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Posted Date: 2 January 2026

doi: 10.20944/preprints202601.0066.v1

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Article

Adsorption of Perfluorooctanoic Acid from Aqueous Media Using an Engineered Sugarcane Bagasse Biochar–Chitosan Composite

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Abstract

Recently, several studies from developing economies have reported the presence of per- and polyfluoroalkyl substances (PFAS) in water bodies, with a dominance of Perfluorooctanoic acid (PFOA), a potential endocrine disruptor. In this study, an engineered sugarcane bagasse biochar–chitosan composite (SBCT) was designed, synthesized, and evaluated as an adsorption medium for the removal of PFOA from aqueous systems at concentrations up to 500 ppb in water. Batch adsorption experiments were conducted to investigate the effects of initial PFOA concentration, contact time, pH, adsorbent dosage, and temperature. Scanning electron microscopy (SEM) showed that SBCT has a significant porous structure. The composite showed over 90% of PFOA removal from water. Further, the presence of peaks corresponding to C-F bonds after adsorption by Fourier transform infrared (FTIR) Spectroscopy analysis confirms the adsorption of PFOA on SBCT. The protonated amine groups (NH_3^+) in chitosan enhanced the adsorption of anionic PFOA through electrostatic attraction with carboxyl groups (COO^-). The Kinetic study revealed that Pseudo-first order best described the adsorption process, with equilibrium adsorption capacity (q_{eq}) of 2.78 mg/g, suggesting that physisorption is the predominant mechanism. The Langmuir Isotherm model gave the best fit, establishing a maximum adsorption capacity (q_{max}) of 9.08 mg/g. Thermodynamic analysis revealed that the adsorption process was spontaneous and exothermic, consistent with physisorption. The regeneration capacity of the SBCT composite demonstrated exceptional reusability across five adsorption-desorption cycles with methanol. The adsorption kinetics, equilibrium behavior, and regeneration efficiency suggest that SBCT is a viable low-cost adsorbent for batch adsorption-based treatment systems targeting PFOA removal, particularly in decentralized and resource-constrained water treatment applications.

Keywords: PFOA removal; adsorption; biochar; chitosan; composite; regeneration

1. Introduction

Perfluorooctanoic acid (PFOA), used as an anionic surfactant, is a long-chain perfluoroalkyl carboxylic acid (PFCA) within the broader class of per- and polyfluoroalkyl substances (PFAS) [1]. PFOA is utilised as a processing aid in making fluoropolymers for the production of non-stick cookware, waterproof clothing, and insulation, and in coatings, fire-fighting foams, and lubricants for their oil, water, and stain resistance [2–4]. The worldwide emissions resulting from commercial PFOA manufacturing were estimated between 90–970 tonnes from 1951 to 2002, 30–430 tonnes from 2003 to 2015, and are projected to reach 630 tonnes between 2016 and 2030, while emissions from

fluoropolymer production were reported between 1,220–6,560 tonnes (1951–2002), 660–3,870 tonnes (2003–2015), with an expected increase to 4,520 tonnes (2016–2030) [5].

Owing to its remarkable chemical stability and surface activity ascribed to its strong carbon–fluorine (C–F) linkages, PFOA is resistant to biodegradation [6,7] and is listed as a persistent organic pollutant (POP) in Annex A of the Stockholm Convention since 2019 [1,8]. Despite its widespread utilization in industrial and domestic applications, multiple studies have reported its toxicological effects, including carcinogenicity, genotoxicity, endocrine disruption, liver toxicity, immunological dysfunction, and reproductive issues [3,9,10]. PFOA is defined as hydrophobic and oleophobic due to its low fluorine polarizability, but it has a robust and particular affinity for human serum albumin, as evidenced by [11] and supported by molecular simulations from [12], highlights its pronounced proteinophilic nature.

The hydrophobic and lipophobic characteristics of PFASs and their longevity provide important challenges in remediation efforts [13,14]. Wastewater treatment facilities are significant sources of PFAS, since PFOA concentrations in effluents (110 ng/L) surpass those in influents (100 ng/L) at a U.S. facility, demonstrating inadequate effectiveness in removal [15]. In China, elevated PFOA concentrations in drinking water are ascribed to upstream wastewater treatment plant discharges into source streams [6,16]. Common treatment procedures are typically inadequate for eliminating trace levels of PFOA from wastewater, while advanced technologies are sometimes prohibitively costly. Adsorption, especially with materials like activated carbon, provides a more cost-effective and efficient solution, delivering enhanced removal performance and less secondary pollution relative to methods such as coagulation, degradation, or filtration [7,17,18]. Adsorption is recognized as a simple, effective, and cost-efficient method for removing PFASs from water. The efficacy of diverse adsorbents in eliminating PFOA has been examined, encompassing nanomaterials, activated carbon, carbon nanotubes, ion exchange polymer resins, and different minerals [19–22]. Biochar, an economically carbon-dense substance generated through the pyrolysis of biomass under low-oxygen conditions, has emerged as a potential adsorbent for removing PFOA from water and wastewater [23–25]. Biochar is preferred over other adsorbents because of its cost-effectiveness, superior adsorption performance, and ecologically friendly properties. Additionally, one of the benefits of employing biochar is its ability to be customized for specific carbon sequestration [26]. Sugarcane bagasse, an agro-industrial byproduct, is considered a suitable raw material for extensive biochar production owing to its plentiful availability, economic viability, substantial mineral content, and inherently porous fibrous structure [27]. Again, Chitosan's non-toxicity, hydrophilicity, biodegradability, and biocompatibility render it a cost-effective, renewable, and eco-friendly sorbent for effective water purification [28]. Chitosan-modified biochar has demonstrated improved efficacy in augmenting the adsorptive capacity for a wide range of contaminants, including arsenic (V)[29], Ofloxacin [30], and heavy metals [31].

In this study, we have synthesized a novel engineered biochar in the form of sugarcane bagasse biochar/chitosan composite (SBCT) for effective removal of PFOA present even in trace levels in water. The SBCT composite material was characterized using FTIR methodology, SEM examination, and BET surface area methods. Adsorption kinetics and isotherm models were employed to evaluate the PFOA uptake capacity. We have further investigated the impact of initial PFOA concentration, adsorbent dosage, pH of the aqueous PFOA solution, and temperature on adsorption effectiveness. Additionally, the potential for reusing the exhausted adsorbent was examined using several regeneration agents. Finally, the effectiveness of SBCT has been compared with other adsorbents for PFOA removal.

