻¹	0.66 ± 0.02	0.67 ± 0.02	0.65 ± 0.04	0.65 ± 0.03	0.67 ± 0.03	0.67 ± 0.05	0.67 ± 0.08	0.65 ± 0.05
Turbidity/NTU	1.0 ± 0.2	1.8 ± 0.7	22 ± 4	34 ± 2	11 ± 1	11 ± 1	17 ± 4	20 ± 6
COD/mg L ⁻¹	92 ± 8	201 ± 6	298 ± 4	270 ± 9	349 ± 1	348.5 ± 0.2	346 ± 2	334 ± 7
DOC/mg L ⁻¹	37 ± 4	69 ± 6	63.0 ± 0.4	65 ± 3	80 ± 17	98 ± 1	98.0 ± 0.3	96 ± 1
DIC/mg L ⁻¹	34 ± 2	34 ± 1	37 ± 1	35 ± 1	37 ± 2	30.8 ± 0.3	19 ± 2	32 ± 2
TDN/mg L ⁻¹	21 ± 2	23 ± 1	25 ± 3	27.2 ± 0.4	19.8 ± 0.8	20.7 ± 0.5	22 ± 1	23.9 ± 0.8
E_{sp} /kWh kg _{COD} ⁻¹	61 ± 1	123 ± 7	296 ± 7	334 ± 7	120 ± 8	194 ± 9	364 ± 5	403 ± 6

Table 3

Characterization of wastewater samples after EO with BDD anode at 50 A m⁻² for different treatment times.

Parameter	Mean Value (± SD)			
	EO_24h	EO_14h	EO_10h	EO_4h
Color (visual)	Non-visible	Non-visible	Non-visible	Non-visible
Abs. 367.5 nm	–	–	–	–
pH	7.8 ± 0.3	7.7 ± 0.3	7.8 ± 0.3	7.3 ± 0.3
EC/mS cm ⁻¹	0.63 ± 0.04	0.59 ± 0.01	0.59 ± 0.02	0.56 ± 0.04
Turbidity/NTU	1.0 ± 0.1	1.7 ± 0.4	2.2 ± 0.4	34 ± 5
COD/mg L ⁻¹	46 ± 6	88 ± 9	170 ± 6	327 ± 8
Biodegradability index	0.65	0.65	0.74	0.35
DOC/mg L ⁻¹	32.6 ± 0.1	49 ± 1	69 ± 6	122 ± 3
DIC/mg L ⁻¹	34 ± 1	36 ± 2	36 ± 2	38 ± 2
TDN/mg L ⁻¹	23.3 ± 0.2	20 ± 3	22 ± 2	22 ± 3
Cl ⁻ /mg L ⁻¹	2.6 ± 0.1	5.51 ± 0.01	8.02 ± 0.03	29 ± 1
TSS/mg L ⁻¹	3.3 ± 0.5	2.0 ± 0.5	6.7 ± 2.5	15 ± 3
TDS/mg L ⁻¹	535 ± 1	515 ± 11	526 ± 9	491 ± 7
EC ₅₀ (48h)/%	29	29	48	50
€/kWh m ⁻³	32 ± 1	18 ± 3	15 ± 1	5.4 ± 0.2

compared to the initial wastewater value (BOD₅/COD = 0.24). Although the improvement after 4 h of treatment was modest, longer treatments (≥10 h) resulted in wastewaters classified as fairly biodegradable (BOD₅/COD >0.6) (Abdalla and Hammam, 2014), with the highest value recorded after 10 h of electrolysis (BOD₅/COD = 0.74). The modest increase observed after 4 h of treatment can be attributed to the relatively low degree of oxidation achieved under these milder conditions (short duration and lower applied current), which limited the breakdown of persistent organic compounds into more biodegradable intermediates. Conversely, the slight decline in the biodegradability index observed for treatment durations exceeding 10 h may result from the formation of oxidation by-products with lower biodegradability or higher toxicity, a hypothesis further supported by the ecotoxicity results. Indeed, although a reduction in the ecotoxicity toward *Daphnia magna* was observed after 4 h of treatment (from 2.3 to 2.0 TU), prolonged electrolysis led to an increase in toxicity, reaching 3.4 TU after 14 h and remaining stable thereafter. This increase in ecotoxicity during EO with BDD electrodes has been previously reported and is attributed to the formation of oxidation by-products that exhibit higher toxicity than the parent compounds (Fernandes et al., 2019; Souli et al., 2023). Such intermediates may include chlorinated organics and chloramines generated through secondary reactions involving chloride and nitrogen-containing species. It should be noted that the maximum ecotoxicity value obtained (3.4 TU) still falls within the toxic category (1 < TU < 10) but remains close to the threshold for non-toxic classification (TU < 1), which is consistent with the relatively low concentrations of chloride and nitrogen in the wastewater. Moreover, the primary objective of the EO treatment was not to produce discharge-quality water but to obtain wastewater suitable for reuse in new dyeing operations. Therefore, the ecotoxicity assessment provides a preliminary indication of potential acute toxicity variations resulting from EO treatment rather than a definitive measure of environmental hazard.

