

Article

Antioxidant Activity, Composition and Application of Paulownia (*Paulownia tomentosa*) Fruits, Flowers and Shells

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Abstract

Paulownia is one of the most useful and sought-after trees worldwide. Various parts (flowers, fruits, leaves, wood, bark, and seeds) have found practical applications. The present study focuses on the characterization of paulownia fruits, flowers and shells, and their potential non-traditional application for pollutant adsorption. The fruits were found to contain higher amounts of flavonoids ($1051.85 \pm 10.32 \mu\text{g mL}^{-1}$) compared to flowers ($401.26 \pm 5.48 \mu\text{g mL}^{-1}$). This reflects the observed antioxidant activity of the fruits: two times higher than that of the flowers. Salicylic acid was the main phenolic acid ($2729.96 \pm 16.89 \mu\text{g mL}^{-1}$) in the flowers, while quercetin ($572.42 \pm 10.32 \mu\text{g mL}^{-1}$) and rosmarinic acid ($352.84 \pm 6.97 \mu\text{g mL}^{-1}$) were present in higher amounts in the fruits. The lipids from the flowers were rich in linoleic (48.1%) and linolenic (20.0%) acids, while the fruit lipids were rich in linoleic (53.9%) and oleic (19.5%) acids. More than half of the fruits and flowers consisted of insoluble dietary fibers. The shells were rich in cellulose and lignin (42.96 ± 0.61 and $31.23 \pm 0.34 \text{ g } 100 \text{ g}^{-1} \text{ DW}$, respectively) and were applied as a natural biosorbent for Cu^{2+} , Cd^{2+} and methyl orange removal (99.05%). Paulownia fruits were effective, removing 98.25% of Cu^{2+} (5000 mg L^{-1}). Hence, paulownia shells and flowers could potentially be utilized as non-traditional biosorbents for pollutant removal.



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Keywords: *Paulownia tomentosa*; antioxidant activity; antimicrobial activity; polyphenols and flavonoids; fatty acids; adsorption

1. Introduction

Paulownia is among the most utilized and treasured trees. In China and elsewhere, it is grown for its multipurpose uses. Though not regarded as a regular medicinal plant species, various parts, such as leaves, flowers, fruits, wood, bark, roots and seeds, of paulownia have been used for treating a variety of ailments and diseases. Each of these parts has been shown to contain one or more bioactive components, such as ursolic acid and mattheucinol in the leaves; paulownin and desamin in the wood/xylem; syringin and catalpinoside in

the bark; and quercetin in the flowers and fruits. The fruits contain fatty oils, alkaloids, and flavonones, as well as flavonoids with antioxidant properties. The flavonoid contents and Trolox equivalent antioxidant capacity value in fresh and dry leaf extracts of paulownia suggest that this species has potential for advanced medicinal use [1].

The genus *Paulownia*, belonging to the monogeneric *Paulowniaceae* family, comprises nine species: *P. albiphloea*, *P. australis*, *P. catalpifolia*, *P. elongata*, *P. fargesii*, *P. fortunei*, *P. kawakamii*, *P. taiwaniana*, and *P. tomentosa*. *Paulownia* species are widely distributed in various regions of Asia, and some of them have also been introduced and cultivated in many countries around the world for their bright ornamental appeal and useful timber [2,3].

Apart from its extensive use as a high-quality wood in the industrial and agricultural areas, paulownia has also been used as an herbal medicine in traditional Chinese medicine. The “Compendium of Materia Medica” (a Chinese Materia Medica written by Li Shizhen in 1578) mentions paulownia bark for the treatment of hemorrhoids and worms and the flowers for reducing swelling, as well as promoting hair growth [4]. Modern medical research has identified several potential applications for the genus *Paulownia* as an antibacterial, anti-inflammatory, thirst-quenching, diuretic, antihypertensive, hemostatic, and insecticidal agent [5]. Insecticidal properties have been linked to chemicals synthesized in glandular trichomes predominantly dispersed on the surface of leaves, flower buds, and flowers [6]. Furthermore, paulownia flowers are traditionally treasured in culinary technology and as a new tobacco flavor in cigarette production [7].

An important usage is the production of bioethanol from paulownia cellulose by two approaches: using microorganisms specially selected for this purpose or by relying on the action of specific enzymes. *Paulownia* can grow at a speed of 2–3 m per year, which makes the species among the fastest-growing trees in the world. Unlike other fast-growing trees, which are characterized by relatively short life spans, a normally developing paulownia can last up to 100 years. It is highly ornamental, with magnificent flowers, blooming profusely in spring before the leaves appear, with large lilac flowers and a beautifully spreading, discreet violet aroma, particularly pleasant, used in perfumery and cosmetics [8].

The paulownia is very treasured for the quality of its wood and for its faster growth. The wood is resistant to twisting (grows straight, clean and smooth); it is resistant to contraction (no splits, no cracks) and to deformations, and it is stable during humidity changes. It is characterized by a fine texture, which makes the paulownia a tree with exceptional wood for boats, pallets, household furniture, as well as plywood, houses, prefabs, semi-products and internal panels for construction [8]. Recently, this tree has received great attention for its beneficial environmental properties and has been presented as a possible solution to the problem of deforestation at a global level, an issue that is central to the debate on climate change, with plantations representing a certain good forestry protection belt. It has very good fire resistance, having a high ignition temperature of 420–430 °C compared with the average of hardwoods, at which the ignition temperature is in the 220–225 °C range. *Paulownia* is an exotic tree from which all parts of the tree, such as the trunk, leaves, flowers, fruits and seeds, are used, and the whole tree, with its remarkable growth, is a treasure for environmentalists [8].

According to the literature data, extracts from paulownia flowers and fruits exhibit inhibitory effects against a range of pathogenic and saprophytic microorganisms due to the presence of various biologically active compounds. Its flowers, leaves, bark, roots, and fruits are used in traditional medicine for their antibacterial and anti-inflammatory properties [9]. Sławińska et al. confirmed the presence of polyphenols and flavonoids with antibacterial and antifungal activity in extracts from the flowers and fruits of *Paulownia tomentosa* [10]. Due to its rich phytochemical composition, *P. tomentosa* represents a promising source of natural bioactive compounds with potential applications in the pharmaceutical, food and

cosmetic industries. The literature survey suggested that most of the investigations focus on the antimicrobial activity of paulownia flowers and seeds, and no data could be found for paulownia fruit shells.

Water pollution by dyes is an environmental problem that is directly related to industrial development, especially in sectors such as textiles, paper, cosmetics, pharmaceuticals, and food. Synthetic dyes are widely used due to their stability, low cost and color intensity, which is why over 700,000 metric tons of synthetic dyes are produced annually, with 100,000 different types available commercially. However, these properties make them difficult to remove from wastewater, and between 12% and 20% of the total dye production is lost during the production process [11]. The textile sector is responsible for a large percentage of pollution (about 15%), as the dyes used are not fixed on the textile fibers during dyeing and go into natural water bodies without appropriate treatment [12].

Methyl orange is a synthetic organic compound from the azo dyes group, widely used in industry and analytical chemistry. It is utilized as an acid–base indicator due to its clear color change, depending on the pH of the medium, making it extremely useful in titrimetric analyses [13]. In addition to its laboratory application, the dye is also used in the textile industry as a colorant, which makes it a significant component of industrial wastewater [11]. A significant amount of dyes, including methyl orange, end up in water bodies as a result of production processes, with some of them remaining unfixed on textile materials and being discharged into the environment [14]. Methyl orange is characterized by high chemical stability and a complex molecular structure, making it difficult to degrade in natural conditions. This makes it one of the pollutants that can persist in aquatic environments for a long time. Human health might be at risk, as it has been found that methyl orange, like other synthetic dyes, can cause serious damage during prolonged exposure [13]. In the field of materials science and nanotechnology, methyl orange is used to study the adsorption properties of new materials. By monitoring the change in its concentration in solution, the effectiveness of various adsorbents, including nanomaterials and composite structures, can be assessed [15].

Copper is an element with an extremely significant role in modern society, technology and infrastructure, due to the physicochemical properties it possesses, and it is third place in importance after iron and aluminum [16]. The continuously increasing consumption of copper resources is accelerated by its prospects, which contribute to the establishment of a sustainable and low-carbon economy. In addition, in recent years, it has attracted considerable attention due to its pronounced antimicrobial properties, which determine its use as an effective material for contact surfaces [17]. Copper was recently classified as a critical and strategic raw material (metal) important for the development of the non-carbon-based industry. Judging by the available historical data, copper is one of the first metals used by human civilizations, since it can be found in its natural form, without the need for smelting [18].

Cadmium is ranked the seventh most toxic metal, having wide industrial usage. Based on studies conducted over the past century, various forms of cadmium exposure have been identified, with anthropogenic activity being the main cause of its presence in the environment [19]. A major ongoing source of pollution is its industrial application, including its use as an anti-corrosion agent, stabilizer in polyvinyl chloride (PVC) materials, as well as in the production of pigments and nickel–cadmium (Ni–Cd) batteries [20,21]. In addition to human activity, cadmium can also enter the environment as a result of natural processes, which highlights its widespread distribution and the emergence of potential environmental and health risks.

With its high efficiency in removing unwanted pollutants in a wide range of pH values and technological ease, adsorption is increasingly emerging as one of the most promising methods for decontamination of wastewater. However, on the other hand, there are significant costs for the production of suitable adsorbents, and some of them,

such as commercially available active carbon, are more difficult to regenerate after use, which limits their application on a larger scale [22–24]. Chemically modified paulownia wood (phosphorylated) was successfully applied as an effective adsorbent for removal of Ni^{2+} in simulated wastewater conditions [25]. In the study, modification was chosen for two main reasons: to improve the adsorption capacity of the wood and to introduce functional groups able to chelate metals. The reported adsorption performance of the phosphorylated paulownia wood (at 2.94 mmol g^{-1} phosphate groups) was $130.2 \text{ mg adsorbed Ni}^{2+} \text{ per g adsorbent}$. Moreover, the paulownia tree was investigated as a bioaccumulator of heavy metals (Cd, Zn and Cu), and it was found that it could be successfully utilized for phytoremediation of contaminated soils [26].

