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Research Article

Removal of 2-naphtol orange by industrial rose and lavender solid by-products in dynamic conditions

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Abstract

Rosa damascena Mill. (RD) and *Lavandula angustifolia* Mill. (L) biomasses – by-products of essential oil manufacturing were utilized as renewable and nature-friendly adsorbents for removal of 2-naphthol orange (2NO) in column adsorption model (dynamic conditions). The amount of proteins in the biomasses were 11.60±0.68% for RD and 8.65±0.20 for L, and the polyuronide content for both residues was around 7.50%. The dynamic adsorption of 2NO was investigated in five solutions with different pH, namely 0.1 N H_2SO_4 (1.21), deionized water (3.63), 50 mM citrate buffer (5.49), 50 mM phosphate buffer (8.30) and 0.05 N NaOH (12.80). Preliminary washing of RD and L residues with 0.1 N HCl increased their removal efficiency compared with water washed biomasses. At pH range from 1.21 to 5.49 the 2NO removal was above 89.0±0.1% for RD and 84.2±0.1% for L. The present work demonstrated that RD and L biomasses were able to remove 2NO in dynamic conditions (column adsorption) from water solutions with 89.0±0.1% and 84.2±0.1% efficiency, respectively.

Keywords

2-naphtol orange, rose (*Rosa damascene* Mill.), lavender (*Lavandula angustifolia* Mill.), column adsorption, dynamic adsorption

Abbreviations

RD – *Rosa damascena* Mill.; L – *Lavandula angustifolia* Mill.; 2NO – 2-naphthol orange

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Introduction

The essential oil manufacturing is emblematic and widely represented in Bulgaria. Essential oil and medicinal plants from numerous families (Rosaceae, Lamiaceae, Asteraceae, etc.) have been grown and processed. Bulgaria, Turkey, France and Iran produce and trade more than 80% of the rose oil worldwide (Kovacheva et al. 2010; Stoyanova 2022). Lavender (*Lavandula angustifolia* Mill.), besides the Damascus rose, is the second grown up essential oil variety in Bulgaria. The amounts of essential oil in the fresh raw plants depending from the climate conditions, crop and cultivar is usually below 0.05% for *Rosa damascena* Mill., and below 1.5% for *Lavandula angustifolia* Mill., which implies that after industrial obtaining of the oil large amounts of waste biomass are produced. In most cases, distilleries dispose of this biomass in the nearby surroundings, where it spontaneously ferments but these waste materials could be reused and valorised (Kovacheva et al. 2010).

Azo dyes are commonly used organic pigments with wide applications in various industries (Rathi and Kumar 2022). 2-Naphthol orange (2NO) is used for dyeing textile, paper goods, printing toners, leather handiworks, hairs cosmetics, etc. Crettaz et al. (2020). After dying process the waste dye solutions should be purified from all the dissolved chemicals before water could be safely disposed. Numerous processes, such as: microbial fermentation, oxidation, and adsorption were utilized for 2NO removal from waste waters. The adsorption is among the most utilized methods. Various materials were used as adsorbents (Khalil et al. 2024; Nistor et al. 2022) and waste/by-products (resulted from electricity generation, agriculture, timber, food, etc.) showed promising potential (Amalina et al. 2022; Kaya and Şahin 2022; Marovska et al. 2023; Naraghi et al. 2017; Slavov et al. 2023). Two main ways of carrying out the process can be distinguished – adsorption under static conditions (when the adsorbent is placed in a suitable vessel and the contaminated solution is added; stirring is often used to intensify the adsorption) (Ouakouak et al. 2021) and under dynamic conditions (when the adsorbent is placed in a suitable vessel (most often a column) and the contaminated solution is passed through it by gravity or with the help of a pump) (Khan et al. 2022). Adsorption under static conditions is easier to perform, but a disadvantage is the serial operation mode. Adsorption under dynamic conditions is performed in a continuous mode, periodically changing the adsorbent after reaching its capacity to retain the target pollutant. Lavender and rose biomasses were employed in several studies for heavy metals removal from waste waters (Slavov et al. 2017) but application for organic pollutants removal are scarce (Echavarria-Alvarez and Hormaza-Anaguano 2014; Rabbani et al. 2016). To the best of our knowledge there are no data for adsorption of organic pollutants by plant by-products performed by column adsorption process. Hence, the present work aimed on investigations for utilization of RD and L biomasses as adsorbents for 2NO removal from water solutions in dynamic conditions (column adsorption).

