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Removal of methylene blue azo dye from aqueous solution using biosorbent developed from floral waste

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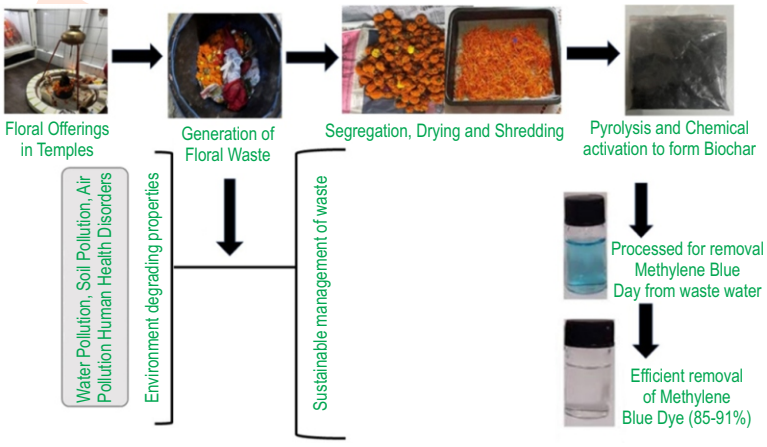
Abstract

Aim: The present study was carried out to prepare biosorbent from temple floral waste (*Tagetes erecta*) by pyrolysis and chemical activation method for removal of methylene blue dye from aqueous solution.

Methodology: Floral waste of *Tagetes erecta* collected from the temples were segregated, washed and dried to form biochar by direct pyrolysis and chemical activation method. Followed by physio-chemical analysis of biosorbents the most efficient biochar was selected for the removal of methylene blue dye from aqueous solution. The adsorbent efficiency and percentage removal of methylene blue dye was studied using various doses of biochar (10, 20, 30, 40, 50, 60, and 70 mg 100 ml^{-1},), effect of pH (2.0 to 4.0, 6.0 to 8.0, and 10.0 to 12.0) and effect of contact time etc.



Results: The comparative physio-chemical analysis of the bio chars suggested that the activated charcoal made from temple flower waste by the direct pyrolysis method showed better performance, with its low moisture content (5.3%), low ash content (4.3%), higher yield, larger surface area, and higher porosity (65.3%) as compared to the biochar obtained from chemical activation. The percent adsorption significantly increased ($p<0.05$) from 76% to 87.0% on increasing biochar dose from 10.0 to 70.0 mg 100 ml^{-1}. On increasing the pH of the solution from 4.0 to 6.0, Methylene blue removal significantly increased ($p<0.05$) from 88.0% to 91.0%.

Interpretation: It is possible to manage floral waste from temples in a sustainable and environmentally responsible manner by converting it into biochar and using it for the treatment of waste water in order to eliminate hazardous dyes.

Key words: Activated carbon, Azo dye, Bioremediation, Biochar, Floral waste, Methylene blue

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Introduction

Water is a crucial component for human existence and is known as the "elixir of life". However, contamination of water has increased tremendously due to industrialisation followed by urbanisation. Anastopoulos *et al.* (2019) reported that various colouring industries release effluent containing dyes, which in turn has become a big challenge as they contaminate water bodies. Over 40% of the synthetic dyes are carcinogenic chemicals pose serious threat to human health. Dye effluents also damages aesthetic beauty of the water bodies. Thus both toxicological and aesthetic concerns make the environmental release of dye effluents distressing. Additionally, as per Hua *et al.* (2018), the usage of several synthetic dyes has negative consequences on the environment and is particularly linked to damage to human health in terms of allergies, toxicity, and cancer Therefore, removal of dye pollutants from water is essential. According to Agarwal *et al.* (2021) and Richard *et al.* (2019) several methods have been explored, nonetheless, adsorption is the most cost-effective and time-saving technique for removal of dyes.