2. Materials and Methods

2.1. Adsorbate

Perfluorooctanoic acid (PFOA), chitosan (deacetylated chitin), and epichlorohydrin (ECH) were sourced from Sigma Aldrich. Every additional reagent was of analytical grade. The stock solution

was initially made with methanol, while Milli-Q water formulate all subsequent working solutions. The concentrations of the prepared solutions were verified before starting the experimental procedures.

2.2. SBCT Preparation

The novel SBCT was synthesized utilizing a modified technique derived from established methods in the literature [30,32]. The raw material, sugarcane bagasse (SB), was procured from a local cane juice seller in Tamil Nadu and divided into smaller pieces. The bagasse was thoroughly cleaned with Milli-Q water to eliminate the surface contaminants and subsequently dried in a hot air oven at 105 °C for 8 hours to eradicate moisture content. The dried bagasse was powdered using a grinder and subjected to isothermal pyrolysis in an oxygen-limited environment in a preheated muffle furnace at 350 °C for 70 minutes. The residual ash and other contaminants from the biochar were removed by washing the biochar with Milli-Q water. The biochar was dried at 80 °C in a hot air oven, screened through a 150 µm nylon mesh, and stored in a sealed container within a desiccator to prevent moisture absorption. In this procedure, 2 g of chitosan powder was dissolved in 200 mL of a 3% (v/v) acetic acid solution while being magnetically stirred to obtain a homogeneous solution. Subsequently, 4 g of biochar derived from sugarcane bagasse was integrated into the 2 g chitosan solution in a 2:1 ratio (SB: CT) and stirred continuously for 2 hours. To facilitate crosslinking, 300 mL of a 2% (v/v) epichlorohydrin (ECH) solution was introduced, and the mixture was stirred in a water bath shaker at 40 °C for 30 minutes. The pH was later adjusted to a range of 8.0 – 10.0 using a 1 mol/L NaOH solution. The resultant composite was meticulously cleaned to eliminate any remaining alkali before utilization.

2.3. Adsorbent Characterisation

The chemical functionality and composition of the SBCT composite were examined via Fourier-transform infrared spectroscopy (FTIR) utilizing a PerkinElmer ALPHA-FT-IR spectrometer, covering a wavelength range of 400–4000 cm⁻¹. Surface morphology and structural properties were assessed using scanning electron microscopy (SEM) employing an FEI Quanta 200 instrument. At the same time, the composition of the elements was analysed through energy-dispersive X-ray spectroscopy (EDAX) using an FEI Nova SEM 450. The Brunauer–Emmett–Teller (BET) surface area was ascertained by nitrogen adsorption-desorption isotherms at 77 K utilizing a Quantachrome Autosorb IQ analyzer. The point of zero charge (pZC) of the SBCT composite was ascertained using the specified methodology given elsewhere [33]. Briefly, a 50 mL solution of 0.1 M NaCl was mixed with 0.5 g of biochar and permitted to remain undisturbed for 24 hours to reach equilibrium. A series of solutions with differing initial pH values (pHi) was generated by adjusting the pH using 0.1 M hydrochloric acid and sodium hydroxide solutions. After the equilibration interval, the final pH (pHf) was documented, and the pH difference ($\Delta\text{pH} = \text{pHi} - \text{pHf}$) was calculated. A graph of ΔpH against pHi was constructed to determine the pZC. All measurements were conducted in triplicate to guarantee repeatability.

2.4. Batch Adsorption of PFOA

Adsorption studies were conducted in duplicate utilizing the SBCT composite as the adsorbent. Simulated wastewater with perfluorooctanoic acid (PFOA) concentrations between 0.5 and 4 mg/L was generated, and 30 mL of this solution was placed into 50 mL Nalgene tubes. Each tube was allocated 0.02 g of the SBCT material and underwent agitation at 150 rpm in an orbital shaker, sustained at ambient temperature (25 °C), for different contact times. After treatment, the suspensions were filtered through a 0.22 µm membrane to isolate the solid and liquid phases. Before analysis, the filtrate was subjected to derivatization, transforming PFOA into its anilide form through the use of 2,4-difluoroaniline (DFA) as the derivatizing reagent and N, N-dicyclohexylcarbodiimide (DCC) as the dehydrating agent [34]. PFOA was quantified in a GC-ECD system (Agilent 7890B) using an HP5

column (30 m length, 0.32 mm internal diameter, 0.25 μm film thickness). Nitrogen was used as the carrier gas, with a total flow rate of 18.8 mL per minute. The temperature of the detector was maintained at 280 $^{\circ}\text{C}$. Sample injections were conducted in splitless mode with a fixed injection volume of 1 μL .

The adsorption capacity at equilibrium, q_{eq} (mg/g), and PFOA removal efficiency, R_{E} (%), were estimated with Eq. (1) and Eq. (2):

$$q_{\text{eq}} = (C_{\text{in}} - C_{\text{eq}})V_0/m_0 \quad (1)$$

$$R_{\text{E}}(\%) = \frac{C_{\text{in}} - C_{\text{tc}}}{C_{\text{in}}} * 100 \quad (2)$$

C_{in} refers to the initial PFOA concentration, while C_{eq} (mg/L) represents the equilibrium state PFOA concentration. The variable C_{tc} (mg/L) indicates the concentration at a given time of contact, m_0 represents the adsorbent's mass in grams, and V_0 denotes the PFOA volume in mL. Each of the experiments was conducted in duplicate under controlled temperature conditions.