Materials and Methods

Materials. The bio-certified rose (*Rosa damascena* Mill.) and lavender (*Lavandula angustifolia* Mill.) biomasses, by-products resulted from industrial steam-water and steam distillation, respectively, were obtained from distillery of Mirkovo (Sofia region, Bulgaria, 2021). 2NO (4-(2-Hydroxy-1-naphthylazo) benzenesulfonic acid sodium salt) was obtained from Merck (Germany). A chromatography glass column with bottom glassy porous support and PTFE stopcock (20×400 mm, NS 29/32; Isolab, Germany) was used for dynamic adsorption.

Methods. The residues were roughly chopped using garden shredder and then finely milled with a laboratory grain-milling apparatus. The biomasses were washed with deionized water and 0.1 N HCl as described (Marovska and Slavov 2023).

Dynamic adsorption was carried out in a 400 x 20 mm column, NS 29/32 (Isolab, Germany), equipped with a porous barrier at the lower end and a Teflon stopcock. Five grams of adsorbent were placed in the column and porous material from glass crucible was used at the upper end to prevent the adsorbent layer from being disturbed during the addition of the liquid phase. After the addition of 100 mL of the dye solution, half an hour was waited (to wet the entire amount of adsorbent) and fractions of 4 ml were collected in 5 ml Eppendorf tubes by opening the stopcock until the entire solution had passed through the adsorbent. The collected fractions were filtered first through a paper filter and then through a CA 0.45 µm syringe filter (Isolab, Germany). The adsorption of the filtrate was measured at 500 nm using LLG-uniSPEC 2 UV-Vis spectrophotometer (LLG Labware, Germany) and the concentration of 2NO remaining after adsorption was calculated using a standard curve prepared with solutions of the dye of known concentrations.

The proteins' amount was assessed by the method of Kjeldahl (MultiKjel K-365 – Büchi, Switzerland). The polyuronide content (PC) and degree of esterification (DE) of the biomasses were determined as described by Marovska and Slavov (2023). The ash was determined by heating samples (placed in a crucibles) at 625°C in a laboratory muffle furnace and measuring the weight of the remaining mass in the crucibles until constant weight achieved. The moisture content was determined with KERN DAB 100-3 analytical balance (KERN&SOHN GmbH, Germany).

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

Results and Discussion

Preliminary characteristics of the RD and L residues. The protein quantities of the RD were 11.60±0.68% and in the L biomass – 8.65±0.20% (Table 1). The L biomass (5.96±0.27%) had two times higher ash content than RD (2.84±0.65%). Tentatively, this observation could be due the type of distillation applied – the RD flowers were steam-water distilled and this process because using large amounts of water could remove part of the inorganic matter. Besides, for obtaining rose oil only the rose flowers were used and for lavender oil the whole plant is steam distilled. The PC, usually associated with pectic polysaccharides in the plant cell walls, for both biomasses were similar (no significant difference): 7.81±0.33% for L and 7.56±0.28% for RD. Table 2 presents information from the mesh size analyses of the biomasses. The dominating fractions were between 100 and 250 µm: more than 70% for both milled biomasses.

