

Article

Olive Leaf Waste from the Olive Oil Industry as Adsorbent Material for the Removal of Methylene Blue from Water

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Abstract

In this study, olive mill leaves (OMLs), a waste generated during the olive cleaning at the mill, were used as an adsorbent for removing methylene blue (MB) from aqueous solution. An Olive Leaf-based Adsorbent (OLA) was prepared from OMLs with minimal processing. The maximum adsorption capacity experimentally measured for OLA was $q_e = 12.8 \text{ mg} \cdot \text{g}^{-1}$. The adsorption of MB by OLA, primarily governed by electrostatic attraction, pore diffusion and weak interactions with functional groups of the leaves, is well described by the Koble–Corrigan isotherm model and pseudo-second-order kinetic model. The performance of OLA under practical environmental conditions, such as in synthetic wastewater, remained unchanged compared to tests carried out in ideal conditions. The possibility of regenerating OLA with methanol, HCl and NaCl solutions was demonstrated, allowing the material to be reused for at least three cycles with unchanged performance. A life cycle assessment was conducted to compare the impacts of OLA production with those of activated carbon, a benchmark for adsorbent materials. Overall, despite its moderate adsorptive performance, OLA stands out as a highly sustainable material for dye removal from polluted waters, owing to its exceptional cost-effectiveness, reusability, and minimal environmental impact.

Keywords: olive mill waste; adsorption; methylene blue; environmental impact; life cycle assessment



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1. Introduction

The olive sector represents a strategic component of global agriculture, with a strong geographical concentration in the Mediterranean basin but with worldwide economic and commercial relevance. According to FAO data, more than 11 million hectares are devoted to olive cultivation, and global olive production in 2023 exceeded 20 million tons per year. The vast majority of this production is destined for the olive oil industry, which places approximately 3 million tons of olive oil on the market annually [1]. The olive oil industry is responsible for the production of a large amount of waste, estimated at around 40 Mt/year

and composed mainly of olive oil mill wastewater, olive pomace and olive stones [2]. The disposal of these olive mill wastes (OMWs) presents significant environmental and technical challenges. In the past, OMWs were simply burned or dumped in the field, causing harmful effects on the environment associated with soil degradation, greenhouse gas emissions, contamination of surface and groundwater, eutrophication of water bodies and the generation of unpleasant odors [3]. Today, increasingly stringent environmental safety regulations require producers to rigorously manage olive mill byproducts through disposal according to authorized methods. This results in a significant increase in milling costs, which depends on the specific regulations in force in the country of production. However, by-products can generate an economic benefit rather than just a cost: for example, olive pomace and stones can be exploited to generate energy (both thermal and electrical), a practice also partially applied to pruning residues. This represents a low-value use of olive biomass, while a transition to high-value applications is desirable [4]. OMWs are indeed rich in organic matter and bioactive compounds. Therefore, research has increasingly shifted toward their valorization rather than their disposal through destructive treatment methods, in line with the principles of the circular economy. Consequently, OMWs are no longer considered merely as waste to be disposed of, but as a resource with significant potential for exploitation [5]. Figure 1 schematically illustrates the main valorization pathways of OMWs, ranging from more traditional approaches to those yielding higher value-added products.

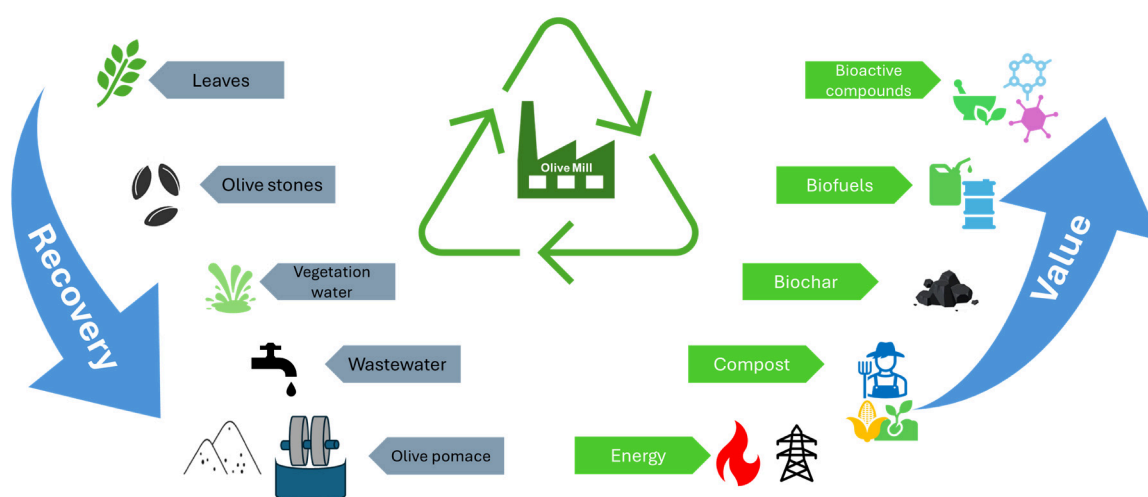


Figure 1. Schematic overview of the main strategies for the valorization of OMWs.

Among the byproducts of olive cultivation, leaves represent a waste product that is still little studied and underutilized. Most studies have focused on the extraction of bioactive compounds, primarily phenolic compounds such as oleuropein, hydroxytyrosol, verbascoside, and so on [6,7]. These molecules are known for their antioxidant, anti-inflammatory, and antimicrobial properties, and have numerous high-value applications in the nutraceutical, pharmaceutical, food, cosmetics, animal nutrition, and other sectors [4,8]. Another important line of research about leaves valorization has focused on the development of bio-adsorbent materials for the treatment of polluted water. Leaves are indeed a biodegradable and non-toxic material, ideal for removing chemical species from aqueous solutions through biological, physical, and chemical mechanisms [9]. In particular, the biomass obtained from olive leaves has proven effective for the removal of organic dyes and heavy metals from wastewater [10–12]. Among organic dyes, methylene blue (MB) was tested as it is a cationic dye widely used as a model molecule in adsorption studies,

due to its high stability, easy spectrophotometric detectability, and representativeness of emerging pollutants originating from the textile industry [10].

In this work, olive mill leaves (OMLs) were used to prepare a biosorbent for the removal of MB from water. OMLs are the leaves separated during the cleaning of olives at the mill. Although no official data are available on the quantity of leaves removed from olives during milling, this fraction can be estimated to range between 5% and 15% of the total weight of the harvested olives [6,13,14]. This variability is influenced by the type of harvester (manual or mechanical), the olive cultivar, climatic conditions, and pruning practices. This percentage is expected to increase due to the growing adoption of intensive and super-intensive cultivation systems, which rely on mechanized harvesting using shakers rather than manual harvesting. It can therefore be regarded as a highly concentrated and emerging industrial waste stream accumulated directly at processing plants that has not yet been valorized to date [7]. Unlike biomass derived from pruning, OMLs offer two major advantages: (1) they represent a much more homogeneous material, consisting primarily of leaves with only a minor proportion of thin branches; and (2) they are a lower-cost resource, as they are concentrated at the mill, reducing transportation costs, and are readily available for use [7]. Therefore, the OMLs supply chain already exists, being the same as that of olives for oil production. The exploitation of OMLs for the production of biosorbents does not compete with the extraction of phenolic compounds, since prolonged exposure to atmospheric conditions in olive mill storage yards causes the OMLs to dry and initiates biomass fermentation. As a result, this material is generally unsuitable for the extraction of bioactive molecules. Moreover, the production of olive leaves and related residues is so abundant that the valorization process we propose does not, in any way, compete with other utilization pathways. An Olive Leaf-based Adsorbent (OLA) was prepared for the first time from OMLs without any chemical treatment and with minimal processing, representing a significant advantage from a circular economy perspective, as no additional energy or material inputs are required for further transformation [2,15]. Furthermore, since OLA is derived from natural, non-chemically modified matrices, it could benefit from simplified procedures in various regulatory contexts, such as the REACH regulation (which would not require registration [16]), strengthening the potential of OLA for future applications in tertiary water treatment.

Adsorption experiments using MB as a model dye pollutant were systematically investigated from a physicochemical perspective in aqueous solutions. Furthermore, adsorption tests were conducted in simulated contaminated waters, and reusability experiments were performed on the same OLA to evaluate performance under a more sustainable operational scenario. Finally, the environmental performance of the proposed process was assessed through life cycle assessment (LCA), with impact categories analyzed and stage-specific contributions quantified to elucidate the relationships among key variables and to identify future research priorities. The environmental impacts of the proposed OLA were then benchmarked against those of conventional activated carbon-based adsorbents, whose production is associated with well-documented environmental burdens that current technological innovations seek to reduce.