Therefore, interest in the utilization of agricultural by-products as potential alternative bioadsorbents for heavy metals and organic pollutants has increased dramatically, as they have a number of advantages over traditional methods, such as low cost, high affinity for metal ions, wide availability, renewability, and biodegradability. In addition, they exhibit both a broad-spectrum metal-binding ability and, in some cases, a selective one [27]. Plant materials are distinguished by high chemical and physical stability due to the content of cellulose, hemicelluloses, lignin and pectins, which contain carboxyl, phenolic, carbonyl and hydroxyl groups [28]. However, despite the significant advantages and their establishment as promising bioadsorbents, their use is still associated with challenges, such as the need for effective regeneration (or proper disposal after use), scaling for their subsequent application in real conditions and assessment of their adsorption capacity, which are decisive for their effectiveness [29].

With its enormous potential for growth and biomass accumulation, paulownia is among the major contributors to the successful development of the circular economy and implementation in practice of its principles. Various parts of the species have found application, but novel approaches and methods for utilization of this treasured tree are constantly being developed and introduced. Hence, the aim of the present study was to characterize several important parts of paulownia, investigating their physicochemical, antimicrobial, and antioxidant properties, and to apply paulownia fruits, flowers and shells as cheap, renewable and nature-friendly bioadsorbents for removal of pollutants (azo-dyes and heavy metals) from wastewater.

2. Materials and Methods

Materials: *Paulownia tomentosa* fruits and shells were collected in October 2025, and the flowers were picked in April 2025 during blooming (University of Food Technologies—Plovdiv, Bulgaria). The raw materials were dried at 50°C in a laboratory drier for 48 h and milled with a laboratory grain-milling machine. The milled raw materials were sieved, and the fraction with particle size between 100 and $200 \mu\text{m}$ was further used.

The following test microorganisms were used: Gram-negative bacteria: *Escherichia coli* ATCC 25922, *Salmonella enteritidis* ATCC 13076, *Proteus vulgaris* ATCC 49132, *Pseudomonas aeruginosa* NBIMCC 1390, *Klebsiella pneumoniae* subsp. *pneumoniae* ATCC 10031; Gram-positive bacteria: *Staphylococcus aureus* ATCC 6538P, *Enterococcus faecalis* ATCC 19433, *Bacillus subtilis* ATCC 19659, *Bacillus cereus* ATCC 14579; Yeasts: *Candida albicans* ATCC 10231; Saprophytic microorganisms: *Aspergillus niger* ATCC 16888, *Aspergillus flavus* ATCC 16883, *Penicillium chrysogenum* ATCC 10106, *Aspergillus oryzae* ATCC 26831, *Fusarium moniliforme* ATCC 48843, *Mucor luteus* ATCC 28932; Baker's yeasts: *Saccharomyces cerevisiae* ATCC 9763. All strains were deposited in the culture collection of the Department of Microbiology and Biotechnology at the University of Food Technologies, Plovdiv, Bulgaria. The Luria–Bertani glucose (LBG) agar with composition: tryptone (10 g L^{-1}), yeast extract (5 g L^{-1}), NaCl (10 g L^{-1}), glucose (10 g L^{-1}) and agar (15 g L^{-1}), and with pH 7.5 was obtained from Laboratorios Conda

S.A. (Madrid, Spain). The LBG agar and saline solutions (5 g L^{-1} NaCl; Merck, Darmstadt, Germany) were sterilized (20 min at 121°C) prior to use. Dimethylsulfoxide (DMSO) and nystatin (8000–13,000 units mL^{-1}) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Amikacin (250 mg mL^{-1}) was obtained from Fillab Ltd. (Plovdiv, Bulgaria).

Active carbon was purchased from Himtex Ltd. (Dimitrovgrad, Bulgaria). Al_2O_3 was obtained from Reanal (Budapest, Hungary), and chromatographic silica gel was from Merck (Darmstadt, Germany). Methyl orange was purchased from PPH Polskie Odczynniki Chemiczne (Gliwice, Poland).

Methods: Total, insoluble and soluble dietary fibers were determined using K-TDFR-100A (Megazyme, Wicklow, Ireland), according to the instructions provided by the manufacturer. Moisture content was determined with the moisture analyzer KERN DAB 100-3 (Kern & Sohn GmbH, Balingen, Germany). Water activity was measured using Labmaster aw Neo (Novasina, Lachen, Switzerland).

The determination of non-cellulosic polysaccharides, anhydrouronic acids, cellulose, and lignin was performed as described by Pla et al. [30].

Crude lipid content was determined after extraction of the samples with n-hexane (Merck, Darmstadt, Germany) using a Soxhlet extractor as described [31]. The extracted crude lipid fractions were subjected to transesterification with methanol in the presence of sulfuric acid, and their fatty acid composition was determined using a gas chromatograph Agilent 8860 (Santa Clara, CA, USA) with a flame ionization detector and a capillary column DB-Fast FAME (Agilent; Santa Clara, CA, USA), according to the procedure described by Teneva et al. [31].

Protein content was determined employing the Bradford method [32], using AMRESCO E535-kit (AMRESCO, Solon, OH, USA), with bovine gammaglobulin as standard. In brief, 100 μL extract was mixed with 1000 μL of the Bradford reagent, vortexed, kept at room temperature for 2 min, and the absorption at 595 nm with a 1 cm cuvette was measured using an LLG-uniSPEC 2 UV-Vis spectrophotometer (LLG Labware, Meckenheim, Germany).

The 70% ethanolic extracts were prepared as follows: In a sealed container of 100 mL 5 g of milled raw paulownia material was placed, and 50 mL 70% ethanol was added. The containers were put on a shaking water bath VWR VLSB18 (VWR International, Radnor, PA, USA), shaken for 1 h at 60°C , and then left for 24 h at room temperature and filtered through filter paper. The solid residue was returned to the containers and extracted one more time, but with 25 mL 70% ethanol under the same conditions. Both extracts were combined, and the volume was measured. The extracts were stored at -18°C before being analyzed.

Total polyphenolic and flavonoid content: The total polyphenolic content (TPC) of paulownia 70% ethanol extracts was assessed according to the method of Ivanov et al. using the Folin–Ciocalteu reagent (Sigma-Aldrich, St. Louis, MO, USA) [33]. Results were expressed as mg equivalents of gallic acid (GAE) per mL of extract. The total flavonoid content (TFC) of paulownia 70% ethanol extracts was evaluated following the method described by Ivanov et al. [33]. The results were expressed as mg quercetin equivalents (QE) per mL of extract.

Individual phenolic acids and flavonoids: Determination of individual phenolic acids and flavonoids in extracts was performed by HPLC with a UV-VIS detector (Waters, Milford, MA, USA). A total of 20 μL of extract was injected into the system, and the separation and determination were performed using a C18 column (Supelco Discovery HS; $5 \mu\text{m}$, $25 \text{ cm} \times 4.6 \text{ mm}$) (Merck KGaA, Darmstadt, Germany). Elution was performed with 1% acetic acid (Phase A) and methanol (Phase B) with the following changes of Solvent A: 0 to 36 min Solvent A decreased from 90% to 78%; 36 to 37 min decrease from 78% to 70%; 37 to 47 min decrease from 70% to 60%; 47 to 58 min decrease from 60% to 54%; 58 to 59 min decrease from 54% to 40%; 59 to 71 min decrease from 40% to 20%; 71 to 72 min increase from 20% to 90%; 72 to 75 min hold to 90%. The mobile-phase flow rate used was 1.0 mL min^{-1} . The gallic,

protocatechuic, vanillic, syringic, *p*-coumaric, and salicylic acids, (+)-catechin, (+)-epicatechin, and hesperidin were detected at $\lambda = 280$ nm, whereas the rosmarinic, chlorogenic, caffeic, and ferulic acids, rutin, quercetin, and kaempferol were detected at $\lambda = 360$ nm. Quantification was done by retention times and calibration curves of external standards.

2.1. Antioxidant Activity

2.1.1. DPPH Radical Scavenging Assay

The DPPH assay was performed using DPPH (2,2-diphenyl-1-picrylhydrazyl) reagent (Sigma-Aldrich, St. Louis, MO, USA) as described [33]. The antioxidant activity was expressed as mmol Trolox equivalents (TE) per gram dry extract.

2.1.2. ABTS Radical Scavenging Assay

The ABTS assay was performed using ABTS (2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid)) reagent (Sigma-Aldrich, St. Louis, MO, USA) as described [33]. The antioxidant activity was expressed as mmol Trolox equivalents (TE) per gram dry extract.

2.1.3. Ferric-Reducing Antioxidant Power (FRAP) Assay

The FRAP assay was performed according to the method of Ivanov et al. [33] using 2,4,6-Tris(2-pyridyl)-s-triazine (TPTZ) (Sigma-Aldrich, St. Louis, MO, USA). The antioxidant activity was expressed as mmol Trolox equivalents (TE) per gram dry extract.

2.1.4. Cupric Reducing Antioxidant Capacity (CUPRAC) Assay

The CUPRAC assay was performed as described [33] using copper (II)—neocuproine (2,9-dimethyl-1,10-phenanthroline) (Sigma-Aldrich, St. Louis, MO, USA) reagent. The antioxidant activity was expressed as mmol Trolox equivalents (TE) per gram dry extract.

2.2. Antimicrobial Activity

Antimicrobial activity was evaluated against the specified test microorganisms. For this purpose, fungi and yeasts were cultivated on LBG agar at 30 °C for a period of 3 to 7 days, while bacterial cultures were incubated at 37 °C for 24 to 48 h.

Spore suspensions from each test microorganism, with a concentration of 10^6 – 10^7 colony-forming units (CFU) mL^{-1} , were prepared and used for deep inoculation on LBG agar medium. After solidification of the medium, wells with 6 mm diameter were drilled, into which 0.06 mL of the corresponding samples were added. The Petri dishes were then incubated at 30 °C or 37 °C, depending on the type of microorganism used.

Following ethanol extraction, the solvent was removed under reduced pressure using a rotary evaporator. The concentrated extracts were then lyophilized to obtain dry residues, which were dissolved in 10% DMSO-water solution prior to antimicrobial activity evaluation.

2.3. Adsorption of Contaminants

Washing of the adsorbents was performed as follows: 100 g of the initial material was placed in a Buchner funnel and washed with 1600 mL (in total) deionized H_2O , 0.1 M H_2SO_4 (Merck, Darmstadt, Germany) or 0.05 M NaOH (Fillab Ltd., Plovdiv, Bulgaria). The solvent was added in portions of 400 mL. After the second portion of solvent was drained, vacuum was applied to remove all of the solvent remaining. The solid residue remaining after washing was dried at 60 °C for 48 h.