2.5. Isotherm Modeling of Adsorption

The adsorption nature and behavior of PFOA on SBCT were evaluated by changing the initial PFOA concentrations between 0.5 and 4 mg/L. The amount of PFOA adsorbed at each concentration level was used to construct adsorption isotherms, which were then analyzed using the nonlinear plotting of the Langmuir model given in Eqs. (3) and (4), the Freundlich model in Eq. (5), and the Temkin model in Eq. (6) as used elsewhere [35].

$$q_{\text{eq}} = q_{\text{max}} K_{\text{Lq}} C_{\text{eq}} / (1 + K_{\text{Lq}} C_{\text{eq}}) \quad (3)$$

$$R_{\text{Lq}} = 1 / (1 + (1 + K_{\text{Lq}} C_{\text{in}})) \quad (4)$$

$$q_{\text{eq}} = K_{\text{fd}} C_{\text{eq}}^{1/n} \quad (5)$$

$$q_{\text{eq}} = B_{\text{Tm}} \ln K_{\text{Tm}} + B_{\text{Tm}} \ln C_{\text{eq}} \quad (6)$$

C_{eq} (mg/L) and q_{eq} (mg/g) denote PFOA concentration and SBCT adsorption capacity at the state of equilibrium, and q_{max} (mg/g) indicates the anticipated maximum monolayer adsorption capacity of PFOA. The ' K_{Lq} ' (L/mg) is the Langmuir constant. The dimensionless separation factor R_{Lq} , calculated using Eq. (4), elucidates the favorability of the adsorption process, where $R_{\text{Lq}} > 1$ indicates adsorption is not favorable, $R_{\text{Lq}} = 1$ denotes adsorption is linear, and $R_{\text{Lq}} < 1$ signifies adsorption is favorable. K_{fd} (mg/g) in Eq. (5) is the Freundlich adsorption capacity constant, n indicates adsorption intensity, a $1/n$ value < 1 indicates favorable adsorption and surface heterogeneity; $1 < n < 10$ also suggests favorability. $1/n > 1$ implies phase agreement, while $n=1$ denotes concentration-independent partitioning. K_{Tm} (L/g) denotes Temkin equilibrium binding constant, and B_{Tm} is associated with heat of sorption (J/mol).

2.6. Kinetic Modeling of Adsorption

Kinetic models were used to elucidate the adsorptive mechanism of PFOA onto SBCT. The three kinetic models applied were: the pseudo-first-order model (Eq. 7), the pseudo-second-order model (Eq. 8), and the Weber–Morris intra-particle diffusion model (Eq. 9) [36].

$$\ln(q_{\text{eq}} - q_{\text{tc}}) = \ln q_{\text{eq}} - k_1 t_c \quad (7)$$

$$t_c/q_t = 1/k_2 q_{\text{eq}}^2 + 1/q_{\text{eq}}(t_c) \quad (8)$$

$$q_{\text{tc}} = 1 + k_{\text{ipd}} t_c^{1/2} \quad (9)$$

K_1 (1/min) in Eq. (7) is the equilibrium coefficient of pseudo-first-order kinetic uptake per minute. The parameters ' q_{eq} ' and ' q_{tc} ' represent the adsorption capacity at equilibrium and at contact time (t_c), respectively. In Eq. (8), ' K_2 ' (1/min), the rate constant for pseudo-second-order kinetics. In Eq. (9), ' k_{ipd} ' is the intra-particle diffusion rate constant, ' t_c ' is the time of contact (min), and ' i ' represents the boundary layer effect.

2.7. Thermodynamic Studies

Temperature significantly affects both the rate and capacity of adsorption. To evaluate the impact on PFOA adsorption, experiments were carried out within the 20 °C to 40 °C range. The typical Gibbs free energy change (ΔG^0) was calculated utilizing the Van't Hoff equation [37].

$$\Delta G^0 = -RT \ln(K_v) \quad (10)$$

$$\ln K_v = -\Delta H^0/RT + \Delta S^0/RT \quad (11)$$

$$\Delta G^0 = \Delta H^0 - T\Delta S^0 \quad (12)$$

Gibbs free energy (ΔG^0 , kJ/mol), enthalpy change (ΔH^0 , kJ/mol), and entropy change (ΔS^0 , kJ/mol) were evaluated by Eq. (10) to Eq. (12), where K_v represents the dimensionless equilibrium constant, R represents universal gas constant, and T denotes the absolute temperature in Kelvin.

3. Results and Discussion

3.1. Novel Engineered SBCT

In general, engineered biochar augmented through physicochemical methods or biological modifications enhance the structural and surface characteristics, including increased surface area, improved ion exchange capacity, refined pore structure, and elevated functional group content [38,39]. It is to be noted that compared to activated biochar, pure, unmodified biochar often has a lower adsorption effectiveness, and its low density and small particle size make separation challenging, which limits its usefulness in water treatment [40,41]. Chitosan, a renewable polymer derived from chitin in crustacean shells and fungal mycelia, is valued for its versatile structure and eco-friendly applications, particularly in wastewater treatment [42]. The inclusion of protonated amine units (NH_3^+) in chitosan is capable of enhancing the electrostatic interactions with the carboxyl groups (COO^-) of PFOA [43]. However, the practical use of raw chitosan in water treatment is limited by its slow rate of adsorption and susceptibility to degradation in acidic environments [44]. Hence, we prepared engineered composites made of biochar of sugarcane bagasse and Chitosan (SBCT) to provide an effective substrate for enhancing sorption mechanisms, particularly for anionic pollutants like PFOA. Such a novel composite was prepared to investigate whether SBCT shows better removal efficacy of PFOA from aqueous medium over unmodified pristine biochar or raw chitosan.

3.2. SBCT Characterization

a. Surface morphology

The surface configuration of SBCT was examined using SEM, and the resulting micrographs are presented in Figure 1. It can be seen that the SEM images revealed that the surface of SBCT showed wrinkles and groove-like structures, significantly increasing the available pore size for PFOA adsorption. The presence of a rough and porous surface contributes to enhanced adsorption efficiency. The BET analysis revealed that the surface area of SBCT was 127.07 m²/g. Additionally, post-adsorption SEM images showed increased surface roughness and thickness, suggesting successful aggregation of chitosan onto the SBCT composite surface. Elemental analysis revealed a significant increase in fluorine (F) content from undetectable levels to 5.3% after adsorption, confirming the successful adsorption of PFOA onto SBCT, as fluorine is a key constituent of its perfluorinated structure.