2. Materials and Methods

2.1. Materials and Reagents

Methylene blue, HNO_3 , HCl , NaOH , NaCl , methanol, urea, K_2HPO_4 , $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, and beef extract were obtained from Merck Italy (Merck Life Science s.r.l., Milan, Italy).

2.2. Olive Leaf-Based Adsorbent (OLA) Preparation

The olive leaves were supplied by an oil mill located in southern Italy (Apulia region, province of Bari), during the 2022–2023 season. The olive leaves were first air-dried for approximately one month. Then, about 100 g of leaves were ground with an electric grinder (Bosch MMR08A1, BSH Hausgeräte GmbH, Munich, Germany) and washed with tap water to remove dust, color and other dissolvable impurities. Once excess water was removed, OLA was oven-dried at 323 K (Universal lab oven UF110plus, Memmert GmbH & Co KG, Schwabach, Germany, volume 108 L) until constant weight.

2.3. OLA Characterization

The particle size of OLA was assessed by sequentially using certified sieves (Endecotts Ltd., London, UK) with mesh diameters of 2 mm, 1 mm, and 500 μm . The fractions obtained were weighed and the size distribution was expressed as $\% W_{\text{fraction}}/W_{\text{tot}}$.

The specific surface area of OLA was determined by means of a gas physisorption technique using a Brunauer–Emmett–Teller (BET) Surface Area and Porosity Analyzer (Tristar II plus, Micromeritics Instrument Corporation, Norcross, GA, USA). Analyses were conducted using nitrogen gas as permeant gas and cooling the sample to approximately 81 K using liquid nitrogen.

The thermogravimetric analysis (TGA) was carried out by heating the sample in a controlled atmosphere inside a thermogravimetric analyzer (Pyris 6, PerkinElmer, Waltham, MA, USA), in a temperature range from 25 °C to 600 °C, with a scan of 10 °C·min^{−1}, and a nitrogen flow rate of 20 mL·min^{−1}.

Fourier transform infrared (FTIR) spectra in attenuated total reflectance (ATR) mode were obtained using Spectrum Two instrument (PerkinElmer, Waltham, MA, USA). The internal reflection element used for the ATR measurements was a 4 mm diameter diamond microprism with one rebound. Shredded leaf samples were analyzed by placing a sufficient amount to cover the crystalline area and using the pressure arm. Thus, 16 scans were performed, at a range of wave numbers between 4000 and 400 cm^{−1}. All spectra were acquired on dried samples.

The Z-potential of OLA in different pH conditions was determined using laser Doppler anemometry (Zetasizer Nano ZS, Malvern Panalytical Ltd., Malvern, UK). For this purpose, OLA was dispersed in deionized water (0.1% *w/w*), the pH was adjusted as required, sonicated in a water bath and in continuous mode for 5 min at 550 W and 37 kHz (Elmasonic S 300, Elma Schmidbauer GmbH, Singen, Germany), and allowed to sediment for 3 min. The supernatant fraction, free of coarse particles, was then used for the determination of the Z-potential.

A scanning electron microscope (SEM) EVO 50 XVP (LEO/ZEISS, Oberkochen, Germany) equipped with an integrated energy dispersive spectrometry (EDS) system and an SD X-MaxN detector was used to examine the surface morphology of OLA before and after adsorption. SEM analyses were performed on leaf fragments mounted on aluminum stubs and then coated with a layer of carbon using the following operating conditions: 15 kV accelerating potential, 500 pA probe current, and 8–9.5 mm working distance. Additionally, EDS was employed to analyze the elemental composition of crystalline particles on the OLA surfaces after adsorption and regeneration treatments.

2.4. Batch Adsorption Experiments

Batch adsorption experiments were carried out by adding 0.2 g of OLA to 20 mL of an aqueous solution containing MB dye in the appropriate concentration. The effect of pH was tested at a 2.0–10.0 pH range (adjusted by HCl or NaOH 0.1 M) using an MB concentration of 22.6 mg·L^{−1} at 298 K. OLA/MB suspensions were left to equilibrate for

240 min under stirring at 125 rpm. Then, an aliquot of the suspension was centrifuged for 2 min at 9000 rpm (Multispin 12 Mini Micro-Centrifuge, Giorgio Bormac S.r.l., Carpi, Italy) to separate the adsorbent material. The supernatant was recovered, and the amount of MB adsorbed per unit of mass of adsorbent materials (q_t , Equation (1)) and the removal efficiency (RE%, Equation (2)) were calculated by monitoring the absorbance peak of MB at 665 nm [17] (see calibration curve in Figure S1):

$$q_t = \frac{(C_0 - C_t)V}{m} \quad (1)$$

$$RE\% = \frac{C_0 - C_t}{C_0} \times 100 \quad (2)$$

where C_0 ($\text{mg}\cdot\text{L}^{-1}$) is the initial concentration of MB, C_t ($\text{mg}\cdot\text{L}^{-1}$) is the concentration of MB after t min of adsorption, and V (L) is the volume of the MB solution, while m is the mass of OLA (g). Once the pH was set at 7.0, the effect of contact time on the adsorption process was investigated by keeping the mass of the adsorbent material constant (0.2 g) and varying the MB concentration between $10.8 \text{ mg}\cdot\text{L}^{-1}$ and $177 \text{ mg}\cdot\text{L}^{-1}$. Then, the effect of the amount of the adsorbent material OLA (0.05, 0.1, and 0.2 g) on the adsorption process was studied, while keeping the MB concentration constant ($22 \text{ mg}\cdot\text{L}^{-1}$). Control MB adsorption tests were performed without OLA (initial MB concentration of $22 \text{ mg}\cdot\text{L}^{-1}$, $T = 298 \text{ K}$, $\text{pH} = 7.0$, equilibrium time of 240 min, stirring at 125 rpm). Since no significant variations in the MB concentration were observed between the initial value and the one after 4 h, it can be assumed that there was no degradation of the dye and that it does not adsorb onto the walls of the vial or the magnet. Consequently, the decrease in dye concentration observed during the adsorption experiments can be solely attributed to the adsorption of MB onto OLA.

To evaluate the influence of the temperature on the process, adsorption experiments were carried out at 278 K, 288 K, 298 K and 308 K, by adding 0.20 g of OLA to 20 mL of a MB solution ($23 \text{ mg}\cdot\text{L}^{-1}$). All experiments were performed in triplicate.

2.5. Regeneration of Biomass-Based Adsorbent

To mimic real dye adsorption conditions, adsorption experiments were carried out using MB solutions prepared in tap water, NaCl 0.1 M, and synthetic wastewater prepared according to OECD guidelines [18]. For the reusability experiments, the OLA adsorbent was washed three times with methanol, NaCl 0.5 M, or HCl 0.1 M (total volume 20 mL), and then rinsed with water, oven-dried at 323 K and reused for subsequent adsorption cycles ($\text{pH} 7.0$, adsorbent mass 0.20 g, MB concentration $23 \text{ mg}\cdot\text{L}^{-1}$, temperature 298 K, equilibrium time 240 min, total volume 20 mL). No significant mass loss of OLA was observed between cycles.

The experiments were carried out in triplicate, and the results were normalized to a reference value obtained in deionized water.

2.6. Life Cycle Assessment (LCA)

The LCA methodology used in this work adheres to the international guidelines established by the International Organization for Standardization (ISO) in ISO 14040/14044 [19,20]. In the examined scenario, it is assumed that the apparatus has a working life spanning over multiple decades; therefore, the environmental impact associated with its production and disposal would be distributed proportionally, with a negligible effect [21]. The system boundary of the study, identical for all the analyzed scenarios, comprises transportation of OMLs, processing, and production of the adsorbent at the gate of the laboratory (from cradle to gate). Secondary data for this study were sourced from the OpenLCA v.2.1 soft-