2.3.1. Adsorption of Copper Cations

The initial Cu^{2+} solution (5000 mg L^{-1}) was prepared by dissolving 19.65 g $\text{CuSO}_4 \times 5\text{H}_2\text{O}$ (Fillab Ltd., Plovdiv, Bulgaria) in 1 L deionized H_2O and stored at room temperature

for use throughout the study. From this solution, a $1000 \text{ mg L}^{-1} \text{ Cu}^{2+}$ solution was obtained by dilution. A total of 1 g of sample and 20 mL of 5000 or $1000 \text{ mg L}^{-1} \text{ Cu}^{2+}$ solution were placed in a test tube. The tubes were placed on an MLW THYS 2 shaker (VEB MLW Labortechnik, Ilmenau, Germany) and shaken for 20 min. After the specified time, the contents of the test tube were filtered twice: first through a pleated filter and then through a syringe filter with a $0.45 \mu\text{m}$ pore size (CA, ISOLAB, Laborgeräte GmbH, Eschau, Germany). Analysis for Cu^{2+} concentration determination was performed with Spectroquant® Copper Test (Merck, Darmstadt, Germany) according to the instructions of the manufacturer. Adsorption was measured at 600 nm using a 1 cm cuvette with an LLG-uniSPEC 2 UV-Vis spectrophotometer (LLG Labware, Meckenheim, Germany), and the concentration of copper ions remaining after adsorption was calculated using a standard curve prepared with aqueous copper solutions of known concentrations.

2.3.2. Adsorption of Cadmium Cations

The initial Cd^{2+} solution ($24,540 \text{ mg L}^{-1}$) was prepared by dissolving 49.82 g $\text{CdCl}_2 \times 2.5\text{H}_2\text{O}$ (Merck, Darmstadt, Germany) in 1.0 L deionized H_2O and stored at room temperature for use throughout the study. From this solution, a $613.5 \text{ mg L}^{-1} \text{ Cd}^{2+}$ solution was prepared by 40-fold dilution. The adsorption procedure was followed as described for the copper cations. Furthermore, in alkaline medium, cadmium ions react with 1-(4-nitrophenyl)-3-(4-phenylazophenyl)triazene, forming a red-colored complex, which could be determined photometrically. The analysis for Cd^{2+} concentration determination was performed with Spectroquant® Cadmium Test (Merck, Darmstadt, Germany) according to the instructions of the manufacturer. The adsorption was measured at 525 nm using a 1 cm cuvette with an LLG-uniSPEC 2 UV-Vis spectrophotometer (LLG Labware, Meckenheim, Germany), and the concentration of cadmium ions remaining after adsorption was calculated using a standard curve prepared with aqueous cadmium solutions of known concentrations.

2.3.3. Adsorption of Methyl Orange

The adsorption of methyl orange dye was carried out as follows: 1 g adsorbent was weighed into 50 mL centrifuge tubes, and 20 mL water solution of methyl orange (1000 mg L^{-1}) was added. The tubes were placed on a laboratory shaker MLW THYS 2 (VEB MLW Labortechnik, Ilmenau, Germany). The shaker was started, and at certain intervals (3, 6, 12, 24, 48, 60, 90, 120 min) a centrifuge tube was removed from the shaker, and the mixture was filtered, first through filter paper, and then through a CA $0.45 \mu\text{m}$ syringe filter (ISOLAB, Laborgeräte GmbH, Eschau, Germany). The adsorption of the filtrate was measured at 465 nm using a 1 cm cuvette with an LLG-uniSPEC2 UV-Vis spectrophotometer (LLG Labware, Meckenheim, Germany). The concentration of methyl orange remaining after adsorption was calculated using a standard curve prepared with known concentrations of methyl orange.

The adsorption capacity of the adsorbents was calculated using the formula (Equation (1)):

$$q = \frac{(C_0 - C_e)V}{m} \quad (1)$$

where C_0 and C_e are the initial and the concentration at the equilibrium point (mg L^{-1}), respectively, V is solution volume (in L) and m is the adsorbent amount used (g).

Infrared spectroscopy with Fourier transformation (IR-FT): The infrared spectra of the paulownia shells (initial and pretreated) were taken using Jasco FT-IR6x with ATR Pro ONE X (Jasco, Tokyo, Japan). The spectra were recorded in the $4000\text{--}400 \text{ cm}^{-1}$ range with a resolution of 2 cm^{-1} , 132 scans and correction for H_2O and CO_2 before the measurements.

Statistical analysis: The experimental data (three replications) are presented as mean value $\pm$ standard deviation. For analysis, a one-way ANOVA test (Tukey's post hoc test; $p < 0.05$) was used with Microsoft Excel 2013 (additional XL Toolbox NG module installed).

3. Results and Discussion

3.1. Composition of Paulownia Fruits, Flowers and Shells

3.1.1. Physicochemical Composition of Paulownia Fruits, Flowers and Shells

The collected paulownia fruits, flowers and shells were subjected to analyses for determination of their proximate composition (Table 1). The amount of total dietary fiber in fruits ($54.30 \pm 2.10\%$) and flowers ($59.88 \pm 1.91\%$) is close, although statistically different, and insoluble dietary fiber prevails (more than 50% of the total fiber content). The shells, not surprisingly, are composed of around 85% dietary fiber, of which more than 95% are insoluble. The highest content of soluble dietary fibers was observed for paulownia flowers: $4.64 \pm 0.6\%$, which tentatively might be considered mostly pectic-type polysaccharides based on the results for anhydrouronic acid content: $4.57 \pm 0.21\%$. Guo et al. reported the presence of soluble fiber in paulownia flowers in the 3.5–6.4% range [7]. Wang et al. reported isolation of heteropolysaccharides from *Paulownia fortunei*, showing potent beneficial effects on chickens (promoting the development of the immune organs, increasing the quantities of leukocytes, and the ratio of lymphocytes, etc.) and which are composed of around 10% uronic acids [34]. The highest amount of cellulose was determined in the shells, and the shells and the flowers contain similar amounts of lignin (around 32%). The flowers had the highest amounts of hexane-soluble substances (crude lipids): $6.67 \pm 0.12\%$. The protein amounts in fruits and flowers were $11.84 \pm 0.78\%$ and $10.35 \pm 0.84\%$, respectively, while in the shells they were relatively low: $2.34 \pm 0.14\%$. Cui et al. [35], investigating the chemical composition of paulownia flowers, reported the presence of 10.8–14.8% proteins, which is in agreement with the present findings. The degree of esterification of polysaccharides in fruits and flowers was 88.15 ± 5.34 and $91.29 \pm 2.26\%$, respectively, and the values were not statistically different.

Table 1. Proximate composition of paulownia fruits, flowers and shells (in % except for a_w).

Parameter	Paulownia Fruits	Paulownia Flowers	Paulownia Shells
Proteins	$11.84^1 \pm 0.78^a$	10.35 ± 0.84^a	2.34 ± 0.14^b
Cellulose	28.62 ± 0.24^b	17.69 ± 0.20^c	42.96 ± 0.61^a
Lignin	25.33 ± 0.41^c	32.70 ± 0.39^a	31.23 ± 0.34^b
NCP	4.51 ± 0.21^b	4.95 ± 0.20^b	8.89 ± 0.19^a
Lipids	2.09 ± 0.05^b	6.67 ± 0.12^a	-
Ash	6.48 ± 0.06^a	4.31 ± 0.05^b	2.81 ± 0.11^c
Moisture	11.24 ± 0.06^a	9.20 ± 0.08^b	9.31 ± 0.19^b
TDF	54.30 ± 2.10^c	59.88 ± 1.91^b	84.23 ± 1.68^a
IDF	50.65 ± 2.13^c	55.24 ± 1.89^b	81.91 ± 1.68^a
SDF	3.65 ± 0.19^b	4.64 ± 0.62^a	2.32 ± 0.18^c
GalA	2.34 ± 0.19^b	4.57 ± 0.21^a	0.27 ± 0.01^c
DE	88.15 ± 5.34^a	91.29 ± 2.26^a	22.45 ± 0.08^b
PUC	15.0 ± 6.82^a	11.93 ± 1.03^b	3.11 ± 0.16^c
a_w , 25 °C	0.458 ± 0.018^a	0.312 ± 0.015^b	0.467 ± 0.015^a

¹ Results are presented as the mean of three measurements; ^{a,b,c} Different letters in rows indicate statistically different values (Tukey's HSD test, $p < 0.05$). NCP—Non-Cellulosic Polysaccharides; TDF—Total Dietary Fibers; IDF—Insoluble Dietary Fibers; SDF—Soluble Dietary Fibers; GalA—Anhydrouronic Acids; DE—Degree of Esterification of polysaccharides; PUC—PolyUronic acid Content; a_w —Water Activity.

3.1.2. Individual Phenolic Acids and Flavonoids in Paulownia Fruit, Flower and Shell Extracts

The extraction of paulownia fruits, flowers and shells yielded 51 ± 0.58 mL, 48 ± 0.67 mL and 55 ± 0.48 mL, respectively. The fruits, flowers and shells extracts' dry matter content was $2.79 \pm 0.12\%$, $2.57 \pm 0.11\%$ and $1.89 \pm 0.12\%$, respectively. Six phenolic acids (protocatechuic, syringic, *p*-coumaric, ferulic and rosmarinic acid) and six flavonoids ((+)-catechin, (-)-epicatechin, hesperidin, quercetin and kaempferol) were identified by HPLC (Table 2) in the 70% ethanol extracts. In all studied extracts (flowers, fruits and shells), salicylic acid predominates. Salicylic acid concentration in the flower extract reached $2729.96 \pm 16.89 \mu\text{g mL}^{-1}$. In the fruit extract, the content of salicylic acid was more than six times lower ($426.98 \pm 5.84 \mu\text{g mL}^{-1}$), while in the shell extract, it was over tenfold lower compared to the flower extract. Protocatechuic acid was identified only in the shell extract, and it was present in negligible amounts. Among the hydroxycinnamic acids, rosmarinic acid was detected at the highest concentration in the paulownia fruit extract ($352.84 \pm 6.97 \mu\text{g mL}^{-1}$). Its concentration in the flower extract was approximately 1.7 times lower. Regarding flavonoids, quercetin was the major compound in the fruit extract ($572.42 \pm 10.32 \mu\text{g mL}^{-1}$), followed by (+)-catechin ($120.93 \pm 4.12 \mu\text{g mL}^{-1}$), and rutin ($101.05 \pm 4.38 \mu\text{g mL}^{-1}$). Rutin ($91.77 \pm 2.91 \mu\text{g mL}^{-1}$) and quercetin ($82.73 \pm 3.91 \mu\text{g mL}^{-1}$) were the predominant flavonoids in the flower extract, while the shell extract contained comparatively low levels of all identified flavonoids. Kaempferol was detected only in the flower extract at a concentration of $1.84 \pm 0.26 \mu\text{g mL}^{-1}$ (Table 2). Quercetin and quercetin-3-O- β -D-glucoside were also found in higher amounts in 70% ethanol *P. tomentosa* flower extracts by Dai et al. [36].