An efficient way to remove dye from aqueous solutions and industrial wastes is through the adsorption process and for water filtration, a variety of adsorbents have been employed. Although expensive, activated charcoal is found to be powerful adsorbent. It has several benefits, including a smaller footprint, increased design flexibility, and improved organic contamination removal. It is estimated that approximately, 800,000,000 metric tons of flowers are used as offerings each year in religious places across our country, and every year, approximately 8 million metric tons of flowers are thrown in the river (Bennurmath *et al.,* 2021). Waghmode *et al.* (2018) reported managing floral waste as a really difficult task anywhere in the world. According to Agarwal *et al.* (2021), flowers are utilized year-round in India at festivals and temples, which results in the production of large amount of floral waste. While dried flower has certain adsorption properties and may be used to purify water, its effectiveness is minimal. Therefore, charcoal produced from the controlled combustion of material is an improved adsorbent for use in water purification as per Mondal *et al.* (2015). Several studies have been done earlier utilising plants fruiting parts including floral waste as adsorbents for dye removal. Batool *et al.* (2021) carried out the study in which toxic dye was removed from water using soy waste that had been lightly treated with silica. The soy waste adsorbent with a silica coating demonstrated improved percentage removal of dye up to 97.1%. The adsorption method to remove harmful dyes (Congo red and Methylene blue) from their aqueous solution by using activated carbons prepared from ashitaba waste and walnut shell was also studied by Li *et al*. (2020).

Activated carbon extracted from the *Borassus aethiopum* flower was utilised as an adsorbent in a research by Nethaji *et al.* (2010). The effectiveness of batch adsorption experiments for Malachite Green removal from aqueous solutions was investigated by adjusting the beginning solution pH, adsorbent dose, initial day concentration, and temperature. Saravanan *et al.* (2021) and Jegan *et al.* (2020) made biochar from the shells of *Arachis hypogaea* (Groundnut shell) and used the same to remove Basic Blue dyes BB41 and BR09 from an aqueous solution. Senthil kumar *et al.* (2006) developed phosphoric and sulfuric acid-treated activated carbons (PAAC and SAAC) from coconut tree male flowers for adsorption of the basic dye Crystal Violet. They were successful to use equilibrium data with the purpose of learning more about the rate and mechanism of dye adsorption on each kind of carbon. For PAAC and SAAC, the adsorption capabilities of crystal violet increased from 19.8 to 96.80% and 7.02 to 48.83%, respectively when temperature was increased from 28 to 48˚C. There is no protocol for the disposal of the tonnes of garbage that arrive from numerous temples, and this article provides a short description of the generation of waste flowers from temples in Greater Noida West city and its consequences on the environment, including environmental and health concerns.

Generally, there are several ways to use floral waste sustainably like production of compost, vermin compost, value added products like incense sticks, natural colours etc., but this study is different from earlier studies as it emphasizes on the method of producing biochar from floral waste for the removal of azo dye from waste water. The development of readily available, potentially more affordable activated carbon is required for removal of recalcitrant dyes. The primary aim of this research was to spread the idea of "Clean and Green Temples" via the responsible handling of floral waste and the subsequent use of biochar made from this waste water treatment substance used to remove azo dye.

Materials and Methods

Collection of floral waste: The flowers of *Tagetes erecta* used in this study were procured from the temples in and around Greater Noida West. Gautam Buddh Nagar, Uttar Pradesh. The flowers were separated from the temple waste. The blossoms were sun-dried for three days after being rinsed three times with distilled water. The powdered, dry ingredients were mashed in a blender to turn it into the useable form.