ware using the ecoinvent database v. 3.7 [22], which provides comprehensive life cycle inventory data, including emissions, resource usage, and environmental impacts for many materials and processes, thereby providing consistency and reliability for the data not directly generated in the study. A system expansion approach was adopted to credit the recycled material. In this way, the environmental benefits of the recycling process are completely included in the system boundaries, avoiding arbitrary attribution rules, which do not comply with the ISO standard. In the case of composting, it is taken into account that applying compost to fields replaces the use of synthetic fertilizers. All stages of the process, from the collection of OMLs to the manufacturing of adsorbent material, are integrated within the system boundaries so that all inputs (e.g., energy, transport) and outputs (e.g., recovered materials) related to recycling are taken into account, but the exclusion criterion (cut-off) was not to take into account the impacts of the production of olive leaves within the boundaries of the system, this is because the practice is to assign the environmental loads to those who produce and not to those who use the by-products. A cut-off allocation approach was adopted as OMLs were considered a residual biomass generated during olive processing and entering the analyzed system burden-free. Consequently, environmental burdens associated with olive cultivation and olive production were not allocated. The system boundaries start when the biomass becomes available at the olive mill and include transport, washing, grinding, and drying. Alternative allocation approaches (e.g., economic or mass allocation) could attribute a fraction of the upstream agricultural impacts to OLA; however, given the waste status and negligible economic value of this material, the cut-off approach was considered the most appropriate for evaluating its valorization as an adsorbent material. Moreover, this avoids the assignment of an unjustified environmental advantage to the olive producer. Our lab scale procedure to generate adsorbent material from OMLs was able to generate about 100 g per production run, and the consumption of electricity for the dehydration of the final material is very high, an unrealistic hypothesis from the point of view of industrial applications. The scale of a process plays a critical role in the assessment of the associated environmental impacts when technology is in its early stages of R&D, such as the case of recycled OMLs to produce adsorbent materials of biomass origin. In lab-scale developmental stages, the efficiency of the process is usually much lower than in the full-scale industrial phase, because of economies of scale. The goal of the experimental setup is a “proof-of-concept” demonstration as processing steps are best suited to obtain maximum performance and aimed to understand the behavior of the new material. Processes are neither automated nor optimized, resulting in manufacturing processes energetically inefficient with huge energy demand. An approach for estimating the essential properties of systems and for managing the scale-up procedure, aiming to support the definition of system boundaries, the choice of the allocation procedure and the evaluation of data requirements is given to us by Caduff [23]. The methodology is notable for using a non-linear power law combined with an extensive collection of Life Cycle Inventory (LCI) data to predict how changes in production scale affect key production variables, such as incoming and outgoing flows. The method proposed is characterized by its versatility: the empirical law of the scaling ratio has been successfully applied to various systems, such as energy conversion equipment, wind farms and heat generation technologies. Each system has specifics, but the methodology allows for the adaptation of the estimated parameters and key properties based on the distinctive characteristics of the examined process. In other words, while the power law provides a common basis for the scaling calculation, the resulting scaling factors must always be adapted to the reference technological system, which varies from power generation plants to industrial plants [24]. Thanks to this adaptability, the method offers concrete support in the design of large-scale systems, allowing the evolution of LCI flows and impact assessment results to be accurately

estimated. For this reason, we chose to analyze the small-scale production of 1 and 10 kg and the full-scale production of 100 and 1000 kg of adsorbent material from recycled OMLs. The scaling-up relationship correlates the power of the scaled equipment to the power of the lab ones using scaling factors. The power consumption of the production equipment was directly estimated from the following equation:

$$P = cSx^f \quad (3)$$

where P represents power, c is a coefficient, S is the target scale of the process, and f is the scaling factor. f and c were obtained from the size and nominal power specifications of machines with different scales. Based on literature data, in our analysis, for heating, washing, sieving and drying equipment, f and c were 0.5688 and 0.6045, respectively [25,26]. The scale-up procedure adopted in this study should be regarded as a prospective assessment intended to estimate the potential environmental evolution of the process from laboratory to industrial scale. Since industrial inventory data for OLA production is currently unavailable, the scaling factors derived from the literature were used as engineering approximations. Consequently, the obtained impact values should be interpreted as indicative estimates, and future studies based on pilot- or industrial-scale data are required to reduce uncertainty, particularly with respect to electricity consumption, drying efficiency, water use and transportation requirements.

For comparison, considering the same system boundary, the production of a functional unit of activated carbon black, the traditional scenario, as this material is commonly used as an adsorbent material, comprising transport, is the one modeled in Ecoinvent [27]. To produce granular activated carbon, hard coal undergoes carbonization and partial gasification (activation). Hard coal is first heated to temperatures over 700 °C, and the activation stage in this dataset is achieved through vaporization at temperatures between 800 and 1000 °C. Washing water has been modeled using the tap water production provided from the Ecoinvent database and the same scale factors used in the energy calculations have been adopted to estimate the consumption of water. Solid waste produced was composted, whereas wastewater from the washing process was treated in a municipal wastewater treatment plant according to the local procedures. The electricity mix used was the Italian one. Impact assessment was conducted using the CML 2001 Baseline method [28].

3. Results and Discussion

3.1. Characterization of the Adsorbent Material

Leaf grinding is almost always a necessary step in the production of biosorbents [9]. Indeed, reducing the size of the adsorbent materials not only facilitates the management of the batch adsorption process, but also increases the surface-to-mass ratio, which generally enhances adsorption capacity. However, excessive grinding to obtain a very fine powder was avoided to facilitate the recovery of the spent adsorbent in a realistic application scenario. The particle size of the OLA material obtained by grinding OMLs was determined using calibrated and certified sieves in sequence, as described in Section 2.3. Table 1 (upper part) shows that 87.2% of the adsorbent material fell within the diameter range from 500 µm to 2 mm, while only 11.6% exceeded 2 mm in diameter. After dimensional characterization, the OLA material was used as prepared for adsorption tests, without any sorting. This was done to avoid generating additional waste, to minimize process steps and, consequently, operational costs.

Table 1. Weight percentage size distribution, pore size and surface area of OLA material. Data expressed as mean $\pm$ standard deviation (SD), $n = 3$.

Property	Fraction	Value
Size distribution	$d > 2 \text{ mm}$	$(11.6 \pm 0.6)\% w/w$
	$1 \text{ mm} < d < 2 \text{ mm}$	$(66.9 \pm 0.3)\% w/w$
	$500 \mu\text{m} < d < 1 \text{ mm}$	$(20.3 \pm 0.1)\% w/w$
Specific surface area		$(0.24 \pm 0.04) \text{ m}^2 \cdot \text{g}^{-1}$
Pore size		$(58 \pm 2) \text{ \AA}$

BET analysis (Table 1, lower part) revealed that the adsorbent material has an average surface area of $(0.24 \pm 0.04) \text{ m}^2 \cdot \text{g}^{-1}$ and an average pore size of $(58 \pm 2) \text{ \AA}$. The values found are consistent with data previously obtained on non-chemically modified olive leaves. The specific surface area values are lower than those of other species, as the olive tree is a sclerophyllous plant, with the leaf surface characterized by a dense layer of cutin and epicuticular waxes that act as an effective barrier against water loss [29,30].

The main structural components of lignocellulosic biomass are cellulose, hemicellulose, and lignin [31]. The TGA curve (Figure 2A) shows an initial weight loss below 110°C , attributable to the release of residual adsorbed water from OLA. A major weight loss of about 50% occurs approximately between 200°C and 400°C , corresponding to the decomposition of cellulose and hemicellulose. Finally, lignin degradation is observed approximately between 400°C and 600°C , during which an additional 35% weight loss is recorded. The FTIR analysis was conducted to identify the chemical properties of the OLA material. In the FTIR spectrum (Figure 2B), the band between 3500 and 3100 cm^{-1} , centered at 3394 cm^{-1} , is associated with the stretching vibration of the hydroxyl groups and can be correlated with the simultaneous contribution of oleuropein, polysaccharides, cellulose, organic acids, etc., and to residual water in the plant material [32,33]. The two bands at 2921 and 2849 cm^{-1} attest to the symmetric and asymmetric stretching of CH_3 and CH_2 of long-chain aliphatic compounds, such as hemicellulose and lignin, on the outer surface of the leaves [32]. The absorption bands located between 1750 and 1600 cm^{-1} can be attributed to the stretching vibrations of the $\text{C}=\text{O}$ and $\text{C}=\text{C}$ bonds of esters, carboxylic acids, amides and aromatic rings [11]. The peak at 1464 cm^{-1} can be attributed to the C-H bending vibration, the small band between 1386 and 1366 cm^{-1} to the C-H and O-H bending [33], while the band centered at 1166 cm^{-1} could be attributed to the C-O-C stretching of polysaccharides. Finally, the intense bands between 1150 and 950 cm^{-1} correspond mainly to the endocyclic and exocyclic C-O stretching vibrations of carbohydrates as well as to organic phosphate groups (P-O-P) and carbonates (C-O) [32,34]. A table listing the assignment of functional groups to the main peaks in the infrared spectrum of OLA is provided in the Supporting Information (Table S1).

To characterize the surface morphology of the OLA material, SEM analysis was performed (Figure 3). As-prepared OLA displayed the typical morphology of olive leaf surface, characterized by densely packed stomata and a uniform distribution of trichomes (Figure 3A). In particular, the SEM images revealed well-defined epidermal cells with distinct cell walls and minimal signs of stress or damage (Figure 3B).