Table 1. General characteristics of RD and L biomass: protein, ash, and moisture content, degree of esterification (DE) and polyuronide content (PC)

Biomass	Protein, %	Ash, %	Moisture, %	DE, %	PC, %
Rose (*Rosa damascene* Mill.), RD	$11.60^{a} \pm 0.68$	$2.84^{a} \pm 0.65$	$10.31^{a} \pm 0.12$	$78.36^{a} \pm 1.34$	$7.56^{b} \pm 0.28$
Lavender (*Lavandula angustifolia* Mill.), L	$8.65^{b} \pm 0.20$	$5.96^{b} \pm 0.27$	$9.47^{a} \pm 0.14$	$74.60^{b} \pm 1.21$	$7.81^{a} \pm 0.33$

*Results are presented as the mean of three measurements;
[a,b] - different letters in columns indicate statistically different values (Tuckey's HSD test, $p < 0.05$)

Table 2. Particle size analysis of RD and L biomasses

Mesh size	Rose (*Rosa damascene* Mill.), RD		Lavender (*Lavandula angustifolia* Mill.), L	
	Fraction weight, g	Fraction percentage, %	Fraction weight, g	Fraction percentage, %
>1 mm	$0.14^{f} \pm 0.01$	$0.15^{f} \pm 0.01$	$0.10^{f} \pm 0.01$	$0.14^{f} \pm 0.01$
710 µm	$1.05^{e} \pm 0.04$	$1.15^{e} \pm 0.04$	$0.38^{e} \pm 0.02$	$0.53^{e} \pm 0.03$
500 µm	$8.16^{d} \pm 0.09$	$8.95^{d} \pm 0.10$	$1.76^{d} \pm 0.15$	$2.47^{d} \pm 0.16$
250 µm	$35.81^{a} \pm 0.07$	$39.30^{a} \pm 0.07$	$18.96^{b} \pm 0.09$	$26.63^{b} \pm 0.09$
100 µm	$29.96^{b} \pm 0.16$	$32.92^{b} \pm 0.15$	$33.05^{a} \pm 0.16$	$46.42^{a} \pm 0.16$
<100 µm	$16.00^{c} \pm 0.17$	$17.56^{c} \pm 0.16$	$16.94^{c} \pm 0.17$	$23.79^{c} \pm 0.16$
Total	91.12 ± 0.24	100	71.19 ± 0.14	100

*Results are presented as the mean of three measurements;
[a,b,c,d,e,f] - Different letters in columns indicate statistically different values (Tuckey's HSD test, $p < 0.05$)