Preparation of biochar by Direct Pyrolysis Process: Finely crushed flowers (10 gm) were used to make floral bio char by direct pyrolysis (FBDP). The crushed flowers were heated in a crucible for 2 hr at 550 °C. After resuming to room temperature, material was washed with distilled water and dried at 110°C in an oven for 6 hrs. The dried flowers were grinded in a mortar and pestle to get a fine powder. The powdered samples were sieved to get a uniform particle size of 110 µm to create direct pyrolysis FB_{DP}. It was then stored in an airtight container for further study as proposed by Singh *et al.* (2018). The physio-chemical analysis of FB_{DP} revealed that the ash content was 4.3%, volatile matter was 17.6%, fixed carbon content was 73.4% and porosity was 65.3%, respectively.

Preparation of chemically activated biochar with H_2SO_4 and

H_3PO_4: Powdered waste flower material (10 gm) was taken in two crucibles and treated with 50 ml of sulphuric acid and 50 ml of phosphoric acid respectively. The materials were set aside for complete charring for 24 hr at room temperature once the reaction stopped. The extra acid was decanted after impregnation. Thereafter, both the samples were transferred to clean crucibles of known weight and heated in a muffle furnace for 2 hrs at 450 °C for carbonization. The products were kept at a room temperature for 3 hrs and later washed with distilled water (pH = 7) to remove colour and contaminants before final drying at 110 °C in an oven. The dried product was crushed to powder with a mortar and pestle and then sieved. Both sulphuric (FB_S) and phosphoric (FB_P) acid-treated floral biochar of uniform particle size (110 µm) were stored in an airtight container for further use as reported by Nethaji *et al.* (2010) . Fig. 1 shows the flowers procured discarded from the temples, sorted, collected, and reused.

Physio-chemical analysis of biochar: The pH and electrical conductivity of the biochar sample was estimated with a Labman LMPH-10 electronic pH meter and a Mettler Toledo portable conductivity meter. FB_{DP}, FB_S, and F_{BP} were evaluated in terms of their moisture content (% by mass), ash content (% by mass), volatile content (% by mass), fixed carbon, bulk density (g l^{-1}), porosity (percentage), specific gravity (g cm^{-3}), iodine number (mg g^{-1}), and methylene blue number (mg g^{-1}). These evaluations were carried out utilizing techniques as per industry standard ASTM (1980); Brunauer *et al.* (1938) and Hauge *et al.* (1927).

Removal of methylene blue dye from water: Methylene Blue was selected as a model pollutant to assess the removal efficiency of biochar made from direct pyrolysis (FB_{DP}) from contaminated water. In order to conduct batch adsorption tests, 100 ml of dye solution of varying concentration and pH was added to the biochar and stirred at room temperature using a magnetic stirrer at constant rpm trials. Following agitation, the samples were removed from the stirrer, and dye solutions and the biochar were separated by centrifugation. By measuring the absorbance at 665 nm before and after the treatment, it was possible to determine the residual dye content in the supernatant. The given Equation (i) was used to establish the percent removal rate of Methylene Blue as mentioned by Nethaji *et al.* (2010).

$$\% \text{ Removal} = \frac{A_t - A_0}{A_0} \times 100 \text{ ----------(i)}$$

where, A_t = Absorbance of dye solution at different time interval (t); A_0 =Absorbance of original dye solution. The efficiency of the bio chars were evaluated at different pH, contact time, different concentration of adsorbent as well as dye concentration.

Statistical analysis: In this study to assess the statistical significance differences between the sample mean values, Tukey test was applied to identify significant differences between the means of different concentration of adsorbent dose, pH values, contact time, initial dye concentration and desorption and adsorbent regeneration. The procedure's level for significance was set at $p<0.05$ (significant). The information is displayed as arithmetic means with standard deviations in the form of bar graphs. Microsoft Excel 2010 and Statistical Packages for Social Sciences (SPSS) 23.0 IBM version Software were used for conducting all the statistical analyses.