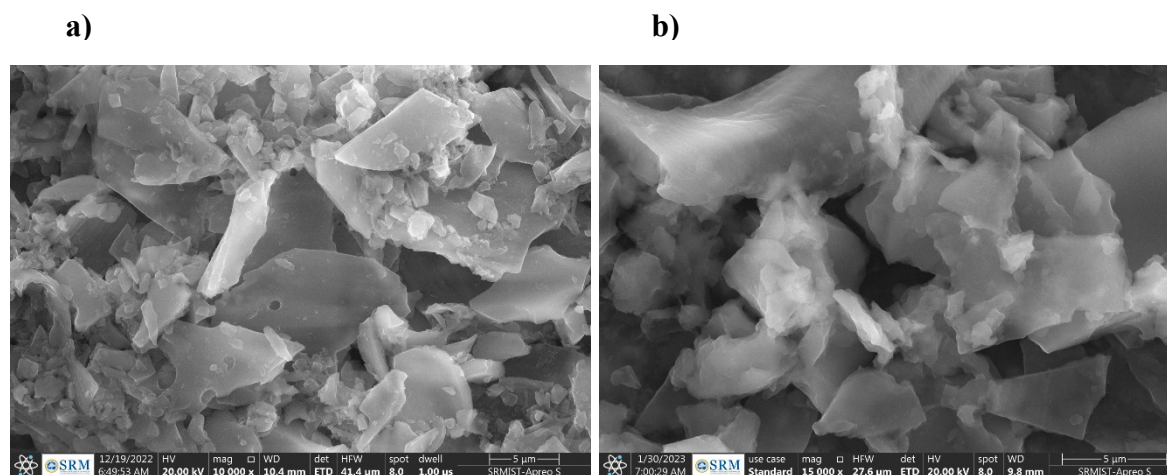


Figure 1. Comparison of SEM images of SBCT (a) before and (b) after adsorption.

b. Point of zero charge (pZC)

The pZC of SBCT was measured as 6.1 (Figure S1). Below pH 6.1, the surface of SBCT acquires a positively charged character, enhancing electrostatic attraction with the anionic form of PFOA. Conversely, at pH above 6.1, electrostatic repulsion may hinder the adsorption efficacy of SBCT. These results indicate that SBCT can function as an effective adsorbent for removing PFOA from aqueous solutions at acidic pH.

c. Chemical Interactions with Functional Groups

The FTIR spectra of the SBCT composite before and after adsorption are represented in Figure 2a and 2b, respectively. The broad absorption bands observed near 3260 cm^{-1} and 3450 cm^{-1} are attributed to the overlapping vibrational modes of N–H and O–H groups. The increased intensity of the O–H band represents the presence of hydroxyl or amine functionalities on the SBCT surface [19]. The absorption band observed at 1580 cm^{-1} is characteristic of the amide (CONH) group, consistent with findings reported by [30]. The peak located between $\sim 1150\text{--}1155\text{ cm}^{-1}$ is attributed to the β -glycosidic linkage in chitosan [45]. Additionally, the band near 1600 cm^{-1} corresponds to the stretching vibration of C=O groups, which are prevalent in carboxylic acids, amides, or ketones. The enhanced intensity of this peak following adsorption suggests effective interaction between PFOA and SBCT, likely facilitated by the carboxyl functional group of PFOA in aqueous media [43]. Peaks appear in this region of $1200\text{--}1140$, corresponding to the C–F bonds in PFOA. This confirms PFOA adsorption [43]. The corresponding C–O Stretch in alcohols, esters, or ethers is given in the signals around $1050\text{--}1100\text{ cm}^{-1}$ [46]. A shift in the N–H absorption peaks toward lower wavenumbers was observed, suggesting that N–H groups may have played a role in the adsorption of PFOA. Thus, the interaction between SBCT and PFOA was collectively confirmed by the spectral changes, supporting the effectiveness of the adsorption mechanism.

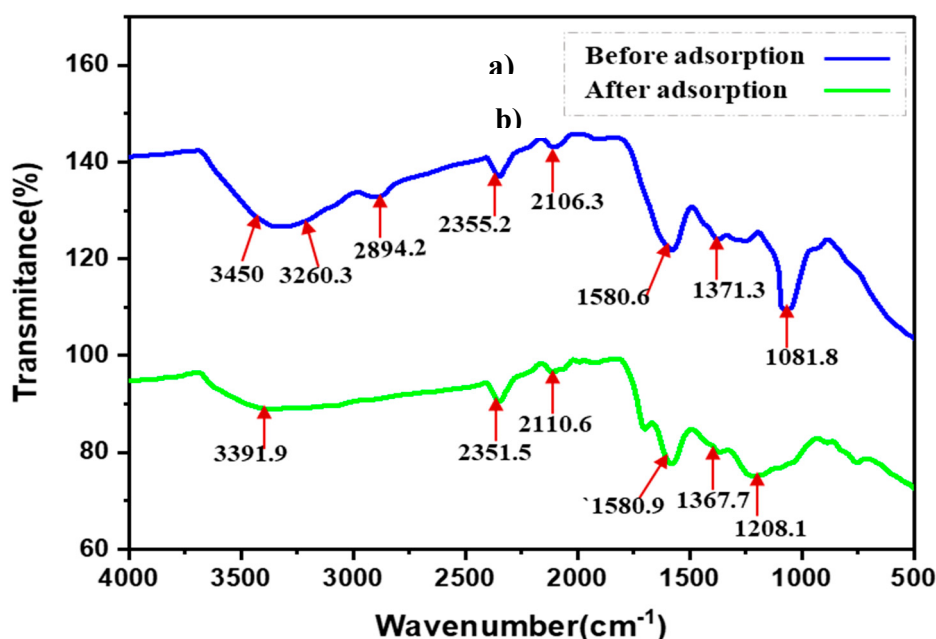


Figure 2. FTIR spectrum for SBCT (a) before and (b) after adsorption.

3.3. Factors Governing PFOA Adsorption

3.3.1. Contact Time

The SBCT composite of 0.02 g was added to the synthetic PFOA solution of 2 mg/L to evaluate the capacity for adsorption of SBCT for PFOA is illustrated in Figure 3. The adsorption capacity of 2.7 mg/g was attained at 240 minutes with maximum PFOA removal of 91%, with a slight increase to 2.71 mg/g at 300 minutes, indicating that equilibrium was effectively reached around 240 minutes. Therefore, all following batch adsorption studies were performed with a contact duration of 240 minutes. In comparison, unmodified pristine sugarcane bagasse biochar (SB) and pure chitosan (CT) demonstrated reduced adsorption capabilities of 1.59 mg/g and 2.05 mg/g, respectively. The SBCT composite was developed by integrating chitosan with sugarcane bagasse biochar in a 1:2 ratio, which possibly enhanced the adsorption capacity of PFOA through electrostatic interactions between protonated positive amine groups (NH_3^+) in chitosan and carboxyl groups (COO^-) in anionic PFOA. The increased surface area, functional groups, hydrophobicity, and synergistic adsorption mechanisms improved the performance of SBCT, as supported by studies showing that chitosan-modified biochar demonstrates significantly higher adsorption capacities than pristine biochar or chitosan alone [30].