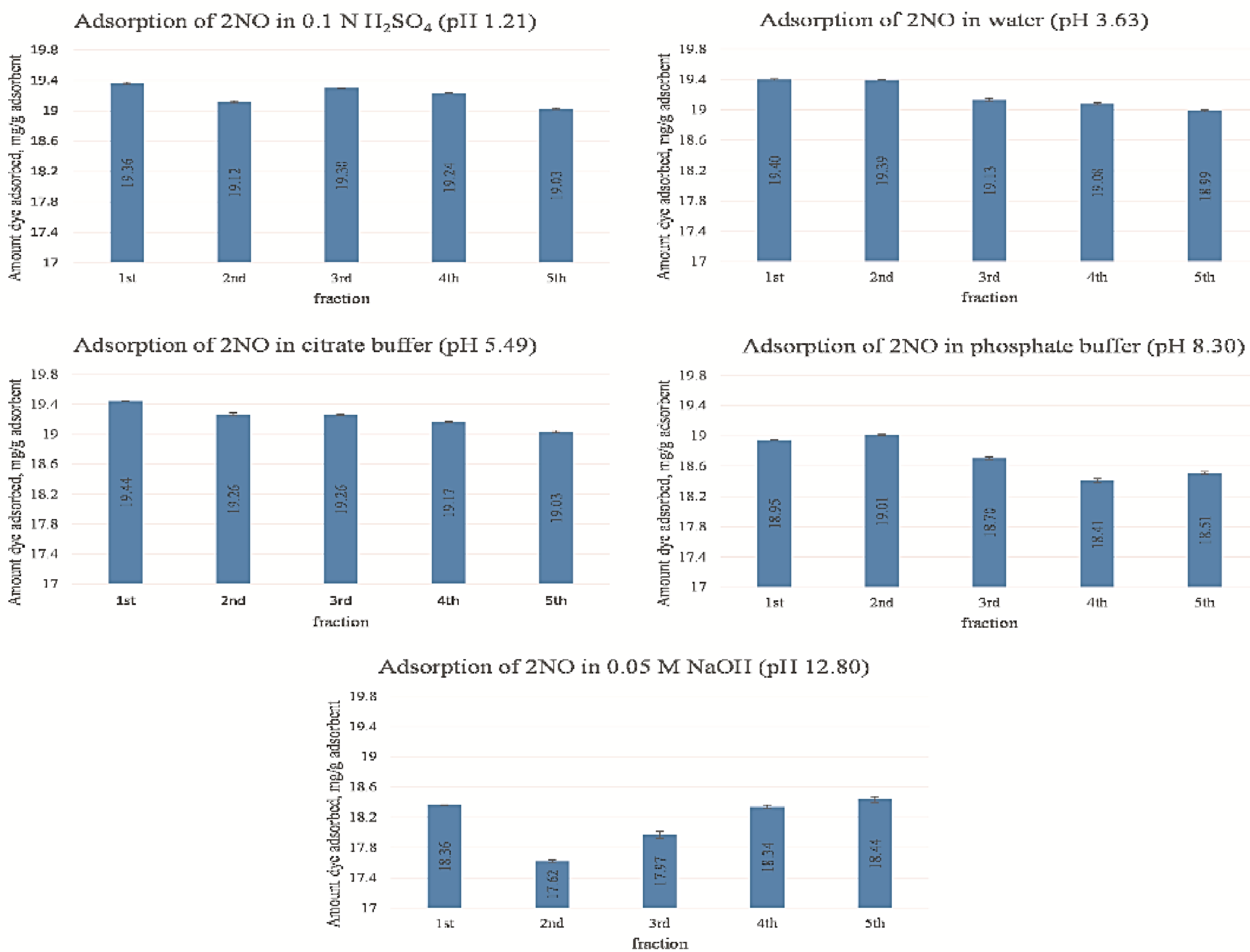

Figure 1. Contact time and pH influence on 2NO removal (column adsorption) by RD biomass pretreated with deionized water