Results and Discussion

After the physicochemical analysis of biochars obtained from different methods *viz.* direct pyrolysis and chemical activation by using sulphuric acid and phosphoric acid, it was found that the biochar obtained from direct pyrolysis (FB_{DP}) had a higher surface area, well-developed porosity, good carbon composition, high methylene blue number and iodine number as compared to carbon biochar made using sulphuric and phosphoric acids. In the present study, the pH value of activated carbons FB_{DP}, FB_S and FB_P were 7.53, 6.54 and 7.23, respectively. Panditha *et al.* (2021) reported similar results where the pH of biochar of *Chrysanthemum morifolium* obtained from pyrolysis at 350°C and 500°C was 7.8 and 8.7, respectively. In comparison to biochar FB_S and biochar FB_P, bio char FB_{DP} had lower amount of moisture (5.3%), ash (4.3%), and volatile matter (17.6%), *i.e.,* low particle density, leading to increased adsorption. The micropore

(A) (B) (C)

Fig. 1: Floral Waste: (A) Collection and segregation of used flowers (B) and Utilization of Flowers to form Biochar (C).

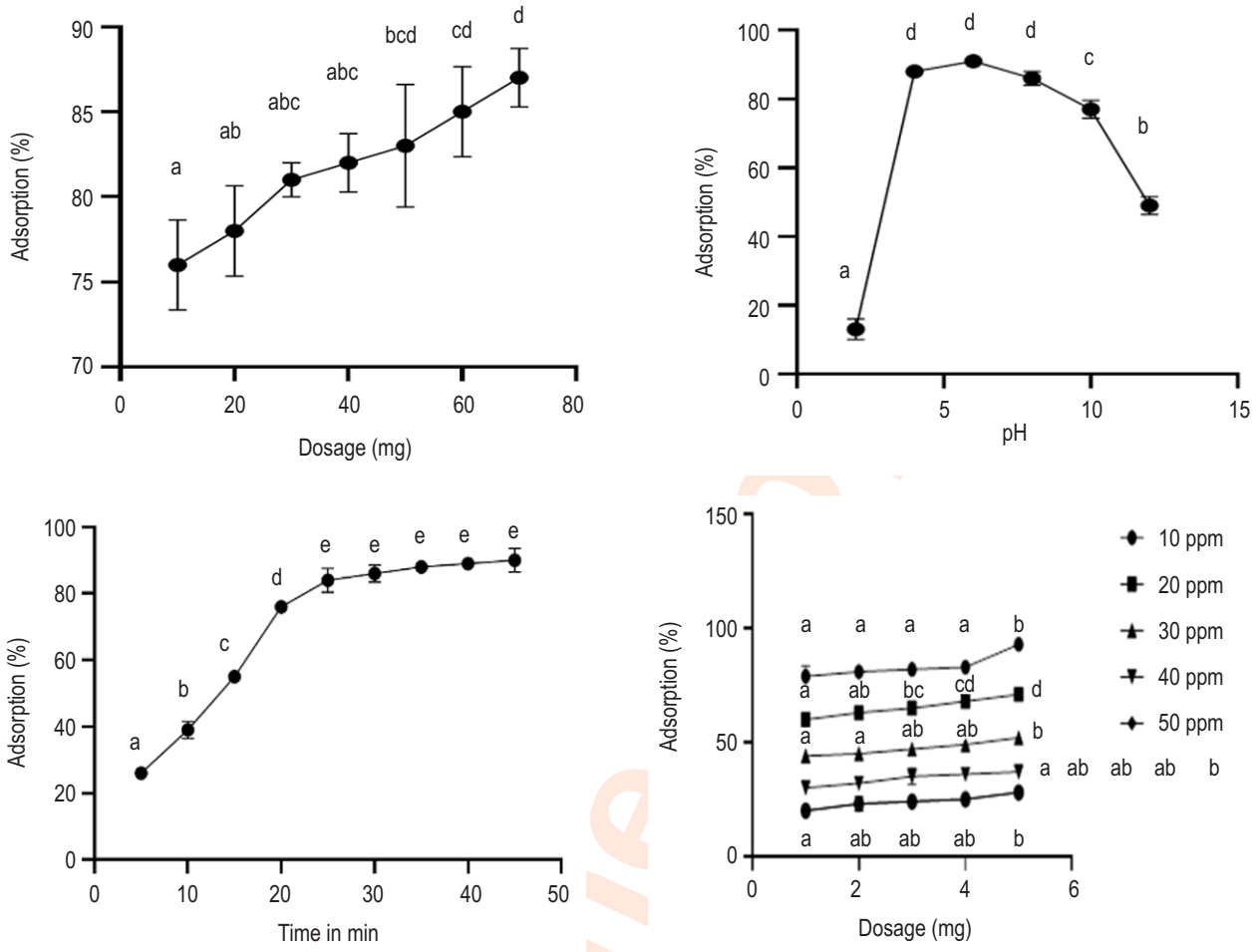

Fig. 2: Effect of biochar dosage, pH, contact time and initial dye concentration on adsorption of methylene blue. Means with different letters are significantly different at $p<0.05$ and the error bars represent standard deviation

content of the biochar was measured in terms of iodine number. The enormous surface area of biochar is due to the presence of micropores. The iodine number for FB_{DP} was 644.0 mg g^{-1}, which was within the range of 600 to 1100 mg g^{-1} as established by ASTM D4607-14 (2021). The methylene blue number and fixed carbon content was highest in FB_{DP} as compared to FB_S and FB_P, clearly