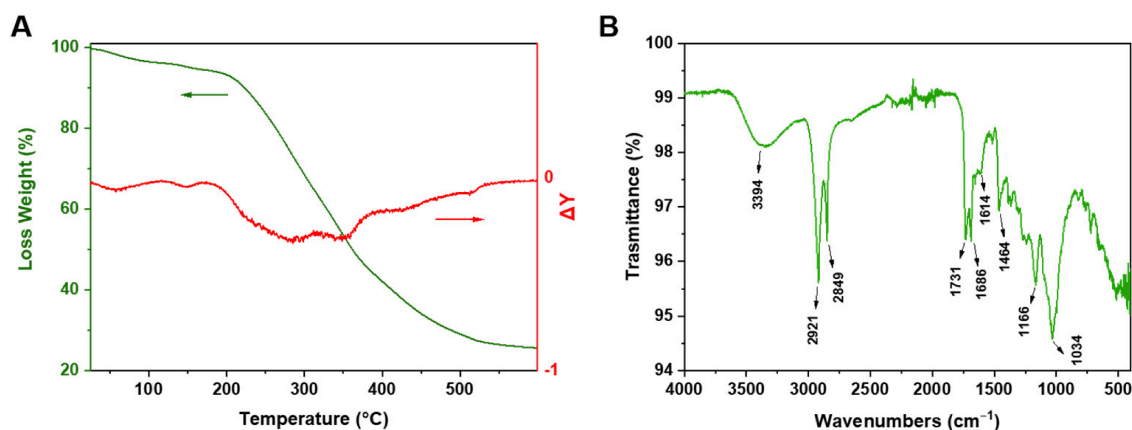


Figure 2. (A) TGA analysis (heating rate 10 °C·min⁻¹, nitrogen flow rate 20 mL·min⁻¹) and (B) FTIR spectrum of OLA.

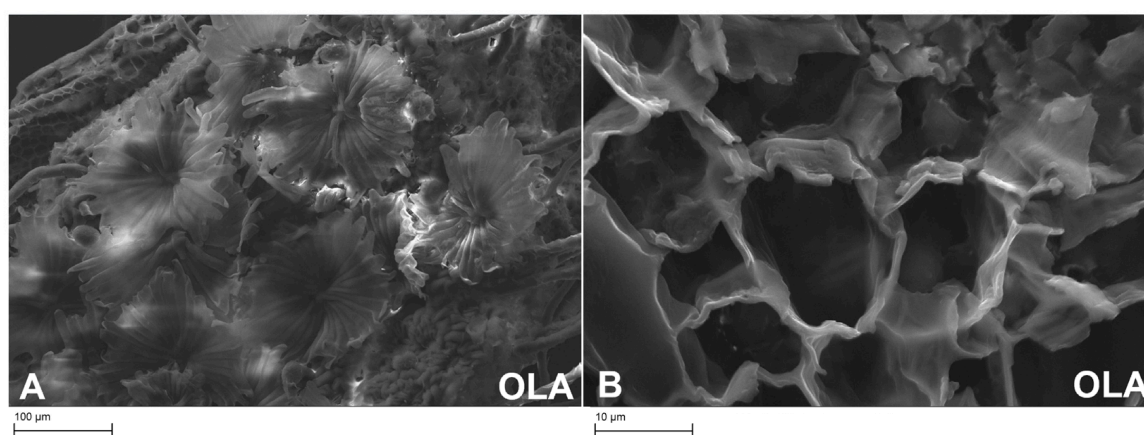


Figure 3. Microstructures of as-prepared OLA. (A) Abaxial and (B) adaxial surfaces observed by SEM.

3.2. Effect of pH on MB Adsorption

It is known that the pH of the solution greatly affects the extent of the interaction between the adsorbent and adsorbate [35,36]. The pH value, indeed, determines the degree of ionization of the adsorbate molecule and the functional groups on the surface of the adsorbent material. Z-potential measurements indicated that OLA material showed negative surface charge values in the pH range 2.0–10.0 (Figure 4A). The charge values became progressively more negative as the pH increased, probably due to the deprotonation of phenolic and carboxylic groups on the surface of the adsorbent material, as already observed for similar matrices [36]. On the other hand, MB (estimated pK_a = 3.14 [37]) exist almost entirely in the cation form in the pH range explored, and the adsorption process is favored by electrostatic interaction. By monitoring the change in equilibrium adsorption capacity of MB (q_e) with varying pH, it was found that a marked increase in q_e is observed when moving from pH 2.0 to pH 3.0, followed by a plateau between pH 3.0 and 8.0 (Figure 4B). The correlation between panels A and B of Figure 4 suggests that the adsorption of cationic MB molecules is favored by the increase in the OLA negative surface charge, primarily through electrostatic interactions. The decrease in adsorption capacity observed at markedly alkaline pH (pH 10.0) could be due to several factors, such as increased ionic strength, degradation of the adsorbent material with loss of porous structure, as well as dye aggregation phenomena, as previously observed [17,38,39]. In summary, the OLA material shows its best adsorption performance over a wide pH range, between 3.0 and 8.0, and is therefore potentially suitable for real wastewater treatment. For simplicity, subsequent adsorption experiments in this work were performed at pH 7.0.

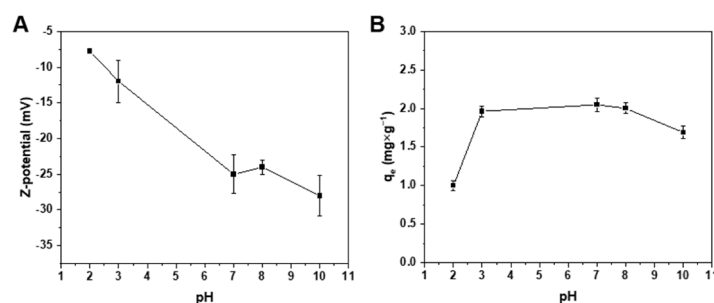


Figure 4. Dependence of the (A) Z-potential of OLA material and (B) MB adsorption on the OLA material on the pH.

3.3. Effect of Contact Time and Kinetic Study

The effect of contact time on the adsorption capacity of MB on OLA was assessed by monitoring the decrease in MB absorbance in the supernatant over time (Figure S2A) and calculated using Equation (1). Figure 5A shows the variation in q_t over time at different initial concentrations (from 10.8 to 36.3 mg·L⁻¹) of MB, whilst keeping the dose of adsorbent material and the stirring speed constant (0.20 g and 125 rpm, respectively). A typical two-stage kinetic pattern was observed for all MB initial concentrations tested. A first, very fast adsorption phase was followed by a progressive deceleration of the process, with the q_t values converging towards a constant value. Minimal variations in q_t values were recorded after the first 100 min, and the adsorption process was assumed to be practically at equilibrium at 240 min. The initial rapid adsorption phase can be attributed to the large number of available active sites on the OLA surface that readily interact with the dye molecules. In the subsequent phase, as these sites begin to fill, the adsorption rate decreases. Furthermore, repulsive interactions among the MB molecules already adsorbed on the OLA surface, as well as between the MB adsorbed molecules and those remaining in solution, contribute to reducing the adsorption rate [17,40]. At equilibrium, most active sites are occupied, the dye concentration gradient at the solution/adsorbent interface was reduced, and further adsorption becomes minimal or no longer occurs [9]. Indeed, the removal of MB was dependent on the dye concentration in solution, as often observed in batch adsorption experiments, even for leaf-based adsorbent systems [41]. Figure 5A clearly shows that q_e values exhibited a steady increase with rising dye concentrations. This trend can be attributed to the larger MB concentration gradient established between the aqueous phase and the OLA surface at higher dye levels, which facilitates the transfer of dye molecules from the bulk solution to the adsorbent and the overall adsorption capacity. Moreover, as MB concentrations grow, the probability of encounters between dye molecules and the active sites on the adsorbent becomes higher, further enhancing the adsorption process. On the other hand, once the available adsorption sites became saturated, the removal efficiency (RE%) values decreased as the dye concentration increased. (Figure S2B). In any case, at all the MB concentrations explored, the RE% at the equilibrium was in the range 80–94%. The effect of different OLA amounts (0.05, 0.1 and 0.2 g), keeping constant MB concentration, was also investigated as shown in Figure 5B,C. RE% values increased consistently with the OLA material, as a larger biomass determined more active binding sites, leading to a lower concentration of free MB in solution (blue and red curves in Figure 5B). Once near-complete removal of the dye is achieved, further increases in the amount of OLA had little effect on the RE%, but resulted in faster adsorption kinetics (red and green curves in Figure 5B). Concurrently, with increasing adsorbent dosage, the equilibrium adsorption capacity decreased due to both the underutilization of available adsorption sites and the reduced concentration gradient resulting from the rapid depletion of free dye in solution (Figure S2C).