Table 2. Phenolic acids and flavonoids ($\mu\text{g mL}^{-1}$) in paulownia fruit, flower and shell 70% ethanol extracts.

Compound	Paulownia Fruits	Paulownia Flowers	Paulownia Shells
Phenolic acids			
Protocatechuic acid	-	-	0.58 ± 0.04 ¹
Syringic acid	2.92 ± 0.05 ^c	6.14 ± 0.13 ^a	4.59 ± 0.03 ^b
Salicylic acid	426.98 ± 5.84 ^b	2729.96 ± 16.89 ^a	249.66 ± 10.32 ^c
<i>p</i> -Coumaric acid	10.92 ± 0.02 ^b	14.47 ± 0.57 ^a	0.89 ± 0.05 ^c
Ferulic acid	10.92 ± 0.02 ^b	15.24 ± 0.91 ^a	4.96 ± 0.20 ^c
Rosmarinic acid	352.84 ± 6.97 ^a	197.41 ± 5.48 ^b	6.54 ± 0.05 ^c
Flavonoids			
(+)-Catechin	120.93 ± 4.12 ^a	13.52 ± 0.91 ^c	26.82 ± 0.94 ^b
(-)-Epicatechin	22.49 ± 2.59 ^a	20.90 ± 3.68 ^a	-
Rutin	101.05 ± 4.38 ^a	91.77 ± 2.91 ^b	9.83 ± 0.48 ^c
Hesperidin	25.54 ± 0.49 ^b	27.51 ± 0.96 ^a	23.91 ± 1.48 ^b
Quercetin	572.42 ± 10.32 ^a	82.73 ± 3.91 ^b	1.35 ± 0.34 ^c
Kaempferol	-	1.84 ± 0.26	-

¹ Results are presented as the mean of three measurements; ^{a,b,c} Different letters in rows indicate statistically different values (Tukey's HSD test, $p < 0.05$).

3.1.3. Total Polyphenolic Compounds and Antioxidant Activity of Paulownia Fruit, Flower and Shell Extracts

The total phenolic content (TPC) was highest in paulownia fruit extract, reaching 5.11 ± 0.10 mg GAE mL⁻¹, followed by flower extract (3.04 ± 0.05 mg GAE mL⁻¹) and shell extract (1.29 ± 0.04 mg GAE mL⁻¹)—Table 3. A similar trend was observed for the total flavonoid content (TF), with the highest value recorded in the fruit extract (1.36 ± 0.01 mg QE mL⁻¹), while the flower extract contained significantly lower amounts (0.77 ± 0.02 mg QE mL⁻¹). Flavonoids were detected in the shell extract in trace amounts.

Table 3. Total polyphenolic compounds and antioxidant activity of paulownia fruits, flowers and shells 70% ethanol extracts.

Parameter	Unit	Paulownia Fruits	Paulownia Flowers	Paulownia Shells
TPC	mg GAE mL ⁻¹	5.11 ¹ ± 0.10 ^a	3.04 ± 0.05 ^b	1.29 ± 0.04 ^c
TF	µg QE mL ⁻¹	1.36 ± 0.01 ^a	0.77 ± 0.02 ^b	traces
AO (DPPH)	mmol TE g ⁻¹	44.59 ± 0.44 ^a	18.25 ± 0.22 ^b	7.47 ± 0.16 ^c
AO (ABTS)		47.50 ± 1.05 ^a	19.20 ± 0.25 ^b	7.97 ± 0.25 ^c
AO (FRAP)		74.64 ± 3.40 ^a	19.16 ± 0.18 ^b	6.18 ± 0.17 ^c
AO (CUPRAC)		114.12 ± 2.11 ^a	64.00 ± 1.19 ^b	18.41 ± 0.65 ^c

¹ Results are presented as the mean of three measurements; ^{a,b,c} Different letters in rows indicate statistically different values (Tukey's HSD test, $p < 0.05$); TPC—total polyphenolic content; TF—total flavonoids; GAE—gallic acid equivalents; QE—quercetin equivalents; TE—Trolox equivalents; AO—antioxidant activity.

The antioxidant activity of the extracts, evaluated by DPPH, ABTS, FRAP, and CUPRAC assays, followed the same pattern as the total phenolic and flavonoid contents (Table 3). The fruit extract exhibited the strongest antioxidant capacity, with values of 44.59 ± 0.44 , 47.50 ± 1.05 , 74.64 ± 3.40 , and 114.12 ± 2.11 mmol TE g⁻¹ dry extract for DPPH, ABTS, FRAP, and CUPRAC assays, respectively. The flowers' extract showed intermediate antioxidant activity, whereas the shells' extract demonstrated the lowest values in all assays. These results indicate a strong relationship between phenolic and flavonoid contents and the antioxidant potential of the investigated paulownia extracts.

Meng et al. [37] investigated antioxidant properties of different ethanolic extracts (20%, 40%, 60%, and 80%) of paulownia flowers by scavenging DPPH, superoxide anions, and hydroxyl radical methods and compared the results with synthetic antioxidants (butylated hydroxyanisole and butylated hydroxytoluene). All extracts showed strong antioxidant properties, especially the 60% ethanolic extract, which exhibited excellent antioxidant activity. Earlier investigations of *P. tomentosa* extracts identified several phenolic acids (*p*-hydroxybenzoic, vanillic, gallic, cinnamic, *p*-coumaric, and caffeic acid in leaves, bark, and wood) and flavonoid compounds (basically C-geranylated flavonoids) and demonstrated substantial antiradical activity; however, comprehensive comparative data for fruits and shells remain limited [38].

3.1.4. Fatty Acid Composition of Glyceride Oils from Paulownia Fruits and Flowers

The isolated hexane-soluble fractions (crude lipids) were subjected to analysis for determination of their fatty acid composition (Table 4). The paulownia shell's hexane extraction did not yield substantial amounts of crude lipids, and it was not further investigated.

Table 4. Fatty acid composition (in %) of paulownia fruits and flowers.

Fatty Acid		Paulownia Fruits	Paulownia Flowers
Saturated fatty acids			
C 8:0	Octanoic acid (Caprylic acid)	-	0.5 ± 0.04 ¹
C 10:0	Decanoic acid (Capric acid)	3.3 ± 0.03 ^a	0.2 ± 0.02 ^b
C 11:0	Undecanoic acid (Undecylic acid)	1.0 ± 0.06	-
C 12:0	Dodecanoic acid (Lauric acid)	1.2 ± 0.04	-
C 13:0	Tridecanoic acid (Tridecylic acid)	0.9 ± 0.02	-
C 14:0	Tetradecanoic acid (Myristic acid)	0.2 ± 0.02 ^a	0.3 ± 0.03 ^a
C 15:0	Pentadecanoic acid (Pentadecylic acid)	0.3 ± 0.01 ^a	0.1 ± 0.01 ^b
C 16:0	Hexadecanoic acid (Palmitic acid)	7.4 ± 0.08 ^b	21.2 ± 0.09 ^a

Table 4. Cont.

Fatty Acid		Paulownia Fruits	Paulownia Flowers
C 17:0	Heptadecanoic acid (Margaric acid)	2.9 ± 0.04 ^a	0.7 ± 0.03 ^b
C 18:0	Octadecanoic acid (Stearic acid)	2.2 ± 0.09 ^a	2.4 ± 0.08 ^a
C 20:0	Icosanoic acid (Arachidic acid)	-	0.1 ± 0.01
C 21:0	Heneicosanoic acid (Heneicosylic acid)	-	0.4 ± 0.03
C 22:0	Docosanoic acid (Behenic acid)	1.2 ± 0.03 ^a	0.1 ± 0.01 ^b
C 24:0	Tetracosanoic acid (Lignoceric acid)	-	0.1 ± 0.01
Unsaturated fatty acids			
C 14:1	cis-9-Tetradecenoic acid (Myristoleic acid)	-	0.2 ± 0.01
C 15:1	trans-14-Pentadecenoic acid	0.2 ± 0.02 ^a	0.1 ± 0.01 ^a
C 16:1	cis-9-Hexadecenoic acid (Palmitoleic acid)	0.2 ± 0.01 ^a	0.1 ± 0.01 ^a
C 17:1	cis-10-Heptadecenoic acid	3.9 ± 0.07 ^a	0.2 ± 0.04 ^b
C 18:1	cis-9-Octadecenoic acid (Oleic acid)	19.5 ± 0.08 ^a	3.6 ± 0.06 ^b
C 18:2	cis,cis-9,12-Octadecadienoic acid (Linoleic acid)	53.9 ± 0.10 ^a	48.1 ± 0.11 ^b
C 18:3 n-6	cis,cis,cis-6,9,12-Octadecatrienoic acid (γ-Linolenic acid)	0.3 ± 0.02 ^a	0.3 ± 0.03 ^a
C 18:3 n-3	cis,cis,cis-9,12,15-Octadecatrienoic acid (α-Linolenic acid)	0.9 ± 0.02 ^b	20.0 ± 0.08 ^a
C 20:1 n-11	cis-9-Icosenoic acid (Gadoleic acid)	-	0.2 ± 0.02
C 20:2	cis,cis-11,14-Icosadienoic acid	-	0.1 ± 0.01
C 20:3 n-6	cis,cis,cis-8,11,14-Icosatrienoic acid (Di-homo- γ-linolenic acid)	0.2 ± 0.02 ^a	0.3 ± 0.02 ^a
C 20:3 n-9	cis,cis,cis-5,8,11-Icosatrienoic acid (Mead acid)	0.1 ± 0.01 ^a	0.1 ± 0.01 ^a
C 20:5	cis,cis,cis,cis,cis-5,8,11,14,17-Icosapentaenoic acid (Icosapentaenoic acid)	-	0.2 ± 0.01
C 22:1	cis-13-Docosenoic acid (Erucic acid)	-	0.1 ± 0.01
C 22:2	cis,cis-13,16-Docosadienoic acid (Docosadienoic acid)	0.2 ± 0.02	-
C 22:6 n-3	cis,cis,cis,cis,cis,cis-4,7,10,13,16,19-Docosahexaenoic acid (Docosahexaenoic acid, DHA)	-	0.1 ± 0.01
Saturated fatty acids, %		20.6 ± 0.09 ^b	26.1 ± 0.09 ^a
Unsaturated fatty acids, %		79.4 ± 0.11 ^a	73.9 ± 0.11 ^b
Mono-unsaturated fatty acids, %		23.8 ± 0.11 ^a	4.7 ± 0.11 ^b
Poly-unsaturated fatty acids, %		55.6 ± 0.11 ^b	69.2 ± 0.11 ^a

¹ Results are presented as the mean of three measurements; ^{a,b} Different letters in rows indicate statistically different values (Tukey's HSD test, $p < 0.05$).