indicating the highest adsorption activity of dye. Thus, in this study, the biochar FB_{DP} made from direct pyrolysis of temple waste flowers can be considered as a reliable dye adsorbent. The results of various parameters are presented in Table 1. Studies were also conducted to find out the effect of dosage, pH, contact time, initial dye concentration, desorption and adsorbent regeneration of biochar FB_{DP}. At neutral pH, the room temperature, and continuous stirring, the percentage adsorption of methylene blue dye of FBDP was investigated at various biochar dosages (10, 20, 30, 40, 50, 60, and 70 mg 100 ml^{-1}, respectively) for 45 min.

Fig. 2 shows the influence of adsorbent dosage on the equilibrium adsorption percentage. On increasing the amount of adsorbent (biochar), the amount of dye adsorption tends to increase. The percentage of adsorption was significantly increased ($p<0.05$) from 76% to 87.0% when the FB_{DP} dosage was raised from 10.0 to 70.0 mg 100 ml^{-1}. With the increase in dosage, the effective adsorbent's surface area also increased. Adsorption rates declined beyond 70 mg 100 ml^{-1} biochar due to saturation of adsorption sites and dye molecules in solution. Similar results were observed by Hameed (2009) on adsorption of methylene blue dye using Jackfruit peel. On increasing the jackfruit peel dose from 0.05 to 0.60 g, the rate of percent removal increased from 58.20 to 89.80%. Similarly, Shakoor (2016) demonstrated that as the concentration of *Citrus limetta* peel adsorbent increased from 0.4 to 2.0 gl^{-1}, the removal efficiency of methylene blue dye increased rapidly from 94.6 to 97.1%, respectively. Similar results were obtained by Singh *et al.* (2018), on increasing the dose of biochar made from marigold flower (0.5