Regarding the energy consumption, it inevitably increases with treatment time, as longer electrolysis implies a greater applied electric charge. The specific energy consumption after 4 h of treatment was approximately 5.4 kWh m⁻³, a value comparable to that reported for industrial EO reactors and similar to the overall energy demand of conventional textile wastewater treatment plants (≈3.5 kWh m⁻³) (Buscio et al., 2019). However, when increasing the treatment time to 24 h, the energy consumption rises to 32 kWh m⁻³. While extended electrolysis improves organic load removal, it does so at the expense of higher operational costs. These results reinforce that the optimal treatment duration must balance pollutant removal, energy efficiency, and wastewater quality to ensure both technical feasibility and

environmental sustainability in water reuse applications. Similar optimization approaches have been successfully applied in advanced oxidation processes using multivariate modeling to identify the most efficient operating conditions, improving process control and energy efficiency (Gupta et al., 2020, 2021).

3.3. Reuse of treated wastewater in new dyeing processes

The wastewater treated by EO with BDD anode, at 50 A m⁻² and different treatment times, originating treated wastewater with different quality levels, was utilized in new eco-dyeing processes. The reuse experiments were carried out utilizing 100 % treated wastewater (COD <50 and 200 mg L⁻¹) and also a 50:50 mixture with fresh water (COD <100 and 400 mg L⁻¹). Despite the small scale of the dyeing experiments (250 mL drums), the methodology adopted reproduces industrial practices, as the same laboratory equipment, dyeing recipes, and auxiliaries used in industrial quality control were employed. This ensures that the findings are representative of actual textile conditions, while maintaining precise experimental control. Such small-scale trials are a recognized step in industrial workflows, preceding pilot and full-scale production, and thus provide robust and transferable data.

The fabrics obtained from reuse experiments were evaluated in terms of colorimetric coordinates, overall color difference (ΔE), and K/S values (Table 4), together with standard ISO fastness tests (Supplementary Material, Table S3). The results demonstrated negligible differences compared with fabrics dyed with fresh water. Most samples showed ΔE values below 1.0, which is considered the threshold of human visual perception (Hu et al., 2016). Remarkably, the lowest ΔE (0.5) was observed for the 50:50 mixture at COD <400 mg L⁻¹, i.e., using moderately treated water. This indicates that effective reuse can be achieved without requiring the highest degree of mineralization, thus reducing treatment time and energy demand. Likewise, K/S values remained practically unchanged across all conditions, confirming that color depth and uniformity were not compromised.

Equally important, all color fastness results (to washing, rubbing, water, and perspiration) were classified as excellent, which is particularly relevant in the case of natural dyes, as these are often associated with poor fixation and fastness limitations (Repon et al., 2024). The fact that high-quality fastness was preserved when reusing EO-treated wastewater is a strong indication of the process's feasibility. Similar findings have been reported for synthetic dye wastewaters treated by EO-BDD (Pinto et al., 2022b), reinforcing the robustness of the present results.

From an economic and environmental perspective, reuse provides significant savings. The energy demand of EO under the selected conditions (5.4 kWh m⁻³ after 4 h treatment at 50 A m⁻²) translates into a treatment cost of approximately 0.97 € m⁻³, considering the average electricity price in Portugal in 2025 (0.18 € kWh⁻¹). This value is similar to reported costs for alternative advanced oxidation or membrane processes used in dyeing wastewater reuse (Çelebi et al., 2021; Yang et al., 2020; Yin et al., 2019). In contrast, the cost of industrial water supply in Portugal ranges between 0.44 and 2.77 € m⁻³ (EMARP, 2025; EPAL, 2025; SMCB, 2025; INDAQUA, 2025), depending on region and consumption band. As for the costs related to textile wastewater treatment and management, they range from 0.29 to 2.37 € m⁻³ (PROWATER, 2007). Wastewater discharge costs into the sewage system vary between 0.052 and 0.10 € m⁻³ (European Commission, 2020). In Portugal, the fees applied to the discharge of industrial effluents are calculated by applying a base value to the amount of pollutants contained in the discharge, which varies from 0.13 to 0.30 € per kg, depending on COD and nitrogen load (Diário da República, 2008). By replacing up to 50 % of freshwater intake and reducing wastewater discharges, EO reuse not only cuts water procurement but also decreases discharge fees.