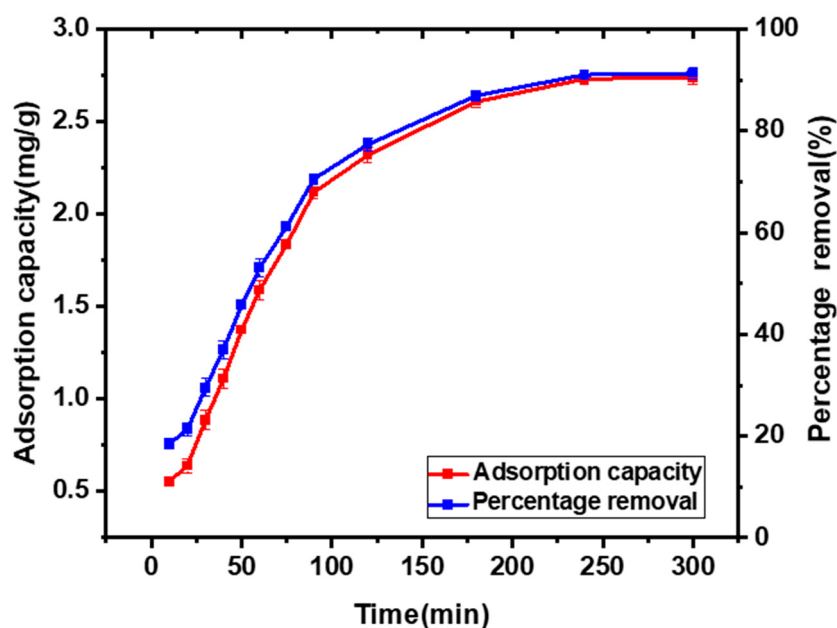


Figure 3. Effect of contact time on adsorption capacity and percentage removal of PFOA using SBCT. Error bars represent the standard deviation of duplicates. (Adsorbent dosage: 0.5 g, PFOA Concentration: 2 ppm, Temp: 25 °C, Time: 240 min).

3.3.2. pH

The effect of change in initial pH levels in the adsorption of PFOA, covering a range starting from acidic (pH 2) to alkaline (pH 12), as illustrated in Figure 4. The adsorption of PFOA declined markedly with the rise in the pH levels is associated with the fact that PFOA predominantly exists in its anionic form under aqueous conditions, bearing a carboxylate group. Hence, hydrogen bonding between the carbonyl functionalities (C=O) of PFOA and the positively charged -NH_3^+ moieties on the composite surface at lower pH leads to pronounced adsorption due to strong electrostatic interactions with the PFOA molecule and SBCT [43,47]. The adsorption process is dominant under acidic conditions associated with this interaction. Our observations are in line with other studies showing higher adsorption of perfluorinated compounds, including PFOA, at acidic pH values, viz., pH 3 [48–50], pH 4 [51], and pH 2 [43]. As the pH increased, these bonds might have started getting disrupted in alkaline conditions, resulting in a decrease in adsorption capacity. When the pH reached 8, the alkaline environment became unfavorable for PFOA adsorption. At pH 10, the adsorption capacity was the least, likely due to the abundance of OH^- ions in a strongly alkaline state. The pZC of SBCT was around 6.1. Below this pH, in more acidic conditions, the surface became positively charged, promoting electrostatic attraction with anionic PFOA and enhancing adsorption. In contrast, at higher pH in alkaline conditions, the surface became negatively charged, thereby reducing adsorption efficiency [30].

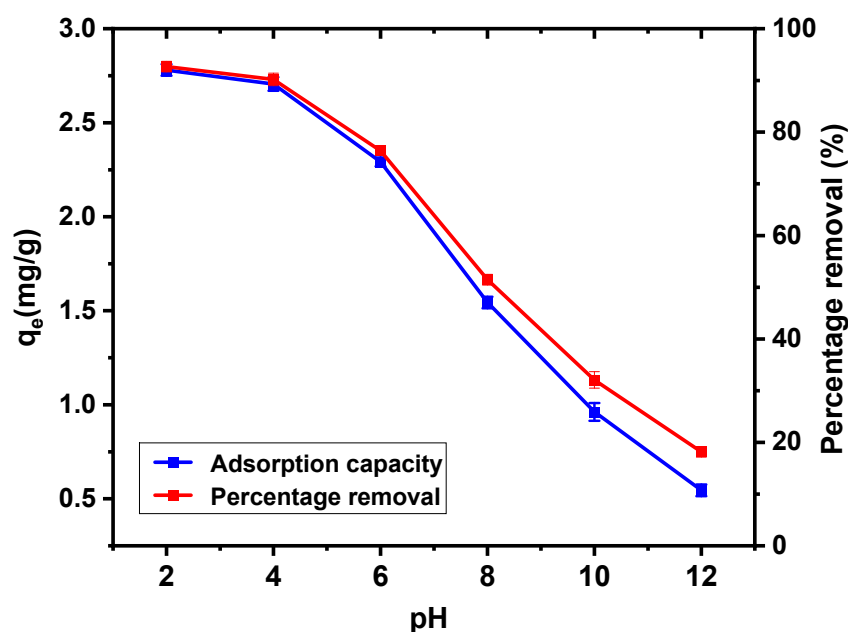


Figure 4. Effect of pH on adsorption capacity and percentage removal of PFOA using SBCT. Error bars represent the standard deviation of duplicates. (Adsorbent dosage: 0.5 g, PFOA Concentration: 2 ppm, Temp: 25 °C, Time: 240 min).