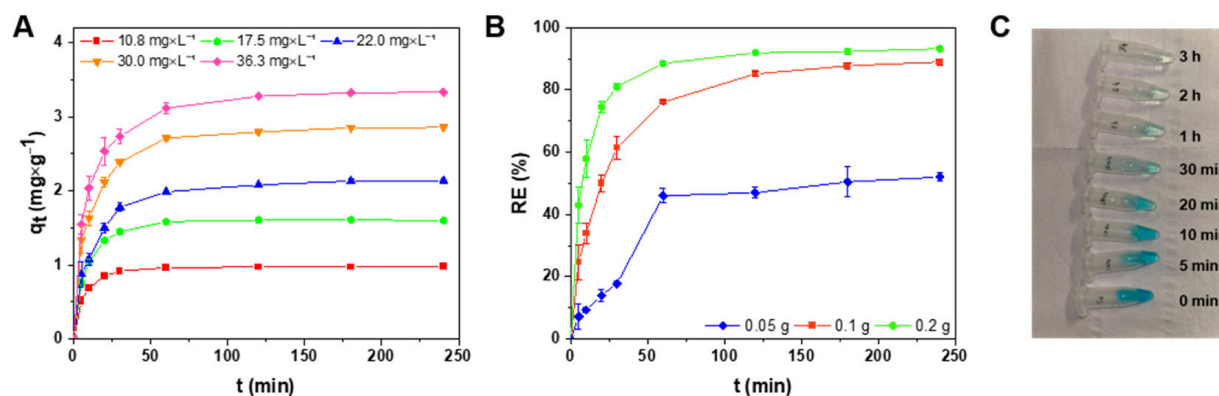


Figure 5. (A) Dependence of MB adsorption capacity onto OLA on contact time, at different values of initial dye concentration (adsorbent mass 0.20 g) and (B) dependence of MB removal efficiency onto OLA on contact time, at different adsorbent mass (MB initial concentration 22 mg·L^{−1}). (C) Decoloration of a 22 mg L^{−1} MB solution during treatment with 0.2 g OLA. All experiments were performed at T = 298 K, pH = 7.0, and results are expressed as mean ± SD, n = 3.

Kinetic studies of the MB adsorption process on OLA were carried out to obtain information on the process reaction pathways. To this purpose, experimental data were fitted with pseudo-first-order and pseudo-second-order kinetic models and intraparticle diffusion model by the linear regression method according to the following equations, respectively:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (4)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (5)$$

$$q_t = k_i t^{1/2} + c \quad (6)$$

where q_t and q_e (mg·g^{−1}) are the adsorption capacities at time t (min) of the adsorption process and at the equilibrium, respectively; k_1 (min^{−1}) and k_2 (g·mg^{−1}·min^{−1}) are the rate constants of the pseudo-first-order and pseudo-second-order kinetic models, respectively; k_i (g·mg^{−1}·min^{−0.5}) is the intraparticle rate diffusion constant.

Figure 6A,B show the application of the pseudo-first- and pseudo-second-order kinetic models to the experimental data, while Table 2 provides the corresponding estimated kinetic parameters. The data show better compliance with the pseudo-second order equation, with a higher regression coefficient R^2 and with a calculated q_e closer to the experimental value. This observation suggests that the adsorption process may involve chemisorption-related interactions, in addition to physical ones [34]. The intraparticle diffusion model was applied to clarify the mass transfer mechanisms occurring during the process. Figure 6C illustrates two independent stages involved in the adsorption of MB. The first stage represents the adsorption of MB onto the OLA surface, while the second stage indicates the diffusion of MB into the available sites within the adsorbent. The data in Table 2 show that when the initial MB concentration increases from 23 to 36.3 mg·L^{−1}, the k_1 value changes slightly (from 0.21 to 0.28), while the k_2 value remains unchanged. This suggests that different initial dye concentrations can affect only the initial stage of adsorption on the adsorbent surface, without influencing the internal diffusion process. In any case, the observed multilinearity and the absence of a straight line passing through the origin suggest that intraparticle diffusion is not the sole rate-limiting step of the adsorption process [12,42,43].

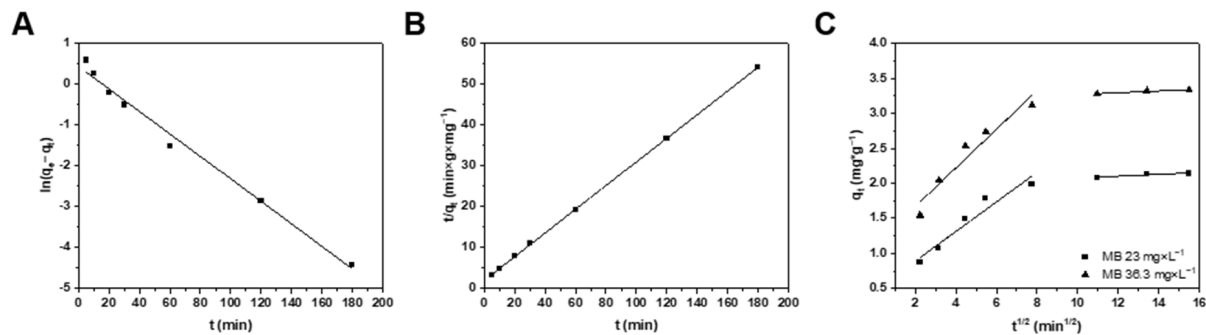


Figure 6. (A) Pseudo-first-order and (B) pseudo-second-order models at MB concentration of $36.3 \text{ mg}\cdot\text{L}^{-1}$ and $T = 298 \text{ K}$; (C) intraparticle diffusion model at 298 K and MB concentration of 23 or $36.3 \text{ mg}\cdot\text{L}^{-1}$. The black squares and triangles indicate the experimental data points, whilst the solid black line represents the fitted curve.

Table 2. Kinetic parameters of the models shown in Figure 6.

Kinetic Model	MB ($\text{mg}\cdot\text{L}^{-1}$)	$q_{e,\text{exp}}$ ($\text{mg}\cdot\text{g}^{-1}$)	$q_{e,\text{calc}}$ ($\text{mg}\cdot\text{g}^{-1}$)	k_1 (min^{-1})	k_2 ($\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1}$)	$k_{1/2}$ ($\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-0.5}$)	R^2	Reduced χ^2
Pseudo-first order	36.3	3.33 ± 0.02	1.537 ± 0.008	0.028 ± 0.001	-	-	0.9891	17.3
Pseudo-second order	36.3	3.33 ± 0.02	3.436 ± 0.017	-	0.379 ± 0.001	-	0.9999	1.5
Intraparticle diffusion	23	-	-	-	-	$k_1 0.21 \pm 0.03$ $k_2 0.013 \pm 0.005$	0.9390 0.8650	1.8 2.6
	36.3	-	-	-	-	$k_1 0.28 \pm 0.05$ $k_2 0.013 \pm 0.003$	0.9324 0.9273	0.8 1.3

3.4. Adsorption Isotherm Study

In this study, the experimental data related to the adsorption of MB onto OLA at equilibrium state under different initial MB concentration conditions were fitted with three isothermal models in order to gain insights into the adsorption mechanism.

The Langmuir model (see Equation (S1)), usually applied to describe monolayer adsorption phenomena occurring onto homogeneous surfaces with no interactions between the adsorbates [44], and the Freundlich model (see Equation (S2)), which assumes that the adsorbent surface is heterogeneous and that adsorption can occur in a multilayer fashion [45], were unable to describe the experimental data accurately as suggested by the high level of uncertainty associated with the parameters extrapolated from the fitting and the lower R^2 values (Figure S3 and Table S2). On the contrary, the Koble–Corrigan model, which is a combination of the aforementioned models, well correlates with the experimental data ($R^2 = 0.9896$, Table 3 and Figure 7A).

Table 3. Koble–Corrigan isotherm parameters for MB adsorption onto OLA at 298 K .

Isotherm	Parameters *	R^2
Koble–Corrigan	$n = 3.3 \pm 0.5$ $A = 0.90 \pm 0.15$ $B = 0.25 \pm 0.04$	0.9896

* Estimated via non-linear fitting (OriginPro 2021), subject to $A > 0$, $B > 0$ and $n > 0$.