The fatty acid profile of the flowers and fruits reveals a highly unsaturated acid composition, dominated by polyunsaturated fatty acids (PUFAs)—69.2 ± 0.11% and 55.6 ± 0.11%, respectively. Amounts of unsaturated fatty acids were higher in the fruits (79.4 ± 0.11%) compared to flowers (73.9 ± 0.11%). Two essential fatty acids, linoleic acid (C18:2—48.1 ± 0.11%) and linolenic acid (C18:3—20.0 ± 0.08%), were in the highest quantity in oil from paulownia flowers, while in oil from paulownia fruits, linoleic acid (C18:2—53.9 ± 0.10%) and oleic acid (C18:1—19.5 ± 0.08%) dominated. The quantity of oleic acid (C18:1) in oil from fruits was five times higher (19.5 ± 0.08%) than that in the oil from flowers (3.6% ± 0.06%). Small amounts of essential icosapentaenoic acid (C20:5—0.2 ± 0.01%) and docosahexaenoic acid (C22:6—0.1 ± 0.01%) were also identified in oil from paulownia flowers. The glyceride oil of flowers was richer in saturated fatty acids: 26.1 ± 0.09% versus 20.6 ± 0.09% for oils from paulownia fruits. Among the saturated fatty acids, hexadecanoic acid (palmitic acid) was present in the highest amounts: 21.2 ± 0.09% (flowers) and 7.4 ± 0.08% (fruits). In oil from paulownia fruits, a relatively high content of medium- and long-chain saturated fatty acids was found: capric acid (C10:0—3.3 ± 0.03%), margaric acid (C17:0—2.9 ± 0.04%) and stearic acid (C 18:0—2.2 ± 0.09%), while in oil from flowers, only stearic acid was found at 2.4 ± 0.08%. The rest of the fatty acids were identified

in minimal quantities from 0.1 to 1.2%. Similar results were reported by Angelova-Romova et al. investigating lipid composition of paulownia seeds (fruits) [39]. The amount of oleic acid in their investigation was 21.2% (compared to $19.5 \pm 0.08\%$ in the present study), while the amount of linoleic acid was higher: 64.1% versus $53.9 \pm 0.10\%$. Schneiderová and Šmejkal found that triglycerides from paulownia possess potent biological activity and could play a role in deterring herbivores [38].

3.2. Antimicrobial Activity of Paulownia Fruit, Flower and Shell Extracts

The extracts were found to be rich in biologically active substances (phenolic acids, flavonoids, triglycerides, etc.), and for this reason, they were investigated for their antimicrobial activity against pathogenic and saprophytic microorganisms causing food spoilage. The results from the analyses are presented in Table 5.

Table 5. Antimicrobial activity of paulownia fruit, flower and shell extracts after 24 h of incubation.

Microorganism	cfu mL ⁻¹	Paulownia Fruits	Paulownia Flowers	Paulownia Shells	Negative Control (10% DMSO)	Amikacin	Nystatin
inhibition zone, mm							
Gram-negative bacteria							
<i>Escherichia coli</i> ATCC 25922	6×10^8	-	10.00 ± 0.00^{1c}	12.66 ± 0.47^b	-	25.80 ± 0.59^a	-
<i>Salmonella enteritidis</i> ATCC 13076	6×10^8	-	11.66 ± 0.47^c	16.00 ± 0.81^b	-	21.33 ± 0.94^a	-
<i>Pseudomonas aeruginosa</i> NBIMCC 1390	6×10^8	7.33 ± 0.47^c	10.00 ± 0.00^b	9.66 ± 0.47^b	-	24.93 ± 0.47^a	-
<i>Proteus vulgaris</i> ATCC 49132	6×10^8	10.00 ± 0.00^b	-	9.66 ± 0.47^b	-	24.80 ± 0.49^a	-
<i>Klebsiella pneumoniae</i> subsp. <i>pneumoniae</i> ATCC 10031	6×10^8	-	-	9.66 ± 0.47^b	-	25.10 ± 0.43^a	-
Gram-positive bacteria							
<i>Bacillus subtilis</i> ATCC 19659	6×10^8	7.66 ± 0.47^d	15.33 ± 0.47^c	17.33 ± 0.19^b	-	22.66 ± 1.25^a	-
<i>Bacillus cereus</i> ATCC 14579	6×10^8	9.00 ± 0.00^d	12.33 ± 0.47^b	9.66 ± 0.47^c	-	23.00 ± 0.82^a	-
<i>Staphylococcus aureus</i> ATC 6538P	6×10^8	14.33 ± 0.47^b	10.00 ± 0.00^c	9.66 ± 0.47^c	-	22.06 ± 0.90^a	-
<i>Enterococcus faecalis</i> ATCC 19433	6×10^8	10.00 ± 0.00^d	11.66 ± 0.47^c	14.16 ± 0.11^b	-	24.33 ± 1.25^a	-
Fungi/Molds							
<i>Candida albicans</i> ATCC 10231	6×10^8	-	10.66 ± 0.47^b	9.66 ± 0.47^b	-		26.16 ± 0.17^a

Table 5. Cont.

Microorganism	cfu mL ⁻¹	Paulownia Fruits	Paulownia Flowers	Paulownia Shells	Negative Control (10% DMSO)	Amikacin	Nystatin
<i>Aspergillus niger</i> ATCC 16888	1.6×10^7	8.00 ± 0.00^b	-	8.33 ± 0.47^b	-		24.60 ± 0.30^a
<i>Aspergillus flavus</i> ATCC 16883	3.2×10^7	7.00 ± 0.00^c	-	12.00 ± 0.00^b	-		24.80 ± 0.57^a
<i>Penicillium crysogenum</i> ATCC 10106	2.1×10^7	10.00 ± 0.00^b	-	10.00 ± 0.00^b	-		19.90 ± 0.65^a
<i>Aspergillus oryzae</i> ATCC 26831	1.1×10^7	7.00 ± 0.00^d	19.33 ± 0.47^b	9.00 ± 0.00^c	-		20.66 ± 1.25^a
<i>Fusarium moniliforme</i> ATCC 48843	2.1×10^7	-		14.00 ± 0.42^b	-		21.53 ± 0.75^a
<i>Mucor luteus</i> ATCC 28932	2.8×10^7	-		14.66 ± 0.22^b	-		20.80 ± 0.59^a

¹ Results are presented as the mean of three measurements; ^{a,b,c,d} Different letters in rows indicate statistically different values (Tukey's HSD test, $p < 0.05$).

In the case of antimicrobial activity against Gram-negative bacteria, the most pronounced effect was reported against *Escherichia coli* ATCC 25922 and *Salmonella enteritidis* ATCC 13076, in which the diameter of the inhibition zones varied approximately between 10.00 ± 0.00 mm and 16.00 ± 0.81 mm, depending on the sample and the incubation time. A less pronounced effect was observed for *Pseudomonas aeruginosa* NBIMCC 1390 and *Proteus vulgaris* ATCC 49132, in which the zones remained within the 9–10 mm range. For *Klebsiella pneumoniae* subsp. *pneumoniae* ATCC 10031, a weak inhibitory effect was observed in the tested samples. The obtained results indicate that extracts from paulownia flowers and shells inhibit the growth of Gram-negative bacteria, despite their generally higher resistance to external factors, which is attributed to differences in the chemical composition of their cell walls. Džugan et al. investigated the antimicrobial activity of paulownia leaf extracts against both Gram-positive and Gram-negative microorganisms [40]. Their findings regarding the effects of the extracts on Gram-positive bacteria are consistent with those obtained in the present study. However, in contrast to our results, their extracts did not exhibit any inhibitory effect against Gram-negative bacteria. Among the Gram-positive bacteria, large inhibition zones were also observed, depending on both the test microorganisms and the type of extract. *Bacillus subtilis* ATCC 19659 and *Enterococcus faecalis* ATCC 19433 exhibited the highest susceptibility to the paulownia shell extract, with inhibition zones exceeding 14 mm. The paulownia flower extract demonstrated the strongest antibacterial activity against *Bacillus subtilis* ATCC 19659, whereas the paulownia fruit extract exerted the greatest inhibitory effect on the growth of *Staphylococcus aureus* ATC 6538P. Yang et al. also observed a strong inhibitory effect of paulownia flower ethanolic extracts against *Bacillus subtilis* and *Staphylococcus aureus* [9]. The pronounced inhibitory activity against *Staphylococcus aureus* observed in the present study is also consistent with the finding of Škovranová et al., demonstrating that *P. tomentosa* fruit extracts and their geranylated flavonoids effectively inhibited the growth of *Staphylococcus aureus* by disrupting their bacterial membranes [41]. Aqueous extracts of fresh paulownia leaves were found to possess antimicrobial effects against *Staphylococcus aureus*, *Salmonella enterica*, *Pseudomonas aeruginosa*, *Paenibacillus alvei*,

Streptococcus pyogenes, and *Candida albicans*, and the inhibitory effect is more pronounced against Gram-negative bacteria [42,43]. From the molds, *Aspergillus oryzae* ATCC 26831 was the most susceptible to the action of the three extracts, with paulownia flowers showing the highest antimicrobial activity: inhibition zone 19.33 ± 0.47 mm. Paulownia shell 70% ethanol extract showed suppressive activity on all the investigated molds and the yeast *Candida albicans* ATCC 10231. The effect of the three extracts on the baker's yeast *Saccharomyces cerevisiae* ATCC 9763 was also investigated. After 24 h of incubation, no inhibition zones were observed, suggesting that the extracts are compatible with *Saccharomyces cerevisiae* and did not inhibit its growth during co-cultivation.