3.3.3. Adsorbent Dosage and Initial Concentration

The adsorptive capacity of PFOA is significantly influenced by SBCT composite dosage and PFOA concentration. In this study, 0.01 g and 0.02 g of SBCT were used to evaluate the adsorbent dose effect on PFOA removal. PFOA showed a higher removal efficiency of 91% at 0.02 g, as illustrated in Figure S2. The PFOA concentration, ranging between 0.5 and 4 mg/L, was examined to reflect both common and severe environmental pollution levels. This range of concentration falls between the range used in other studies [52,53] for PFOA removal. The results reflected that higher PFOA concentration led to a rise in the adsorption capacity. The maximum adsorption capacity ($q_{\max}$) of 5.1 mg/g was recorded at a PFOA concentration of 4 mg/L, showing a gradual increase with rising concentrations. The maximum percentage of PFOA removal was achieved at the lowest concentration tested (0.5 mg/L, Figure S3). The slight reduction in the removal efficiency at higher PFOA concentrations can be attributed to the increasing saturation tendency of active adsorption sites on the SBCT surface. This phenomenon is associated to the aggregation of semi-micelles and micelles at higher concentrations, which can accumulate on the adsorbent surface and influence the adsorption process, as noted in other studies [50,51].

3.4. Adsorption Modelling of PFOA Using SBCT

3.4.1. Isotherm Study

Isotherm models were applied to evaluate the interaction among PFOA in aqueous solution and SBCT at equilibrium concentrations (C_{eq}) at room temperature (25 °C). The data was evaluated via a nonlinear regression model as outlined by [54], was used to construct model parameters and correlation coefficients (R^2). Experimental data and model values are presented in Figure 5, with detailed results in Table 1. The Langmuir isotherm model, which assumes adsorption is a monolayer on a homogeneous surface with finite adsorption sites [55]. The model provided the best fit to the data ($R^2 = 0.95$), with the calculated maximum adsorption capacity ($q_{\max}$) of SBCT being 9.01 mg/g, and the Langmuir constant (K_{Lq}) was 6.80 L/mg, indicating a strong affinity between SBCT and PFOA.

The adsorption process is favourable, as confirmed by the separation factor R_{Lq} of 0.18, which falls between 0 and 1. The Freundlich model [56], suitable for heterogeneous surfaces, also fit the data reasonably well ($R^2 = 0.92$), with a Freundlich constant (K_{fd}) of 7.28 and a $1/n$ value of 0.318, reflecting favorable adsorption intensity. The Temkin model is based on the assumption that the adsorption energy decreases with increasing surface area [57]. With the SBCT composite, the Temkin constants $B_{Tm} = 1.73$ and $K_{Tm} = 65.78$ L/mg, fitted with a $R^2 = 0.91$, suggest substantial interaction between PFOA and SBCT. Thus, the Langmuir model indicates that the adsorption behavior of PFOA on SBCT's monolayer is in line with other adsorbents [16,58].

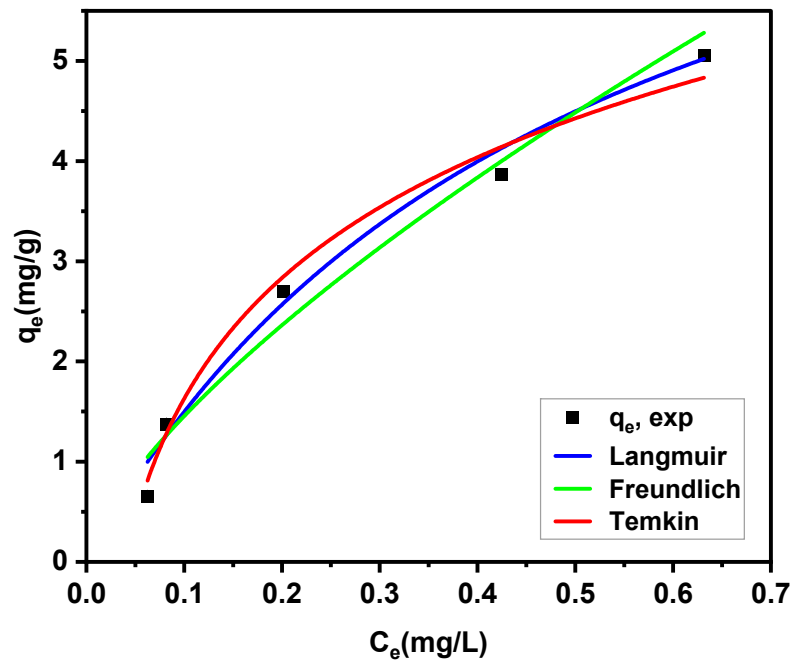


Figure 5. Adsorption isotherms for PFOA onto SBCT.

Table 1. Parameters of adsorption isotherm models.

Nonlinear Isotherm Models	Langmuir				Freundlich				Temkim		
	q_{max} (mg/g)	K_{Lq} (L/mg)	R_{Lq}	R^2	n	$1/n$	K_{fe}	R^2	B	K_{Tm} (L/mg)	R^2
SBCT	9.01	6.80	0.18	0.95	0.69	0.318	7.28	0.92	1.73	25.51	0.91

3.4.2. Kinetic Study

Three kinetic models were evaluated to understand the adsorption process of PFOA using SBCT. The parameters and the coefficient of determination were evaluated (Figure 6), and the values are summarised in Table 2. The pseudo-first-order model showed a strong correlation with the experimental data, with an R^2 value of 0.992 and a rate constant (K_1) of 0.014 hr^{-1} , indicating a good fit. The equilibrium adsorption capacity ($q_{eq} = 2.784$ mg/g) closely matches the experimental value ($q_{exp} = 2.727$ mg/g). Thus, the adsorptive mechanism is primarily driven by physisorption because the model assumes that the adsorption rate is directly proportional to the number of adsorptive sites available [36]. The R^2 value of the pseudo-second-order model was 0.981, which is slightly lower than first-order kinetics with a rate constant (K_2) of 0.004 g/mg/hr. The q_{eq} value of 3.477 mg/g derived from the second-order model is not close to the experimental values, suggesting that chemical

interactions may be a contributing factor, though not a dominant mechanism [36]. The intraparticle diffusion rate constant (k_{diff}) was 0.166 mg/g/hr, and the intercept(C) was 0.167, suggesting boundary layer resistance influences the process. The correlation coefficient ($R^2=0.90$) implies that intraparticle diffusion contributes to the overall mechanism, but not the sole rate-limiting step [59]. The observations are in line with the findings of other studies using adsorbents such as magnetic carbonized fiber [58] and chitosan-based hydrogel [43], where the pseudo-first-order model provided a superior fit, signifying that physisorption played a significant role during adsorption. Thus, SBCT proves an efficient and promising adsorbent for water purification applications, demonstrated with an effective and favorable kinetic profile for PFOA removal.