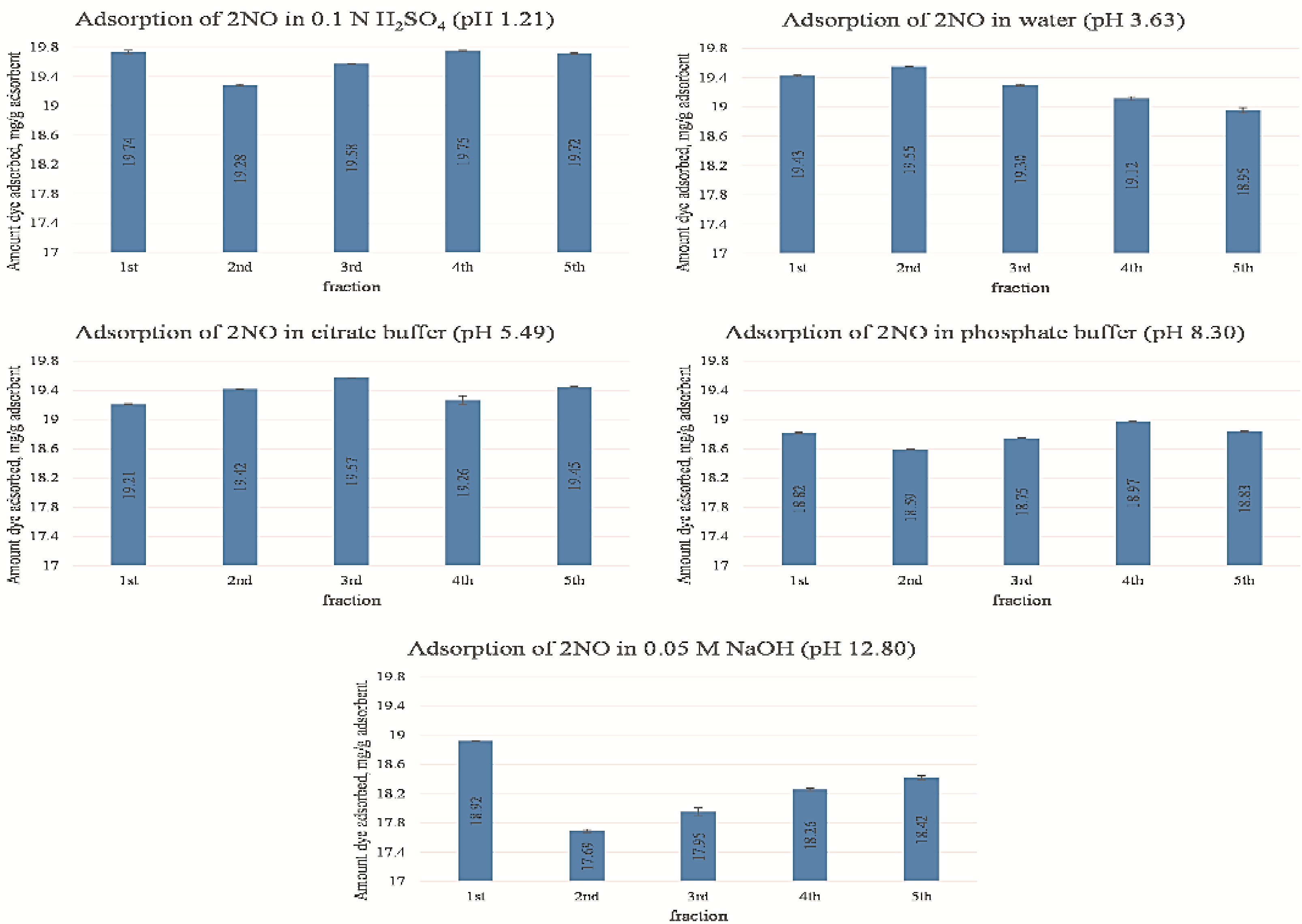

Figure 2. Contact time and pH influence on 2NO removal (column adsorption) by L biomass pretreated with deionized water