The negative control (10% DMSO solution in water) did not produce any inhibition zone, indicating that the solvent had no inhibitory effect on tested microorganisms.

3.3. Adsorption of Contaminants Using Paulownia Fruits, Flowers and Shells

3.3.1. Adsorption of Methyl Orange by Paulownia Fruits, Flowers and Shells

The physico-chemical composition of paulownia fruits, seeds and shells (Table 1) suggested they are rich in cellulose and lignin and could be potentially used as bioadsorbents. The amount of anhydrouronic acids, often associated with the presence of acidic biopolymers in the cell walls, in the flowers is slightly lower than 5%. Hence, in the next experiments, paulownia shells, fruits and flowers were investigated as potential bioadsorbents for removal of the azo dye methyl orange from water solutions, and the results for kinetics of the process are shown in Figure 1. For comparison, the adsorption abilities at the same conditions of three industrially available adsorbents (active carbon, chromatographic aluminum oxide and chromatographic silicagel) are also included.

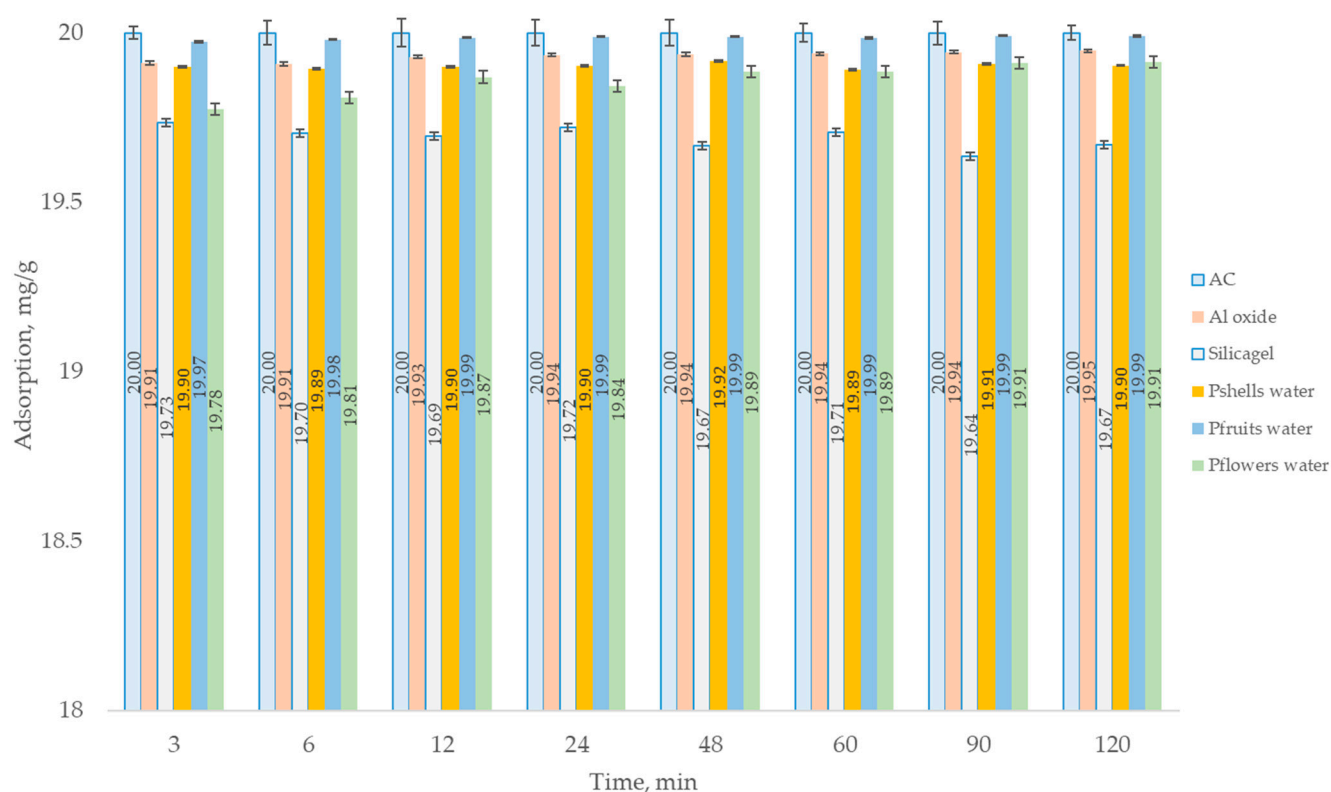


Figure 1. Kinetics of methyl orange adsorption. AC—active carbon; Al oxide— Al_2O_3 ; Silicagel—chromatographic silica gel; Pshells water—paulownia shells washed with deionized H_2O ; Pfruits water—paulownia fruits washed with deionized H_2O ; Pflowers water—paulownia flowers washed with deionized H_2O .

From the three water-washed paulownia materials, paulownia fruits showed the best methyl orange removal capacity (more than 95% after the 12th minute contact time and 98% after 90 min). The calculated adsorption capacity at these experimental conditions is 19 mg g^{-1} for paulownia fruits. These results are comparable with the adsorption ability of activated carbon, which is a well-known adsorbent for removal of various contaminants. The other two paulownia materials (shells and flowers) performed almost equally during adsorption and are comparable with the adsorption shown by chromatographic Al_2O_3 (Figure 1). The pH of the initial mixtures was 5.50 ± 0.06 , 5.54 ± 0.08 and 5.25 ± 0.10 for water-pretreated paulownia shells, fruits and flowers, respectively, and after the end of the adsorption process was: 5.84 ± 0.08 , 5.86 ± 0.12 and 5.67 ± 0.09 , respectively. Deniz and Saygideger investigated the adsorption of Acid Orange 52 (which has a similar structure to methyl orange) using *Paulownia tomentosa* leaf powder as a natural bioadsorbent [44]. The maximum amount of dye adsorbed at an initial concentration of 100 mg L^{-1} was 20.8 mg g^{-1} , while using 20 mg L^{-1} as the initial concentration, the adsorption capacity was only 0.7 mg g^{-1} adsorbent.

Paulownia shells are relatively underinvestigated, and not much data in the literature could be found. In an attempt, paulownia shells were treated in the presence of ZnCl_2 to obtain active carbon [45]. The experiments suggested that the methylene blue and phenol adsorption values can reach up to 300 mg g^{-1} and 451 mg g^{-1} , respectively, and the copper ions adsorption rate can reach 99%. Although the adsorption capacity was higher than that presented in the current work, it should be taken into account that the paulownia fruits, flowers and shells in the present study were used with minimum pretreatments. An additional advantage is that the spent adsorbents (with the methyl orange adsorbed into the lignocellulosic matrix) could be further decomposed and decontaminated using the potential of certain higher fungi to consume and degrade aromatic-type structures, including the azo dyes [46].

3.3.2. Adsorption of Cu^{2+} and Cd^{2+} by Paulownia Fruits, Flowers and Shells

Furthermore, the ability of paulownia shells, fruits and flowers to adsorb and concentrate metal cations was tested in a batch mode. Copper is considered both a pollutant (although it is necessary to be accepted in certain amounts) and a critical raw material important for the development of the economy and the green transition [47]. Recovering copper from low- and high-concentration solutions is important for the successful reuse of this valuable element. The results presented in Figure 2a revealed that paulownia fruits (water-pretreated) were effective in the removal of Cu^{2+} and comparable to active carbon and chromatographic Al_2O_3 . The adsorption capacity (at an initial concentration of Cu^{2+} at 5000 mg L^{-1}) was 98.25 mg g^{-1} . Paulownia shells showed relatively moderate capacity in copper uptake: around 20 mg g^{-1} adsorbent. The pH of the initial mixtures was 4.95 ± 0.11 , 5.72 ± 0.12 and 5.16 ± 0.12 for paulownia water-pretreated shells, fruits and flowers, respectively, and after the end of the adsorption process: 5.15 ± 0.09 , 5.95 ± 0.13 and 5.21 ± 0.10 , respectively.

Larous et al. [48] found that sawdust could adsorb copper cations (initial Cu^{2+} concentration 10 mg L^{-1}) up to 3.5 mg g^{-1} adsorbent. In our experiments (Figure 2b), paulownia shells (water-washed) were able to adsorb $19.91 \text{ mg Cu}^{2+} \text{ g}^{-1}$ adsorbent (initial Cu^{2+} concentration 1000 mg L^{-1}), which is more than four times higher than the adsorption capacity shown by Larous et al. [48]. The paulownia shells' (water-washed) adsorption ability was close to that of active carbon and aluminum oxide. Moreover, the influence of initial pretreatment of the paulownia shells (water, $0.1 \text{ M H}_2\text{SO}_4$ and 0.05 M NaOH) was investigated (Figure 2b). The shells (water-pretreated) showed the best adsorption potential, absorbing almost all of the copper cations from the solution. The pH of the

initial mixtures was 7.52 ± 0.13 , 2.73 ± 0.10 and 5.53 ± 0.09 for paulownia alkali-pretreated shells, acid-washed shells and water-washed shells, respectively, and after the end of the adsorption process, was 7.69 ± 0.10 , 2.79 ± 0.10 and 5.67 ± 0.08 , respectively. The worst adsorption capacity was shown by the acid-pretreated paulownia shells: 14.69 mg g^{-1} . These results are in accordance with the observations of Larous et al. [48] that strong acidic or alkaline conditions do not favor the adsorption process, and this could be due to the interaction of Cu^{2+} , $\text{Cu}(\text{OH})^+$ and $\text{Cu}(\text{OH})_2$ with the functional groups on the surface of the adsorbents.