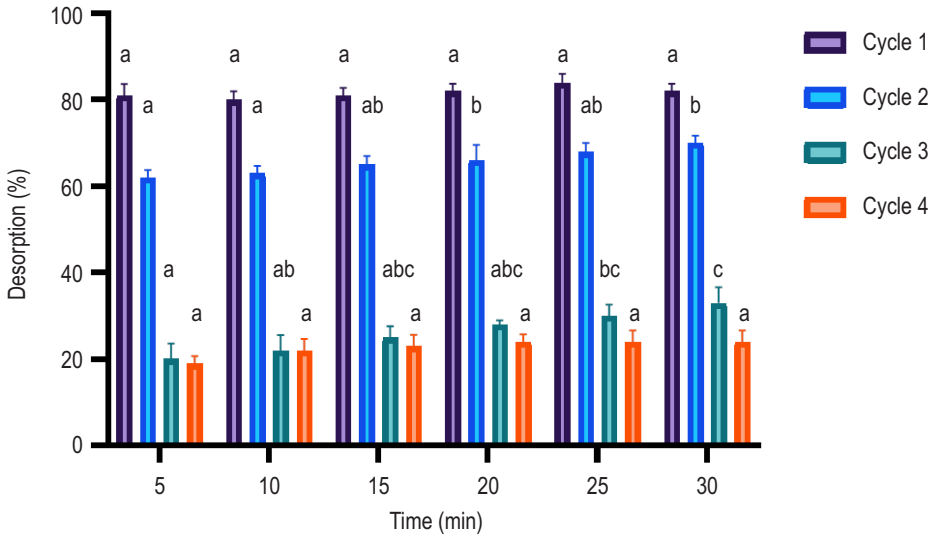

Fig. 3: Desorption (regeneration) studies plot for cycle 1 to cycle 4. Means with different letters are significantly different at p<0.05 and the error bars represent the standard deviation.

Table 1: Physico-chemical analysis of biochars obtained from various methods (by direct pyrolysis, chemical activation by sulphuric acid and phosphoric)

Parameters	FB_{DP}	FB_S	FB_P	Standard values as per ASTM
pH	7.53±0.19	6.54±0.07	7.23±0.19	6-8
Electrical conductivity (S m^{-1})	0.27±0.008	0.20±0.06	0.10±0.04	Depends upon the material
Moisture content (%)	5.3±0.24	7.8±0.08	10.6±0.38	5-8
Ash content (%)	4.3±0.16	6.5±0.40	9.2±0.33	5-15
Volatile matter (%)	17.6±0.24	20.4±1.52	23.5±1.48	37. 5 ± 0.03
Fixed Carbon (%)	73.4±1.06	69.2±0.41	57.4±1.18	Depends upon the material
Porosity (%)	65.3±1.51	58.9±1.32	60.71±1.98	48-85
Bulk density (g ml^{-1})	0.48±0.02	0.44±0.02	0.49±0.02	0.25
Specific gravity	0.80±0.08	0.95±0.02	0.84±0.09	≈1.8
Iodine number (mg g^{-1})	644±2.0	439±7.2	191±1.6	600-1100
Methylene blue number(mg g^{-1})	519±1.24	422±2.8	139±3.6	≈450

Values are mean ± S.D.

to 2.0 gl^{-1}), the amount of methylene blue adsorption was increased to 89% within a time period of 2 hr. The optimal pH required for FBDP biochar to remove maximum colour from methylene blue dye was found to be 6.0. Akar *et al.* (2013) reported that the adsorption rate is significantly influenced by the pH of dye solution. The pH of the solution was adjusted from 2.0 to 4.0, 6.0 to 8.0, and 10.0 to 12.0 by adding either HCl or NaOH as done by Hameed (2009). On increasing the pH from 4.0 to 6.0, there was a significant increase ($p<0.05$) in dye removal from 88.0% to 91.0%, as depicted above in Fig. 2. Since methylene blue dye is a basic dye, the efficacy of removal steadily diminishes on increasing the pH levels. Similar trend of increasing removal efficiency of azo dyes with increase in pH has been also reported by Babaei *et al*. (2016), Hameed (2009), Saravanan *et al*. (2021) and Singh *et al*. (2018) using biochar as adsorbent.