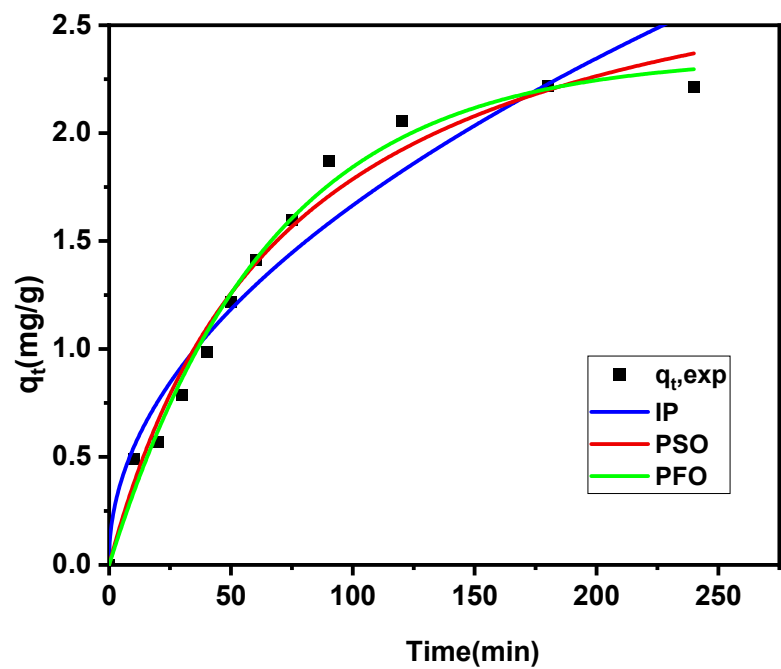


Figure 6. Adsorption kinetics for PFOA onto SBCT.

Table 2. Parameters of Kinetic Models.

Kinetic Models	First order			Second order			Intraparticle diffusion model		
Parameters	q_{eq} (mg/g)	K_1 (1/hr)	R^2	q_{eq}	K_2	R^2	K_{ipd}	C	R^2
Value	2.784	0.014	0.992	3.477	0.004	0.981	0.166	0.167	0.90

3.4.3. Thermodynamic Parameters

The change in temperature impacting the adsorption process tested across the temperature range of 20 - 40°C is presented in Table 3. The Gibbs free energy change (ΔG°) ranged between -6.65 kJ/mol and -5.72 kJ/mol, indicating that the adsorption process occurs spontaneously. The adsorption mechanism is consistent with physisorption rather than chemisorption, where the values of ΔG° are above -20 kJ/mol [37]. Further, the reaction is exothermic and physisorption-driven, supported by the low enthalpy change (ΔH°) of -21.8 kJ/mol, as chemisorption typically involves more significant heat changes (i.e., $\Delta H^\circ < -80$ kJ/mol). These thermodynamic results are consistent with kinetic data indicating a pseudo-first-order adsorption model representing physisorption. The increased disorder at the PFOA–SBCT interface, likely resulting from the release of structured water molecules as PFOA

adsorbs onto the SBCT surface, is supported by the positive entropy change (ΔS°) value of 51.3 J/mol·K. This rise in molecular randomness supports the thermodynamic favorability of the process. The PFOA adsorption proceeds spontaneously and is accompanied by heat release, suggesting it is energetically favorable and primarily driven by physical interactions demonstrated by the thermodynamic model. Studies using activated maize tassel [60] and iron-modified biochar [61] as adsorbents for PFOA removal also support this phenomenon, with negative ΔH° and positive ΔS° values indicative of favorable physisorption [51].

Table 3. Parameters of adsorption thermodynamic models.

Parameters	ΔG° (KJ mol ⁻¹)					ΔH° (KJ/mol)	ΔS° (J/mol/K)
	293°K	298°K	303°K	308°K	313°K		
Value	- 6.65±0.22	-6.67±0.26	-6.09±0.15	-5.95±0.19	-5.72±0.13	-21.8	-51.3

Note:All values denote mean ± standard deviation,.

3.5. Regeneration Experiments

The regeneration capability of the adsorbent is a very important factor in determining its long-term viability in practical use. The stability and reusability of the SBCT composite were assessed by performing five adsorption-desorption cycles using three different methanol concentrations of 50%, 75%, and 100% along with deionized water (DI). Following the initial cycle, the adsorption efficiency gradually dropped. The adsorption capacity declined up to 8% on the fifth cycle for 100% methanol and 65% for DI water, demonstrating more effective desorption and better regeneration with methanol compared to DI water. Figure 7 depicts the gradual loss of active adsorption sites with repeated use of the adsorbent. Hence, the regeneration study proved that SBCT can retain a significant amount of its adsorption efficiency through methanol-based regeneration. The composite material demonstrated its continued reusability, especially when regenerated using higher methanol concentrations, highlighting its potential for real-world practical applications [62].

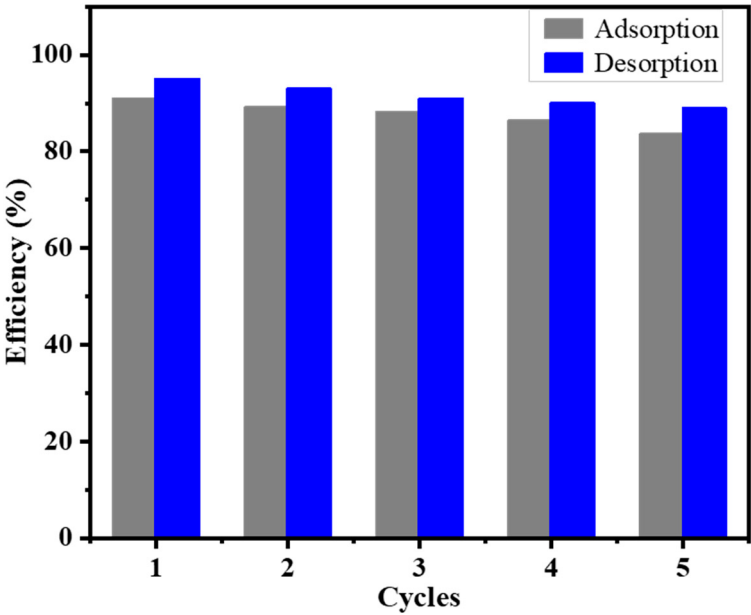


Figure 7. Regeneration and reuse of SBCT by washing with MeOH.