The Koble–Corrigan isotherm model is represented by the following three-parameter equation [46]:

$$q_e = \frac{AC_e^n}{1 + BC_e^n} \quad (7)$$

where A ($L^n \cdot mg^{1-n} \cdot g^{-1}$) and B ($L \cdot mg^{-1}$) ^{n} are the Koble–Corrigan isotherm constants, C_e ($mg \cdot L^{-1}$) is the concentration at the equilibrium, n is the adsorption intensity and q_e ($mg \cdot g^{-1}$) is the adsorption capacity at the equilibrium. The value of $n = 3.3$ extrapolated from the non-linear fitting (Table 3) indicates that the contribution of the Langmuir model is small ($n > 1$), confirming the extremely heterogeneous nature of the adsorbent material, which significantly affects the adsorption mechanism.

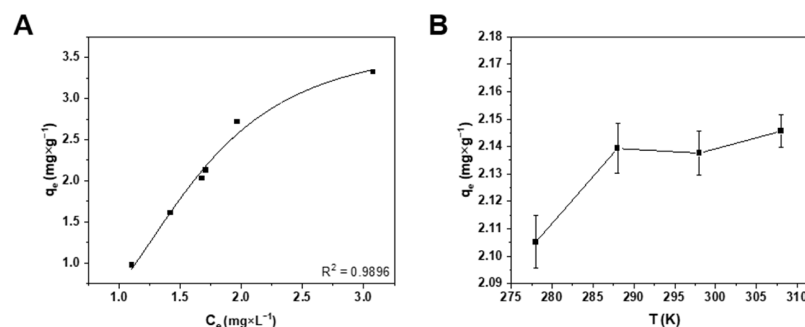


Figure 7. (A) Application of Koble–Corrigan isotherm model to the adsorption of MB onto OLA ($T = 298$ K), where the black squares indicate the experimental data points, whilst the solid black line represents the fitted curve, and (B) effect of temperature on q_e .

Comparison of adsorbent performance is generally based on the maximum adsorption capacity extrapolated from the Langmuir model. Since this model fails to describe adsorption in our case, we derived a maximum adsorption capacity from the experimental data and compared it with that of adsorbent materials derived from non-chemically treated leaves tested under similar conditions. (Table 4). The q_e value obtained for OLA is highly comparable to that of Sumac leaves and roughly twice as high as Neem leaves. It is worth noting that higher capacities reported for Banana and Plane tree leaves were achieved using significantly lower adsorbent dosages (0.0015 and 0.0016 $g \cdot mL^{-1}$, respectively). In batch adsorption, such a low adsorbent dosage typically maximizes q_e value by inducing site saturation. Conversely, the dosage used in this work (0.01 $g \cdot mL^{-1}$) prioritizes overall treatment and removal efficiency by providing an excess of active sites, which is expected to lead to a more moderate q_e value due to site unsaturation. Furthermore, OLA demonstrates competitive performance under environmentally friendly and realistic conditions, operating at neutral pH (7.0) and room temperature (298 K), unlike Plane tree leaves that require energy-intensive temperatures (333 K) or extreme pH adjustments (pH 12) to function effectively.

Table 4. Comparison of the adsorption performance of adsorbent materials derived from non-chemically treated leaves toward MB.

Leaves Source *	Adsorbent Dosage ($g \cdot mL^{-1}$)	MB Concentration ($mg \cdot L^{-1}$)	pH	T (K)	q_e ($mg \cdot g^{-1}$)	Ref.
Spent tea	0.0035	200	Not specified	303	53.16	[47]
Banana (<i>Musa sapientum</i>)	0.0015	150	Not specified	303	90.05	[48]
Neem (<i>Azadirachta indica</i>)	0.01	70	Not specified	300	6.41	[42]
Plane tree (<i>Platanus orientalis</i>)	0.0016	180	12	333	68.95	[49]
Sumac (<i>Rhus Coriaria</i> L.)	0.006	100	7.0	298	12.88	[50]
Olive (<i>Olea europaea</i>)	0.01	177	7.0	298	12.82	This work

* non-chemically treated.

3.5. Effect of the Temperature

The effect of temperature on the adsorption of MB onto OLA was investigated in the range 278–308 K, by keeping constant the MB concentration, the adsorbent mass and the pH of the solution. Figure 7B shows that the temperature has a minimal effect on the process within the considered range, with a slight increase in the adsorption capacity with increasing temperature. The determination of thermodynamic parameters by applying the van't Hoff equation to the experimental data in this context would lead to significant error propagation; consequently, the numerical reliability of the extrapolated values of ΔH and ΔS would be limited. Nevertheless, from a physicochemical perspective, the limited influence of the temperature on the q_e suggests that the interactions involved are weak.

3.6. Proposed Removal Mechanism—Comparison of Adsorbent Material Before and After Adsorption

Elucidating the adsorption mechanism is a complex challenge, since the OLA–MB interaction is evidently influenced by the simultaneous combination of several factors. Diffusion within the pores of the adsorbent material, electrostatic attraction, and other interactions with the functional groups exposed on the leaf surface are the main mechanisms involved in MB adsorption.

SEM images acquired before MB adsorption (Figure 8A,B) highlighted for OLA a typical leaf surface morphology, characterized by densely packed stomata and a uniform distribution of trichomes. The presence of such structures on the OLA surface can enhance MB adsorption by increasing the available surface area (trichomes) and creating microcavities (stomata) that facilitate pore filling and access to functional groups. Following the adsorption of MB (Figure 8C,D), SEM analysis showed only minor morphological changes, indicating that the adsorption process left the plant cells' integrity largely intact. As discussed in Section 3.2, the deprotonation of the carboxyl and phenolic groups that occurs at neutral-alkaline pH results in a negative charge on the OLA surface that is the basis of the electrostatic interaction with MB. In addition, the OLA material, rich in cellulose, hemicellulose, and lignin [31], exhibits numerous surface functional groups capable of interacting with the MB through other types of interactions as well. Aromatic structures, mainly attributable to lignin, allow for the establishment of π – π interactions between the aromatic rings of the MB and the adsorbent surface. Hydroxyl, carbonyl, ether groups, etc., can form hydrogen bonds with the dye. To better investigate this aspect, the FTIR spectrum of MB-adsorbed leaves was compared with those of MB and as-prepared OLA (Figure 8G). After adsorption, the peaks of the stretching vibration of the hydroxyl groups on the adsorbent material were shifted from 3394 cm^{-1} to 3390 cm^{-1} , indicating the involvement of O–H in the adsorption process of MB [12]. The band at 1614 cm^{-1} related to C=C stretching was attenuated, while the band at 1686 cm^{-1} related to C=O stretching was intensified and shifted to 1685 cm^{-1} . In addition, minor peaks appeared at 649 cm^{-1} (C–H aromatic groups) in the OLA material after MB adsorption, as previously observed in similar matrices [12]. Overall, the results of the FTIR analysis suggest that, in addition to π – π interactions, carbonyl and hydroxyl groups included on the surface of the adsorbent material may also be involved in the interaction with the cationic dye [51]. A schematic representation of the proposed mechanism is depicted in Figure S4.