Therefore, integrating EO treatment with reuse provides a double benefit: (i) reduced environmental burden by lowering freshwater demand and pollutant discharge, and (ii) economic savings across water

Table 4

Colorimetric analysis of the fabrics dyed with fresh water and with treated wastewater under different conditions.

Parameter	Mean Value ($\pm$ SD)				
	Fresh water	100 % COD<50 mg L ⁻¹	50 % COD<100 mg L ⁻¹	100 % COD<200 mg L ⁻¹	50 % COD<400 mg L ⁻¹
L*	75.7 $\pm$ 0.4	74.6 $\pm$ 0.5	75.5 $\pm$ 0.8	74.5 $\pm$ 0.1	75.2 $\pm$ 0.4
a*	2.9 $\pm$ 0.1	2.5 $\pm$ 0.2	2.4 $\pm$ 0.1	2.6 $\pm$ 0.2	2.7 $\pm$ 0.2
b*	15.7 $\pm$ 0.4	16.9 $\pm$ 0.9	15.6 $\pm$ 0.2	16.07 $\pm$ 0.04	15.5 $\pm$ 0.3
C*	16.0 $\pm$ 0.4	17.0 $\pm$ 0.9	15.8 $\pm$ 0.2	16.28 $\pm$ 0.01	15.7 $\pm$ 0.3
h	79.5 $\pm$ 0.4	81.7 $\pm$ 0.3	81.2 $\pm$ 0.4	81.0 $\pm$ 0.7	80.1 $\pm$ 0.7
ΔE		1.1 $\pm$ 0.4	0.92 $\pm$ 0.04	0.8 $\pm$ 0.1	0.5 $\pm$ 0.2
K/S	1.19 $\pm$ 0.03	1.3 $\pm$ 0.1	1.2 $\pm$ 0.1	1.24 $\pm$ 0.02	1.2 $\pm$ 0.1

procurement and discharge taxation. Importantly, EO technology has already been demonstrated at an industrial scale. In the ECUVal project, laboratory findings led to the development of a 0.6 m² EO reactor operating at 100 A, capable of treating 4 m³ h⁻¹ of dyeing wastewater (Buscio et al., 2019), confirming the scalability of the process. These findings are consistent with the broader trend of applying advanced oxidation to complex water matrices, as demonstrated by recent studies (Gupta and Gupta, 2022; Kummar et al., 2025).

Altogether, the present results validate the technical feasibility of reusing EO-treated eco-dyeing wastewater in new dyeings without compromising color quality or fastness. Considering energy efficiency, reuse performance, toxicity control, and cost-benefit aspects, the optimal condition for industrial application corresponds to EO treatment with BDD anode at 50 A m⁻² until COD < 400 mg L⁻¹, followed by reuse in a 50:50 mixture with freshwater. This condition balances wastewater quality, treatment cost, and industrial relevance, offering a practical and sustainable solution for circular water management in textile dyeing.

4. Conclusions

This work provides experimental evidence that EO can effectively treat and enable the reuse of real wastewater from the eco-dyeing of linen with a natural dye extracted from *Punica granatum*. Despite the use of a biodegradable colorant, the dyeing wastewater exhibited low biodegradability and moderate toxicity, mainly due to the presence of dyeing auxiliaries, confirming the need for advanced treatment. Among the anode materials evaluated, BDD demonstrated the best performance, combining high organic load removal and substantial mineralization with moderate energy requirements, even in the presence of scavenging compounds such as chitosan. The treatment conducted at 50 A m⁻² for 4 h provided the most balanced outcome, slightly improving biodegradability and reducing toxicity while maintaining energy consumption at 5.4 kWh·m⁻³. Although longer treatments achieved higher COD removal and mineralization, they also increased both energy demand and the formation of toxic oxidation by-products. The reuse trials demonstrated that this 4 h treatment, applied to a 50:50 treated/fresh-water blend, ensured excellent dyeing reproducibility, color stability, and fastness, fully meeting industrial standards.

These findings demonstrate that EO with BDD constitutes a technically and environmentally viable strategy for closing the water loop in textile dyeing using natural colorants. Future work should address pilot-scale operation, by-product characterization, and system hybridization to further improve process sustainability and industrial feasibility.

CRedit authorship contribution statement

Bárbara Saraiva: Writing – original draft, Visualization, Investigation, Formal analysis, Data curation. **Nuno Belino:** Validation, Supervision. **Rui Miguel:** Project administration, Funding acquisition. **Víctor López-Grimau:** Methodology. **Valentina Buscio:** Methodology. **João Carvalho:** Validation. **Annabel Fernandes:** Writing – review & editing, Validation, Supervision, Methodology, Investigation, Conceptualization.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2025.127908>.

Data availability

Data will be made available on request.

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