The adsorption of methylene blue dye by biochar FB_{DP} was rapid initially and then, decreased gradually until a steady state was attained. The results demonstrated that saturation significantly increased ($p<0.05$) adsorption up to 90.6% after 45 min of contact time. Fig. 2 depicts the relationship between contact time and percent adsorption. Similar results have been observed by Mohram *et al.* (2022) where adsorption percentage of methylene blue and crystal violet dyes increased rapidly by 98% in the first 15 minutes of contact time. Shakoor (2016) also studied citrus limetta peel as a low-cost adsorbent biochar for removal of methylene blue dye and it was reported that adsorption capacity of methylene blue dye increased when the contact time of biochar citrus limetta peel was increased from 10 to 30 min. Similarly, Sudan *et al.* (2023) also found the adsorption capacity of biochar made rice husk to remove Eriochrome black T

dye increased up to 94% after the contact time was increased up to 2 hr. This indicates that the majority of FB_{DP} adsorption sites present in the biochar were on its outer surface, where the methylene blue dye could readily reach them (Mittal *et al.,* 2016; Baek *et al.,* 2010).

Adsorption of methylene blue dye was carried out at pH 6 and dye concentration ranging from 10 to 50 ppm, using an optimal amount of 70 mg of bio char FB_{DP} for 30 min. The percentage adsorption of methylene blue dye significantly increased ($p<0.05$) from 28 to 93% when the dye concentration was reduced from 50 to 10 ppm. Fig. 2 demonstrates the concentration of azo dye influencing the adsorption percentage, the reason being that the available adsorption sites of biochar are incapable of adsorbing more concentrated dye molecules. Similar results were observed by Hameed (2009), when the adsorption of methylene blue reduced from 93.33 to 86% jack fruit peel as biochar, when the initial concentration of the dye was increased from 35 to 400 mgl^{-1}. The increased dye concentration most often leads to decrease in adsorption capacity of biochar as with increasing dye concentration adsorption sites of biochar tends to get saturated and can no longer carry out further adsorption with increasing concentration. The cost of the overall procedure and the amount of pollutant recovered are both affected by the desorption and adsorbent regeneration phase. Based on these results the percentage of dye molecules as a function of desorption percentage (%) vs time (in minutes) is shown in Fig. 3. A significant ($p<0.05$) decreasing trend was observed in the desorption percentages (81>62>20>19) from cycle 1 to cycle 4. Mahadevaswamy *et al.* (2016). Demonstrated that the percentage desorption of hazardous azo dye decreased in the order 87 >72 > 33 > 21 for every recycling procedure in the order of Cycle 1 > Cycle 2 > Cycle 3 > Cycle 4 using activated carbon made from Parthenium flower.

Activated biochars can be efficiently used as adsorbent due to their high surface area and adsorption capacity, presence of macropores and micropores. In the current study, biochar was prepared and used to study the adsorption efficiency of hazardous azo dye (methylene blue dye) from its aqueous solution. This study is one of the waste reduction ways for making biochar from waste marigold flowers through direct pyrolysis and chemical activation under ideal conditions using sulphuric acid and phosphoric acid. After comparing the outcomes of different biochars, it was found that the biochar made through direct pyrolysis showed better results than the biochar made through an acid procedure as it had higher surface area, well-developed porosity, and a good carbon composition compared to biochar made from sulphuric and phosphoric acids. So, from this study it can be concluded that biochar produced by direct pyrolysis of temple waste flowers can be employed as a effective adsorbent for remediation of methylene blue dye (upto 91%) from industrial wastewater as it is a cheapest activation method, and it also has a fair adsorptive capacity as compared to acid procedure. The present work has significant dual importance of resource recovery from floral waste as well as for dye removal from wastewater to be implemented commercially also.

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Authors' contribution: S. Agarwal: Writing— original draft; **N. Rana:** Data Curation; **P. Bhardwaj:** Data Curation, Writing-Software, review and editing; **G.N. Tiwari:** Review and editing; **A. K. Yadav**: Review and editing; **M. C. Garg:** Review and editing; **A. Mathur:** Writing-review and editing; **A. Tripathi:** Conceptualization, Methodology, Software, Validation, Writing-review and editing, Funding acquisition.

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