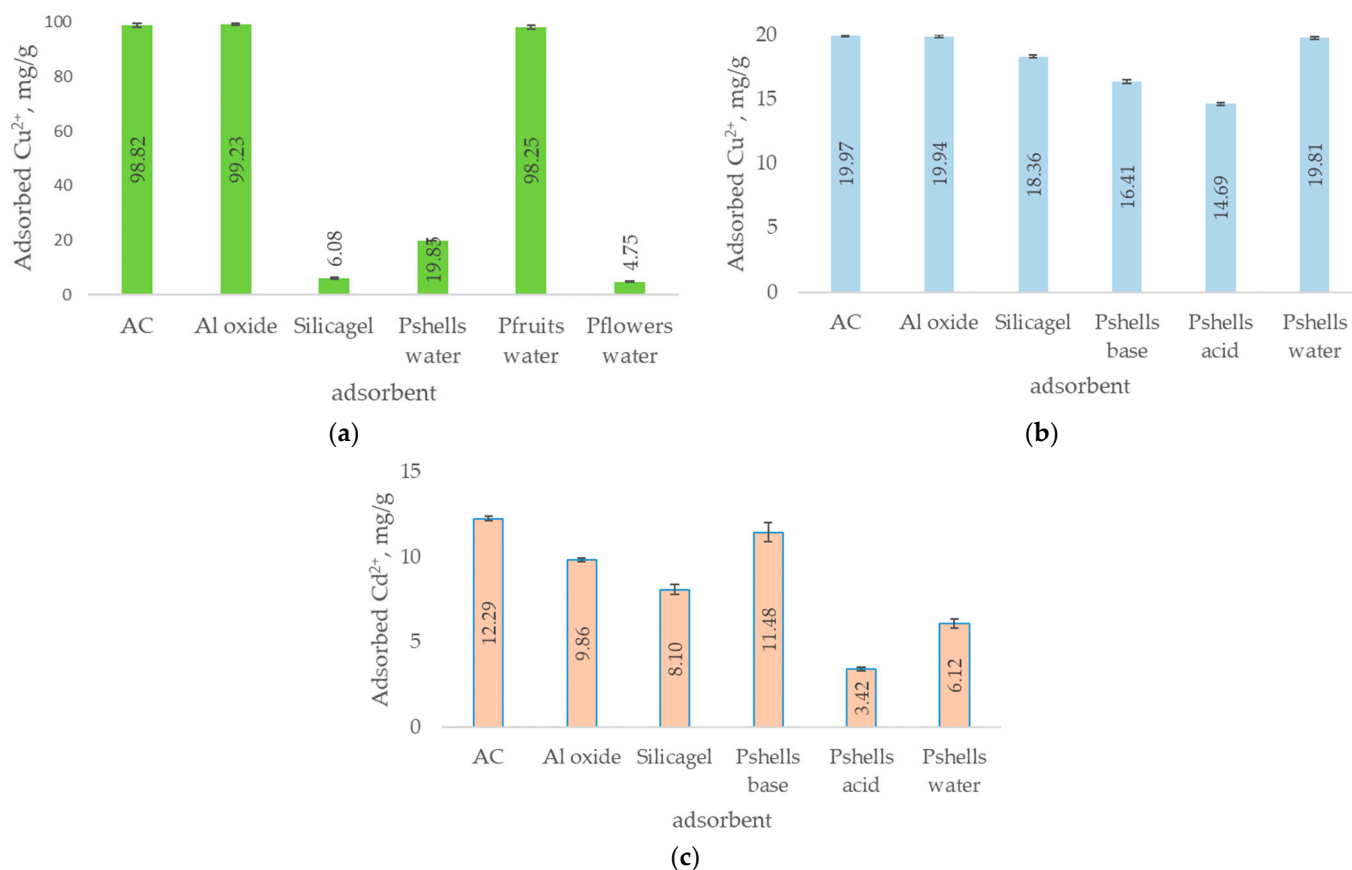


Figure 2. Adsorption of Cu^{2+} and Cd^{2+} from water solutions. (a) Initial Cu^{2+} concentration 5000 mg L^{-1} . AC—active carbon; Al oxide— Al_2O_3 ; Silicagel—chromatographic silica gel; Pshells water—paulownia shells washed with deionized H_2O ; Pfruits water—paulownia fruits washed with deionized H_2O ; Pflowers water—paulownia flowers washed with deionized H_2O ; (b) Initial Cu^{2+} concentration 1000 mg L^{-1} . AC—active carbon; Al oxide— Al_2O_3 ; Silicagel—chromatographic silica gel; base—paulownia shells washed with 0.05 M NaOH ; Pshells acid—paulownia shells washed with $0.1 \text{ M H}_2\text{SO}_4$; Pshells water—paulownia shells washed with deionized H_2O ; (c) Initial Cd^{2+} concentration 1000 mg L^{-1} . AC—active carbon; Al oxide— Al_2O_3 ; Silicagel—chromatographic silica gel; Pshells base—paulownia shells washed with 0.05 M NaOH ; Pshells acid—paulownia shells washed with $0.1 \text{ M H}_2\text{SO}_4$; Pshells water—paulownia shells washed with deionized H_2O .

Cadmium removal was performed using various natural or modified plant materials/by-products such as hazelnut shells, peanut hulls, walnut shells, green coconut shells, base-treated rice husk, treated juniper fibers, corncob modified with citric acid, modified peanut shells, succinic anhydride-treated sugarcane, etc. [21]. In our studies, paulownia shells (water-washed and pretreated with $0.1 \text{ M H}_2\text{SO}_4$ and 0.05 M NaOH) were employed for the adsorption of cadmium (at 613.5 mg L^{-1} initial concentration)—Figure 2c. The alkali-treated shells showed the best adsorption capacity: 11.48 mg g^{-1} adsorbent (higher than the result for chromatographic silica gel), while acid-washed shells showed the

lowest adsorption ability: only 3.42 mg g^{-1} adsorbent. Apparently, the free carboxyl groups of biopolymers present in the cell walls of paulownia might play a significant role in the adsorption process. The pH of the initial mixtures was 6.63 ± 0.09 , 2.41 ± 0.08 and 4.89 ± 0.11 for paulownia alkali-pretreated shells, acid-washed shells and water-washed shells, respectively, and after the end of the adsorption process was 6.15 ± 0.12 , 2.38 ± 0.11 and 4.62 ± 0.07 , respectively. Similar observations were made by Iqbal et al. [49] investigating adsorption of Pb^{2+} and Cd^{2+} by mango peel waste. They found that blocking the carboxyl and hydroxyl groups by chemical modification significantly reduces the adsorption capacity of the adsorbent. The reported adsorption capacity for Cd^{2+} removal was 16.55 mg g^{-1} adsorbent (initial Cd^{2+} concentration 50 mg L^{-1} , 2.5 g waste mango peel and pH 5.0). The lower pH suppresses Cd^{2+} adsorption, while pH in the 6–8 range was optimal. These observations are in accordance with the results of the present study. Bulut and Tez reported adsorption of cadmium by ground hazelnut and almond shells [50]. The observed adsorption capacities were 5.42 and 3.18 mg g^{-1} for hazelnut and almond shells, respectively. Krishnani et al. employed rice husks as an adsorbent for removal of heavy metals and observed an adsorption capacity of 14.4 mg g^{-1} (initial Cd^{2+} concentration 200 mg L^{-1} and pH = 6.0 ± 0.1) [51].

3.3.3. Infrared Spectroscopy of Paulownia Shells

In the next experiments, the paulownia shells (initial and pretreated with deionized water, $0.1 \text{ M H}_2\text{SO}_4$ and 0.05 M NaOH) were subjected to infrared spectroscopy (IR) analysis, and the spectra are presented in Figure 3. The spectra comparison suggests that the alkali pretreatment increases the exposed $-\text{OH}$ groups (the band around 3447 cm^{-1}), which might be the reason for the increased adsorption of Cd^{2+} . Alkali pretreatment usually exposes more $-\text{OH}$ groups, mainly from cellulose, by decreasing masking surface components like lignin and hemicelluloses. Based on earlier observations [40,52], specific main ranges of wave numbers can be assigned to the main cell wall biopolymers: $2840\text{--}2870 \text{ cm}^{-1}$ to proteins (peptide bonds) and $3270\text{--}3290 \text{ cm}^{-1}$ to carbohydrates.

Several peaks in the $1200\text{--}1700 \text{ cm}^{-1}$ region could be distinguished and differentiate the biomass components as follows: 1623 to 1635 cm^{-1} could be associated with adsorbed water or aromatic $\text{C}=\text{C}$ stretching from lignin moiety; around 1458 cm^{-1} , C-H asymmetric bending in lignin and carbohydrates; 1260 cm^{-1} , C-O stretching in lignin (guaiacyl ring) [53].

The peak near 895 cm^{-1} is a classic marker for the presence of β -glycosidic linkages between glucose units in cellulose. The sharp, variable peaks appearing around 669 cm^{-1} (especially prominent in the acid/water pretreated paulownia shells) are typically due to C-H out-of-plane bending [54].

Alkali pretreatment led to the most significant changes in the lignocellulosic paulownia shells matrix. It enhanced overall absorption intensity across almost all functional bands (specifically the $-\text{OH}$ band around 3447 cm^{-1} and the band around 1027 cm^{-1} , which in general could be associated with carbohydrates (C-O and C-O-C stretching vibrations) present in the shells. Larous et al. found that the adsorption of copper using sawdust was most effective in the pH range 2–8, with 6 being the most favored, and the presence of $-\text{OH}$ groups contributed significantly to the process [48].

Acid and water pretreatment led to very similar structural changes compared to the initial paulownia shells (orange line), but still exhibited slight modifications in peak depths, indicating partial hydrolysis or washing away of water-soluble substances from the surface without significantly altering the bulk lignocellulosic structure.