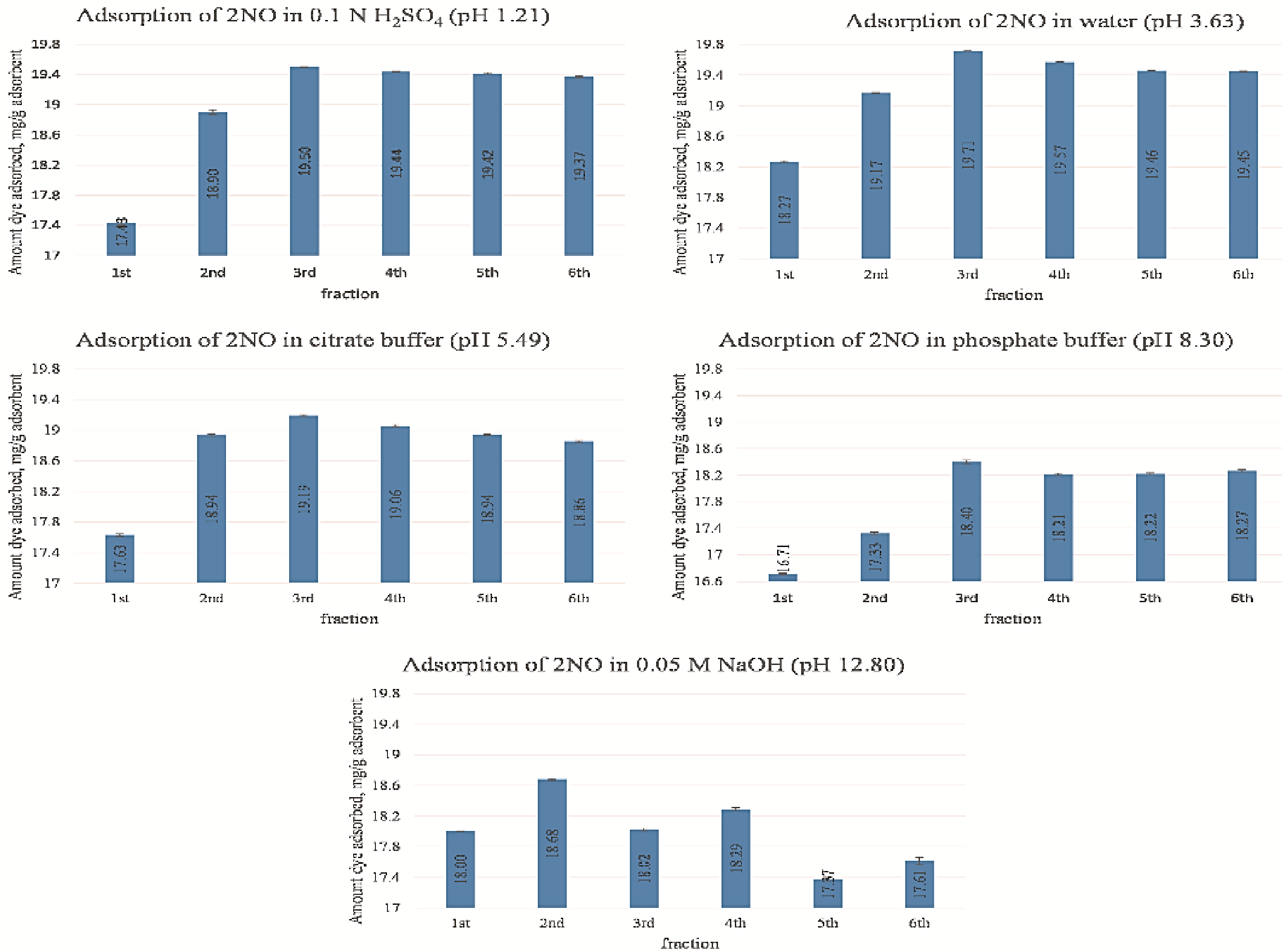

Figure 3. Contact time and pH influence on 2NO removal (column adsorption) by RD biomass pretreated with 0.1 N HCl