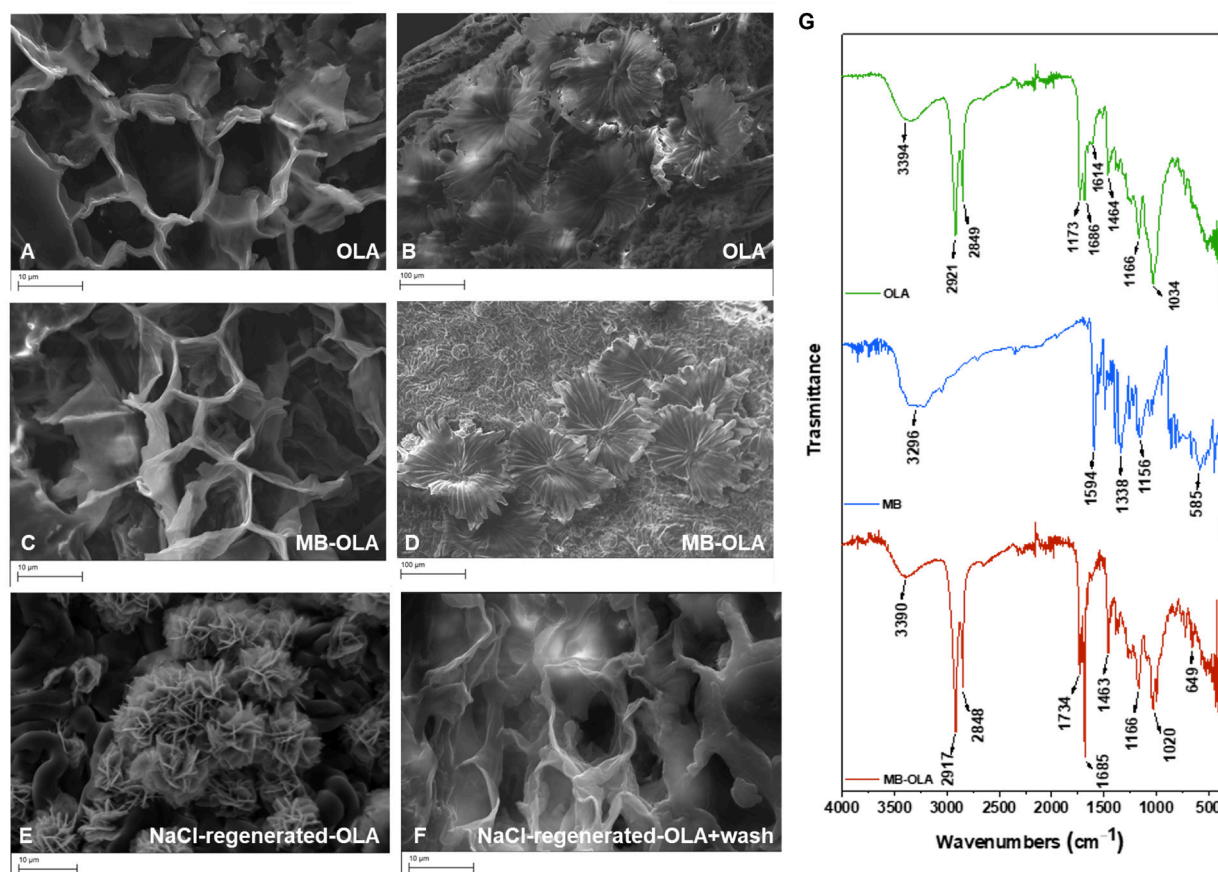


Figure 8. SEM micrographs of OLA (A,B) before, (C,D) after the adsorption of the MB, (E) after regeneration with NaCl 0.5 M and (F) after further washing with water. respectively. (G) FTIR spectra of OLA, MB and MB-adsorbed OLA.

3.7. Performance in Model Complex Matrices and Reusability

The composition of wastewater is generally complex, and the results obtained from adsorption experiments conducted in the laboratory may not represent the performance of the adsorbent material in real-world settings. To better estimate the performance of OLA under practical environmental conditions, MB adsorption tests were performed in tap water, in water supplemented with 0.1 M NaCl, and in synthetic wastewater prepared following the OECD guidelines for the testing of chemicals [18]. The results were compared with the q_e value obtained in deionized water and expressed as a relative q_e for easier interpretation (Figure 9A). In 0.1 M NaCl, a reduction in adsorption capacity of approximately 10% was observed. Ionic strength, indeed, influences both electrostatic and hydrophobic interactions in dye adsorption [52], and the overall effect can be complex and dependent on the nature of the adsorbent used [53]. However, Figure 9A shows that the adsorption capacity of OLA material towards MB remained high in all tested conditions, even in synthetic wastewater, which, in terms of compositional complexity, comes closest to a real matrix.

The suitability of an adsorbent for industrial applications depends not only on its adsorption capacity and removal efficiency, but also on its regenerability and reusability. Effective regeneration prevents the spent material from becoming single-use waste, thereby reducing operational costs and minimizing environmental impact [9]. For these reasons, adsorbent materials are tested in multiple adsorption–desorption cycles to assess whether and when significant losses in adsorption capacity occur. Furthermore, the desorption stage, in addition to regenerating the adsorbent, can simultaneously lead to the recovery of valuable adsorbates. Regeneration is typically carried out by rinsing the adsorbent material

with an organic solvent, with water or aqueous saline or acidic solutions [54]. Although OLA is an extremely cheap material, in this study, three different regeneration conditions for OLA were investigated in parallel. In detail, three successive rinses were carried out with (i) methanol, (ii) HCl 0.1 M, or (iii) NaCl 0.5 M. The desorption mechanism of MB varies by eluent: HCl promotes surface protonation of the OLA carboxyl and hydroxyl groups, triggering electrostatic repulsion with the cationic dye. Conversely, NaCl operates via a competitive ion-exchange where Na^+ ions displace MB. Lastly, MeOH breaks down hydrophobic and π - π stacking interactions between MB aromatic rings and lignin, driven by the high solubility of MB in organic solvents. Regenerated OLA was then reused in adsorption tests with MB solution, and the results were compared with the q_e value obtained with as-prepared OLA ($q_{e, \text{rel}}$). Figure 9B shows that OLA regenerated with all treatments tested maintained its adsorption capacities practically unchanged for at least three cycles of use and regeneration. SEM images acquired after the regeneration phase showed that treatment with 0.5 M NaCl caused noticeable damage to the OLA surface, indicating dehydration and osmotic stress. In addition, small crystals appeared on the leaf surface after the saline treatment (Figure 8E). EDS analysis identified these crystals as containing sodium, potassium, calcium, phosphorus, chlorine, and sulfur, suggesting they were mineral deposits from the salt wash. A subsequent rinse with tap water removed these crystals and restored the OLA original structure, indicating that the observed surface changes were reversible (Figure 8F). Given the toxicity and operational difficulties of methanol and hydrochloric acid, respectively, the use of non-toxic NaCl as a regenerating solution could be a promising strategy, although further and more in-depth desorption studies are necessary to avoid generating secondary pollutants and recover the high-value adsorbate.

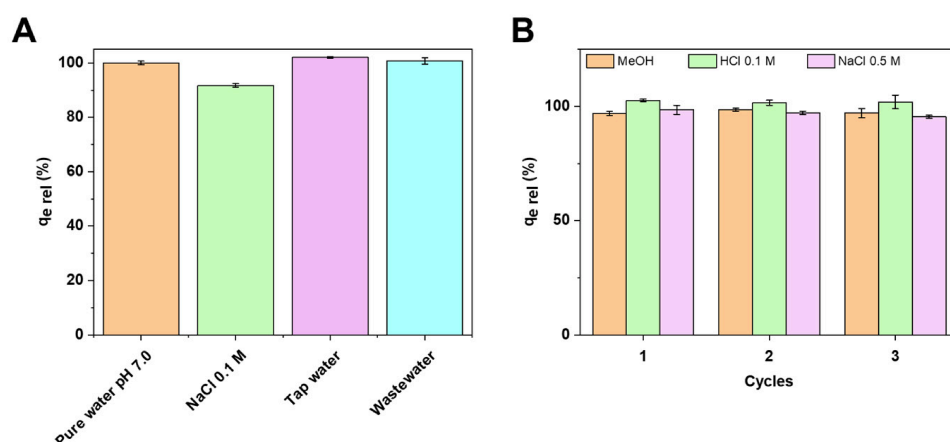


Figure 9. (A) Adsorption experiments in complex matrices and (B) Reusability of OLA in terms of adsorption of MB after washing with MeOH, HCl 0.1 M or NaCl 0.5 M.

3.8. LCA Analysis

The inventory table for the preparation of the OLA material is reported in Table S3 whereas the results of life cycle impact assessment (LCIA) using CML 2001 Baseline method, of the different analyzed scenarios (traditional activated carbon adsorbent, and small-scale (1–10 kg), and full-scale (100–1000 kg) production of 1 kg of an adsorbent material made from recycled olive leaves (functional unit) are reported in absolute values (Table 5) and as relative category indicator results (Figure S5).

Table 5. Results of life cycle impact assessment (LCIA) using CML 2001 Baseline method, of the different project scenarios (traditional activated carbon adsorbent, small-scale (1–10 kg), and full-scale (100–1000 kg) production of 1 kg of OLA material made from recycled olive leaves (functional unit).

Scale Production of OLA	OLA Material			Lab Scale Scenario 1 kg	Activated Carbon	Category Indicator
	1000 kg	100 kg	10 kg			
Impact Category						
Abiotic depletion	5.25×10^{-6}	1.40×10^{-5}	3.76×10^{-5}	1.01×10^{-4}	5.20×10^{-5}	kg Sb-eq.
Abiotic depletion (Fossil fuel)	1.51	3.72	9.69	25.8	77.2	MJ
Acidification	3.18×10^{-4}	5.95×10^{-4}	1.34×10^{-3}	3.36×10^{-3}	$9.30 \cdot 10^{-3}$	kg SO ₂ -eq.
Eutrophication	4.67×10^{-4}	1.11×10^{-3}	2.83×10^{-3}	7.49×10^{-3}	$3.51 \cdot 10^{-3}$	kg PO ₄ ³⁻ -eq.
Freshwater aquatic ecotoxicity	1.28×10^{-1}	3.40×10^{-1}	9.12×10^{-1}	2.46	$2.64 \cdot 10^{-1}$	kg 1,4-DB-eq.
Global warming (GWP100a)	1.07×10^{-1}	2.61×10^{-1}	6.76×10^{-1}	1.80	1.92	kg CO ₂ -eq.
Human toxicity	5.19×10^{-2}	1.29×10^{-1}	3.39×10^{-1}	9.04×10^{-1}	$4.97 \cdot 10^{-1}$	kg 1,4-DB-eq.
Marine aquatic ecotoxicity	68.0	173	457	1223	493	kg 1,4-DB-eq.
Ozone layer depletion	3.06×10^{-8}	7.81×10^{-8}	2.06×10^{-7}	5.53×10^{-7}	$9.79 \cdot 10^{-7}$	kg CFC-11-eq.
Photochemical oxidation	1.58×10^{-5}	3.83×10^{-5}	9.91×10^{-5}	2.63×10^{-4}	$4.05 \cdot 10^{-4}$	kg ethene-eq.
Terrestrial ecotoxicity	4.62×10^{-4}	1.21×10^{-3}	3.22×10^{-3}	8.66×10^{-3}	$1.07 \cdot 10^{-3}$	kg 1,4-DB-eq.