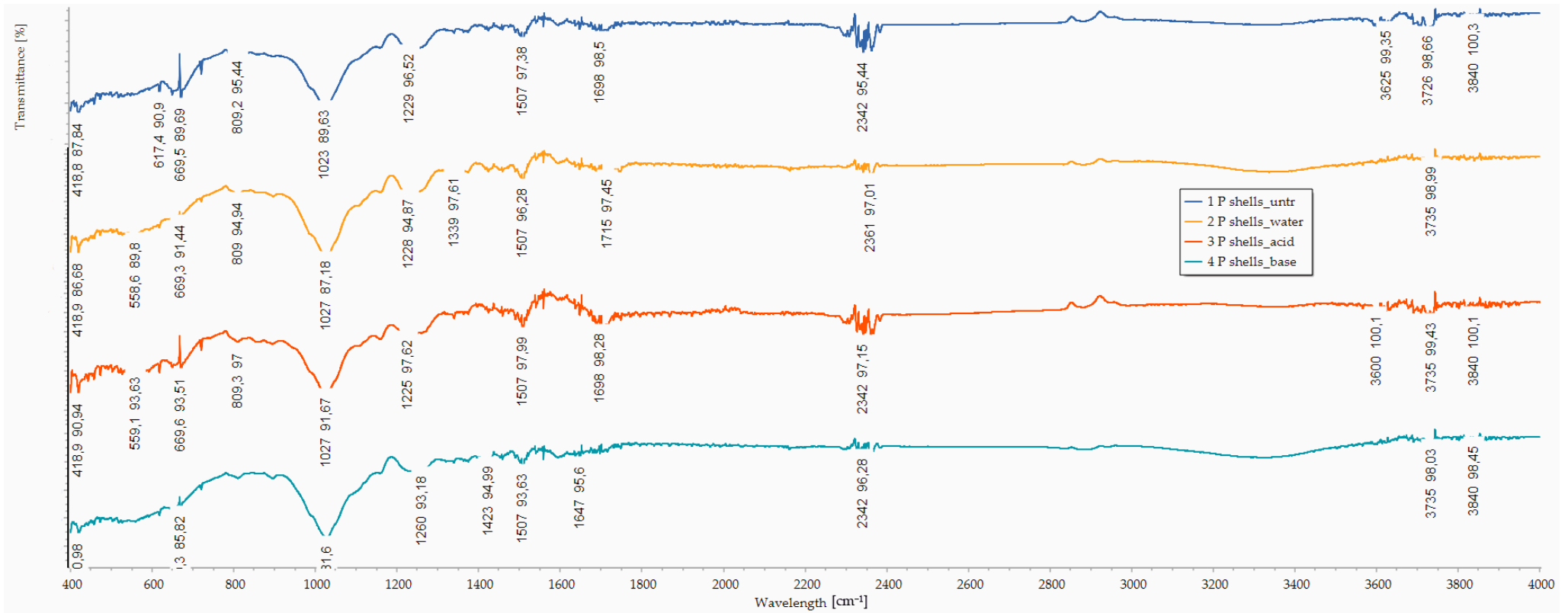


Figure 3. Cont.

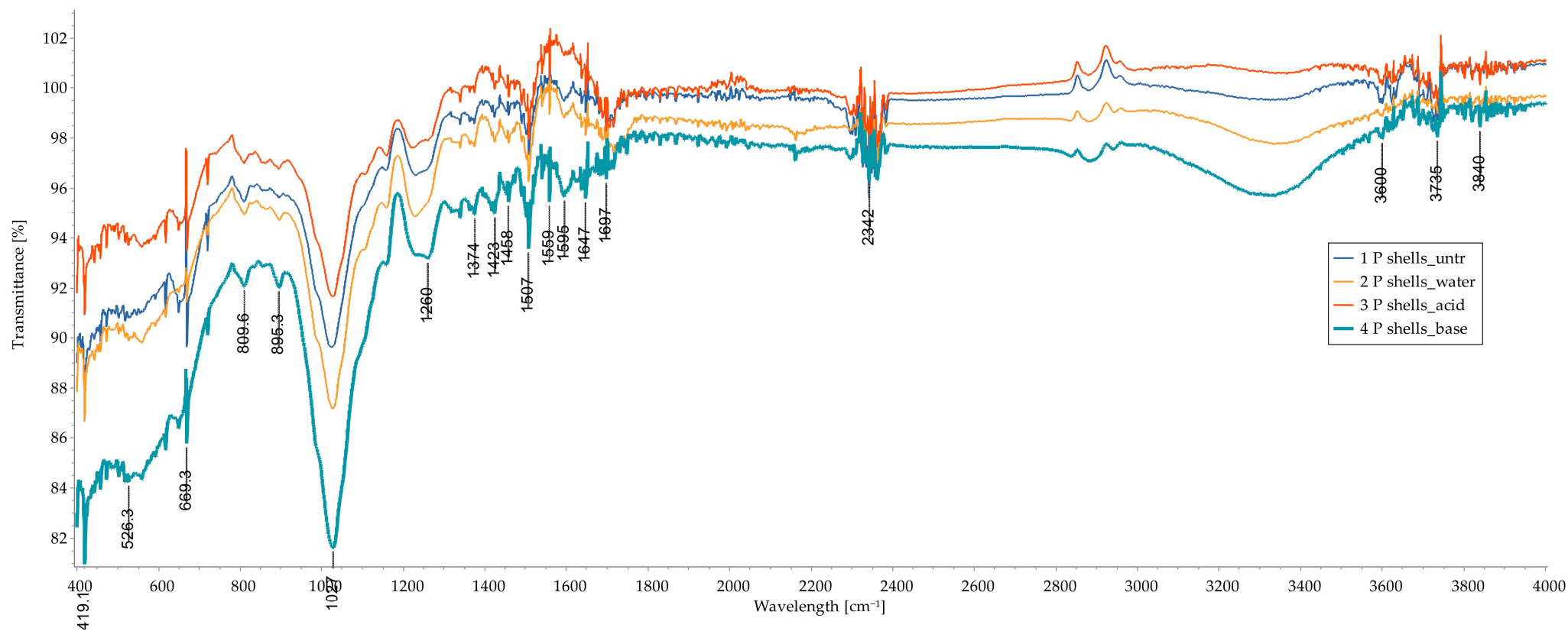


Figure 3. IR spectra of: P shells_untr—paulownia shells untreated; P shells_water—paulownia shells washed with deionized H₂O; P shells_acid—paulownia shells washed with 0.1 M H₂SO₄; P shells_base—paulownia shells washed with 0.05 M NaOH.

4. Conclusions

The growing worldwide demand for wood has led to the cultivation of fast-growing trees, such as paulownia, an important and treasured species, and has led to a constant accumulation of non-timber waste materials. Paulownia flowers, fruits and shells are among these seasonal by-products that can be successfully valorized and better utilized. For centuries, paulownia flowers and fruits have been used in folk medicine and for culinary purposes. Nowadays, flowers are treasured for their significant use in products such as food, animal feed, and cosmetics. Paulownia fruits and seed shells are rich in lignocellulosic biomass and have the potential to be used as natural and renewable bioadsorbents.

The present study demonstrated that different morphological parts of *Paulownia tomentosa*—specifically fruits, flowers, and shells—possess significant chemical and antimicrobial value. The physico-chemical analyses revealed that these components are rich sources of diverse bioactive metabolites, such as phenolic acids, flavonoids, and unsaturated fatty acids, while simultaneously offering highly effective structural configurations for wastewater treatment. The fruits and flowers contain substantial levels of total dietary fiber ($54.30 \pm 2.10\%$ and $59.88 \pm 1.91\%$, respectively) and valuable lipid fractions dominated by polyunsaturated fatty acids ($55.6 \pm 0.11\%$ in fruits and $69.2 \pm 0.11\%$ in flowers). The fruit 70% ethanol extract exhibited the highest flavonoid content ($842.43 \mu\text{g mL}^{-1}$), which directly correlated to an antioxidant capacity twice as high as that of the flower extract measured by DPPH ($44.59 \pm 0.44 \text{ mmol TE g}^{-1}$ dry extract), ABTS ($47.50 \pm 1.05 \text{ mmol TE g}^{-1}$ dry extract), FRAP ($74.64 \pm 3.40 \text{ mmol TE g}^{-1}$ dry extract), and CUPRAC ($114.12 \pm 2.11 \text{ mmol TE g}^{-1}$ dry extract) assays. In contrast, the flowers are uniquely characterized by exceptionally high concentrations of salicylic acid ($2729.96 \pm 16.89 \mu\text{g mL}^{-1}$). The 70% ethanol extracts of all three plant parts showed potent antimicrobial activity against some food-spoilage pathogens and saprophytic microorganisms, while showing no harmful influence on beneficial baker's yeast (*Saccharomyces cerevisiae*). The antimicrobial activity of *P. tomentosa* extracts varies significantly between bacterial categories, with Gram-positive bacteria demonstrating much higher susceptibility than Gram-negative bacteria. Extracts from fruits and flowers produced larger inhibition zones against Gram-positive strains like *Staphylococcus aureus* and *Bacillus cereus*. Their cell walls lack an outer membrane, allowing the plant's hydrophobic bioactive compounds (such as flavonoids and phenolic acids) to easily disrupt the cell membrane. Strains like *Escherichia coli* and *Salmonella enterica* showed much smaller inhibition zones or complete resistance. Their outer lipopolysaccharide membrane acts as an effective permeability barrier, blocking the penetration of the active antimicrobial metabolites. Preliminary adsorption studies suggested *P. tomentosa* by-products could be applied as promising, renewable, and low-cost natural biosorbents. Due to their rich cellulose and lignin content, they exhibited capabilities (although for some of them moderate) in removing organic and inorganic pollutants with minimal pretreatment. Specifically, the fruits were able to remove the azo dye methyl orange, while pretreated flowers and shells showed moderate to good absorbability of copper and cadmium cations (even at high initial concentrations). Alkali-washing pretreatment, tentatively exposing some functional chemical groups (carboxyl, hydroxyl, etc.) of shell cell wall biopolymers, showed the highest cadmium adsorption ability. These preliminary screening adsorption experiments using model solutions suggested that paulownia by-products need further examination of the potential of paulownia morphological parts (by-products) as adsorbents for the removal of pollutants. With the utilization of paulownia resources, new products and technologies based on by-products with economic value are continuously promoted, and the interest in creating and developing new applications of paulownia is increasing.

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Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

ABTS	2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid)
DPPH	2,2-diphenyl-1-picrylhydrazyl radical
FRAP	Ferric-reducing antioxidant power
CUPRAC	Cupric-reducing antioxidant capacity
DMSO	Dimethylsulfoxide
HPLC	High-performance liquid chromatography

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