3.6. Comparative Analysis with Other Studies

The comparison of PFOA removal using sugarcane Bagasse Biochar/Chitosan composite (SBCT) is provided in Table 4. The initial PFOA concentrations in the different studies vary, making it difficult to compare the adsorption capacities of the SBCT composite directly with different adsorption studies. The present study investigated the performance of SBCT under relatively low concentrations ranging between 0.5 and 4 mg/L. The PFOA adsorption using adsorbents like Rice Husk Biochar [19] and Chitosan-Hydrogel [43] was evaluated at much higher concentrations of PFOA (100–2000 mg/L), naturally leading to higher adsorption capacities, which are not comparable with our results. The studies conducted at similar low initial PFOA concentrations, such as GAC ranges between 0.5 and 10 mg/L [63], Multiwalled carbon-nanotubes @ molecularly imprinted polymers (MWCNTs@MIPs) of 0.1 to 20 mg/L [62], and sulfur polymer-supported Powdered activated carbon (PAC) of 0.25 to 5 mg/L [53] is similar to our study. The SBCT developed in our study exhibited maximum adsorption capacity of 9.01 mg/g, which was comparable with MWCNTs@MIPs of 12.4 mg/g [62], and higher than sulfur-supported PAC of 0.355 mg/g [53], in addition, SBCT reached equilibrium within 4 hours, significantly faster than GAC [63] for 120 hours and comparable to MWCNTs@MIPs [62] of 2 hours, with the advantages of low-cost, eco-friendly materials and simple synthesis without the need for complex modifications like molecular imprinting or sulfur polymer support. However, the maximum PFOA adsorption capacities will differ based on the testing conditions. The novel engineered SBCT composite stands out by combining environmental sustainability, fast kinetics, and effective PFOA removal even at low concentrations.

Table 4. Comparison of adsorption kinetics and adsorption capacities of various adsorbents.

Adsorbent	Initial concentrationC ₀ (mg/L)	pH	Equilibrium time t _{eq} (h)	Equilibrium adsorption capacity q _{eq} (mg/g)	Maximum adsorption capacity q _{max} (mg/g)/ Isotherm model	Reference
Rice husk BC	100 - 200	5	5	550	1085/L _q	[19]
Multi-walled carbon nanotubes (MWCNT)	10-500	5	4	12.4	5.4 /F _e	[64]
Bamboo AC	20-250	5.0	24	576	544/F _e	[65]
Starch-stabilized Fe ₃ O ₄ nanoparticles	0-4	6.8	0.5	15.53	62.5/L _q	[66]
Granular activated carbon (GAC)	0.5-10	5	120	12.6	52.8/L _q	[63]
Chitosan + hydrogel	100-2000	-	< 24	1050.83	1275.9/L _q - F _e	[43]
Sulfur polymer support activated carbon	0.250 –5	5	-	-	0.355 / L _q -F _e	[53]
Magnetic carbonized fiber	25 –250	3	2	41.85	204.7/L _q	[58]
Hierarchically microporous biochar (HMB)	50–150	2-10	0.5	423	1269/L _q	[67]

Calotropis Gigantea fiber	25–250	-	3	54.76	232.8/L _q	[68]
Polyurethane-modified magnetic sorbent	0.05-1	5.5	46	243.9(μg/g)	2.480/Sips	[69]
Multi-walled carbon nanotubes @Molecular imprinted polymers (MWCNTs@MIPs)	0.10–20	5	2	1.44	12.4/L _q	[62]
Sugarcane bagasse biochar/Chitosan composite (SBCT)	0.5-4	4	4	5.1	9.01/L _q	Our study

Note: L_q– Langmuir Isotherm F_e– Freundlich Isotherm L_q- F_e - Langmuir Freundlich Isotherm.

3.7. Limitations and a Sustainable Way Forward

The sustainable, cost-effective SBCT composite provides a solution for eliminating PFOA from water, where traditional treatment technologies are inaccessible or economically impractical, and is suitable for use in developing economies. Such an adsorbent can be used as a good filter unit near the open drains or sewer outlets before it reaches the water bodies from the decentralized treatment system. However, several limitations must be addressed for widespread application. If not properly managed, saturated composites may pose environmental risks and serve as a secondary pollution source. The post-treatment of the adsorbent is necessary because the adsorption technology can only transfer PFOA from aqueous media to the solid phase. The application of advanced oxidation processes (AOPs) such as UV/H₂O₂ or ozone treatments enables complete degradation of adsorbed contaminants by promoting sustainable reuse or safe disposal, and helps to address the disadvantages associated with the composite disposal. The performance of the composite may be hindered in complex wastewater matrices due to the presence of competing ions and organic matter, which can be avoided by further field studies. The proper management and regeneration are also required due to finite adsorption capacity. Therefore, optimizing adsorbent recovery, integrating AOPs for contaminant destruction, and developing cost-effective regeneration protocols to ensure environmental safety and economic feasibility are necessary for the successful implementation of treatment.

4. Conclusions

We designed and prepared engineered biochar composite-SBCT for the effective removal of PFOA, which is effective even at a very low concentration (0.5mg/L) from aqueous media. The SBCT composite achieved enhanced PFOA adsorption capacities under varied environmental conditions, outperforming many conventional materials. The removal of PFOA was effective under acidic pH conditions. The adsorption mechanism of PFOA on SBCT is further evaluated using an isotherm study and kinetic modeling, demonstrating favorable alignment with pseudo-first-order kinetics and the Langmuir isotherm model with a maximum adsorption capacity of 9.01 mg/g. Hence, SBCT composite supports the applicability as a low-cost and sustainable adsorbent for decentralized water treatment systems, with potential for further scale-up and process optimization.

Supplementary Materials: The following supporting information can be downloaded at the website of this paper posted on Preprints.org.

Author Contributions: K Pavithra: Methodology, Data curation, Software, Formal analysis, Writing: original draft; Paromita Chakraborty: Fund acquisition, Conceptualization, Methodology, Supervision, Writing: Review and editing.

Funding: K Pavithra would like to acknowledge the support of the **CSIR-Senior Research Fellowship** for the financial support (**CSIR Fellowship/141-4354-8502/2K23/1**). Paromita Chakraborty was supported by the India–Norway cooperation project on capacity building for reducing plastic and chemical pollution in India (**INOPOL Project No: 220260, IND-22/0007**).

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 have no competing interests.

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