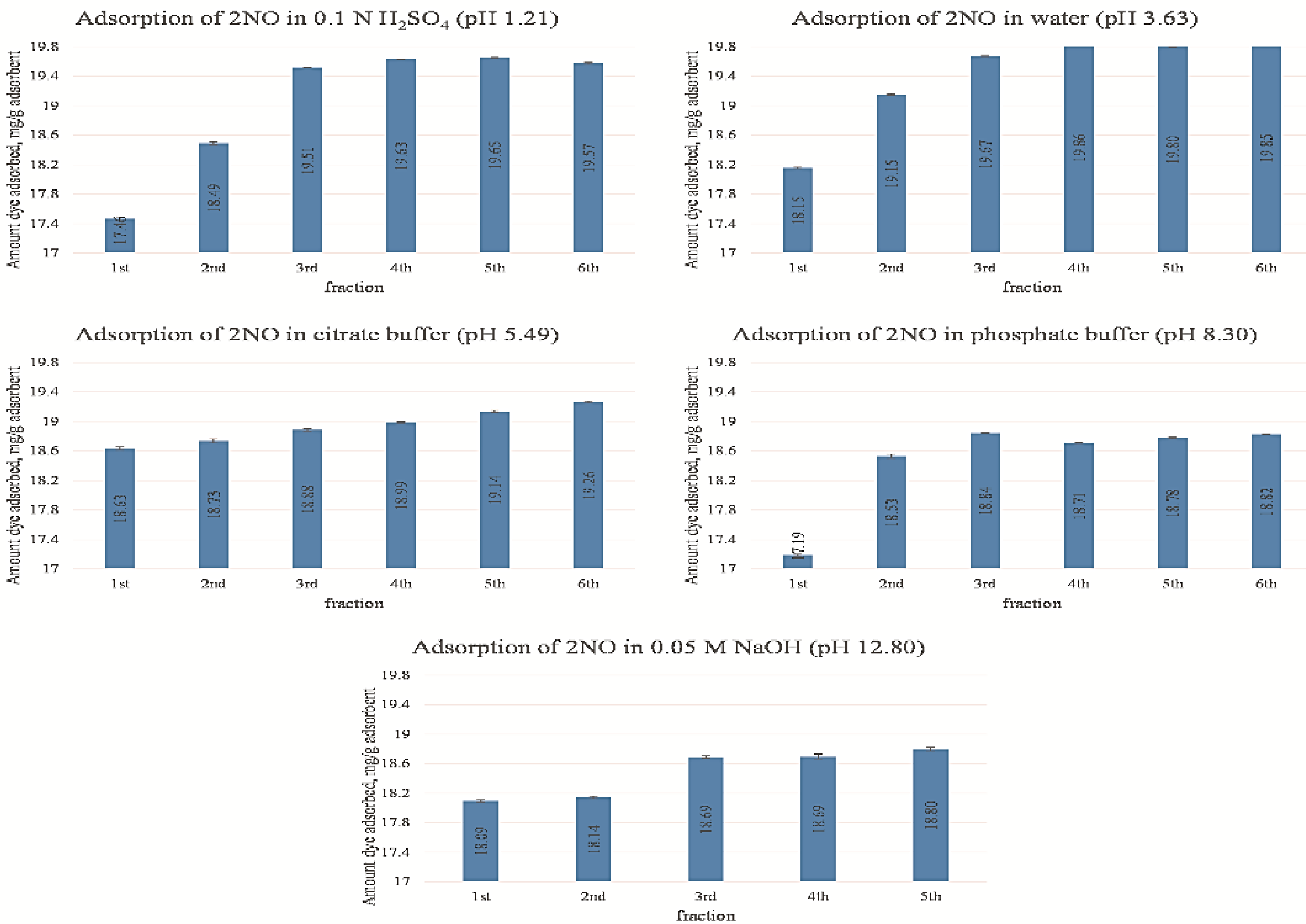

Figure 4. Contact time and pH influence on 2NO removal (column adsorption) by L biomass pretreated with 0.1 N HCl

Contact time and pH influence on 2NO adsorption with RD and L residues pretreated with deionized water. The RD and L residues were washed with deionized water. This treatment was performed to remove low-molecular substances present in the plant matter.

Adsorption of 2NO was performed in five solvents with different pH (from highly acidic – 1.21 to high basic pH – 12.80). First as adsorbents were used water-washed residues. The experimental results are presented in Fig. 1 for RD and Fig. 2 for L biomass. The process relies on physical and chemical interactions of the biomasses and the dye. The predominating functional groups in the adsorbent and the compounds adsorbed play vital role and determined the effectiveness of the process. These interactions are stronger and adsorption is fast in the beginning (first collected fractions) because the unoccupied groups on the surface of the adsorbent predominates (Marovska et al. 2023). The data clearly showed that lower pH (5 or less) increase adsorption effectiveness for RD biomass. The highest percentage removed 2NO was observed at lower pH (1.21) – almost 92% for RD and around 81% for L. The effectiveness of RD as adsorbent was kept in the range of pH from 1.21 to 8.30 which could be seen as constantly lowering the values with increasing the fraction number. At higher pH (12.80) the effectiveness was diminished and this tendency was not observed. At this pH also the percentage of the absorbed 2NO was the lowest (less than 45% for both RD and L biomasses). Similar observations and conclusions were made in the studies (Hambarliyska et al. 2025; Marovska et al. 2023; Wu et al. 2011) focusing on β-naphtol orange removal in static conditions using rose and lavender (residues from industry for essential oil production), brewery spent yeasts, and melissa and yarrow wastes (also from industry for essential oil production), respectively. Collection of fractions beyond 5th and 6th showed that the processes of adsorption/desorption are entering equilibrium and it is difficult to obtain better removal of the 2NO.