Considering the same functional unit, activated carbon adsorbent has quite the same environmental impact as the proposed adsorbent material made from recycled olive leaves (Table 5). By scaling the process considering the production of a functional unit, in small-scale (1–10 kg) and full-scale (100–1000 kg) capacities, the values of the category indicators decrease considerably, making it profitable from the point of view of implementing the principles of the circular economy in the production chain of oil and table olives. Traditional activated carbon adsorbent impacts mostly in fossil fuel abiotic depletion, acidification, global warming, ozone layer depletion and photochemical oxidation impact categories. This result is due to the carbon production chain, which starts from hard coal. As an example, global warming decreases by 94% from lab-scale to full-scale process (which gives 1 t of the final product) (Table 5). Contribution analysis (Figure 10A) reveals that in lab-scale production, electricity for machine operations is responsible for most of the impacts in the impact categories examined (except for ozone layer depletion potential as the production of washing water plays an equal role with respect to electricity). In the up-scale production of 1000 kg of OLA, the contribution of electricity generation decreases but remains the main cause of the impacts (except for ozone layer depletion potential), whereas the contributions of transport and composting increase (Figure 10B). Composting contributes to generating environmental credits (negative contribution), which, however, do not play a relevant role.

The scaling procedure is fundamental in the eco-design of new processes to obtain new, alternative materials, as the experimental setup is not useful to evaluate the environmental impacts in practical applications and when inventory data of producing plants are lacking. It would remain essential to understand the efficiency of the adsorption power of the new material in this study, compared to activated carbon, provided the comparison would be made in the same experimental conditions for both products, to understand exactly what the environmental advantage is in developing the new technology, as it essentially depends on the type of application in which the new adsorbent will be used. It should be noted that the present LCA evaluates the environmental burdens associated with the production of adsorbent materials and does not compare the environmental performance of both on the basis of an equivalent adsorption service. Although OLA showed satisfactory adsorption performance and reusability toward methylene blue, the amount of adsorbent required to achieve the same pollutant removal as activated carbon would depend on the specific operating conditions and adsorption capacities of the materials. Therefore, the environmental advantages identified in this study should be interpreted as production-

related benefits. A service-based comparison, expressed for example per unit mass of pollutant removed or volume of treated water, would require side-by-side adsorption and regeneration experiments performed under identical conditions and should be addressed in future work.

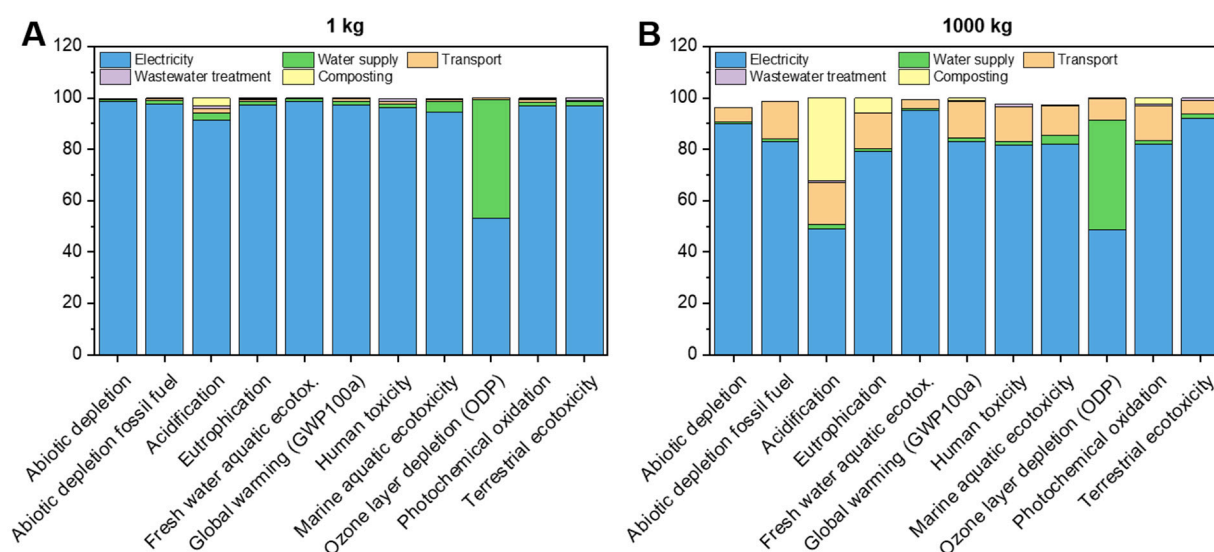


Figure 10. Contribution analysis of the processes to the category indicator results of Life Cycle Impact Assessment (LCIA) using CML 2001 Baseline method, of two different scenarios: (A) small scale (1 kg) (B) and full-scale (1000 kg) production of 1 kg of adsorbent material made from recycled olive leaves (functional unit).

4. Conclusions

The proposed study highlights the potential of OLA obtained from OMLs for the sustainable treatment of water contaminated by emerging pollutants such as MB. OLA achieved a maximum experimental adsorption capacity of $12.8 \text{ mg} \cdot \text{g}^{-1}$ for MB. Experimental data suggested that the process follows the Koble–Corrigan isotherm and pseudo-second-order kinetics. According to the complexity of OLA, the interaction mechanism between the adsorbent and the adsorbate is determined by a combination of multiple factors, including electrostatic interaction, pore diffusion, and weak interactions of the dye with the functional groups of the adsorbent.

Successful tests conducted in simulated contaminated waters, and the possibility of regenerating and reusing OLA at least three times before disposal, further increase its attractiveness. The LCA analysis enabled the assessment of the environmental impacts associated with OLA production and their comparison with those of benchmark activated carbon, demonstrating that the transition from laboratory-scale to full-scale production leads to improvements across all the impact categories considered.

Overall, despite a moderate adsorption capacity, the proposed adsorbent material has the potential to be a sustainable solution not only economically, but also from a social and regulatory perspective. Indeed, it is important to consider that the production of OLA material is based on simple and low-cost technologies, making the process accessible even to small-scale producers in Mediterranean regions. Future research to address the transition to practical applications will focus on testing the performance and selectivity of OLA with real wastewater matrices, exploring its effectiveness against a broader range of emerging contaminants under competitive conditions, and also investigating continuous-flow column studies to assess its full-scale applicability.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/su18147024/s1>, Figure S1: UV-VIS calibration curve for MB in water; Figure S2: Dependence of removal efficiency on contact time, at different values of initial MB concentration and dependence of MB adsorption capacity on contact time, at different adsorbent mass; Figure S3: Application of Langmuir and Freundlich isotherm models to the adsorption; Figure S4: Proposed mechanism of MB adsorption onto OLA material; Figure S5: Relative category indicator results of Life Cycle Impact Assessment (LCIA); Table S1: Main FTIR adsorption signals observed of OLA; Table S2: Langmuir and Freundlich isotherm parameters for MB adsorption; Table S3: LCA inventory data for the preparation of the adsorbent material.

Author Contributions: Conceptualization, V.D.L.; formal analysis, A.M.M., M.M. and P.G. (LCA modeling); investigation, A.M.M., G.V., M.C.D.T., S.S. and V.D.L.; resources, G.V. and L.C.; data curation, A.M.M.; writing—original draft preparation, A.M.M. and V.D.L.; writing—review and editing, A.M.M., P.G., G.V., M.C.D.T., S.S., M.M., V.D.L. and L.C.; visualization, A.M.M. and V.D.L.; supervision, P.G., V.D.L. and L.C.; funding acquisition, L.C. All authors have read and agreed to the published version of the manuscript.

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