Contact time and pH influence on 2NO adsorption with RD and L residues pretreated with deionized water. In the subsequent experiments the RD and L residues were washed with 0.1 N HCl. The acidic pretreatment serves for activation of functional groups of biopolymers from the plant matrix, i.e. protonation of carboxylic acids and quarterisation of amino groups. The second transformation of non-charged to positively charged groups is important, since the 2NO is also charged compound due to presence of sulfo groups, bearing negative charge.

The adsorption process in these cases was different from previous experiments (Fig. 3 and Fig. 4). In the beginning the adsorption rate was lower but with advancement of the fraction collection (after the second fraction) was clear that effectiveness of the adsorption increased compared to RD and L biomasses pretreated with deionized water. This tendency was kept for RD till pH 5.49 and for L – even in the highest pH (12.80). The observed results could be tentatively explained with predominating physical character of adsorption of the dye onto the biomasses and the time necessary in the begging for increasing the effectiveness. The data, however, suggested that biomasses pretreated with hydrochloric acid showed better adsorption effectiveness (95% for RD and 89% for L) than RD and L washed with water – 92% and 81%, respectively. Similar observations were reported by Hambarliyska et al. (2025) investigating removal of β-naphtol orange with melissa and yarrow residues and Marovska and Slavov (2023) investigating adsorption of β-naphtol orange using rose and lavender biomasses in static conditions.

Conclusions

The present investigation demonstrated removal of the azo dye 2NO in dynamic conditions (column adsorption) with two industrial essential oil biomasses – *Rosa damascena* Mill. and *Lavandula angustifolia* Mill. Residues pretreated with 0.1 N HCl demonstrated better adsorption capacity for 2NO removal but the process was slower in the beginning of the adsorption. A possible explanation of this data is protonation of the free carboxyl groups of polyuronic acid biopolymers present in the plant matrix and lowering the repulsion forces bearing in mind that the 2NO has charged sulfo groups. The equilibrium of adsorption/desorption was observed after collection of the 5th and 6th fraction. The pH is important factor influencing the adsorption effectiveness and below pH 5 higher efficiency was observed. The most optimal conditions for 2NO removal could be summarized as follow: pH below 5, initial pretreatment of the plant materials with dilute acids, and contact time at least 30 min.

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Author Contributions

Conceptualization, I.H., A.S.; methodology, A.S., M.T. and I.H.; formal analysis, A.S., I.H.; investigation, A.B., A.S., M.T. and I.H.; resources, A.S. and I.H.; data curation, A.S, I.H. and M.T.; writing - original draft preparation, A.S. and I.H.; writing - A.S., I.H. and A.B., review and editing, A.S., and I.H., visualization, A.S. and M.T.; supervision, A.S. and I.H.; project administration, A.S; funding acquisition, A.S. All authors have read and agreed to the published version of the manuscript.

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Institutional Review Board Statement

The experimental protocol used in this study was designed in compliance with the guidelines of the European and Bulgarian legislation regarding the protection of animals used for experimental and other scientific purposes (Directive 2010/63; EC, 2010 - put into law in Bulgaria with Regulation 20/2012). The protocol was based on the permit for the use of animals in experiments No. 277 of the Bulgarian Food Safety Agency (Statement No. 193 of the Bulgarian Animal Ethics Committee, prot.No.18/02.07.2020, 2 July 2020).

Informed Consent Statement

Not applicable.

Data Availability Statement

The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding authors.

Conflicts of Interest

The authors declare no conflicts of interest.

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