

Article

Evaluation of *Annona diversifolia* Seed Extract as A Natural Coagulant for Water Treatment

Ibrahim Muntaqa Tijjani Usman ^{1,2}, Foo-Wei Lee ^{3,*}, Yeek-Chia Ho ^{1,*}, Han-Ping Khaw ¹, Qi-Wen Chong ¹, Yong-Ming Kee ¹, Jun-Wei Lim ⁴ and Pau-Loke Show ^{5,6,7,8}

- ¹ Centre for Urban Resource Sustainability, Institute of Self-Sustainable Building, Civil and Environmental Engineering Department, Universiti Teknologi PETRONAS, Seri Iskandar 32610, Perak Darul Ridzuan, Malaysia
 - ² Agricultural and Environmental Engineering Department, Faculty of Engineering, Bayero University Kano, Kano 700241, Nigeria
 - ³ Department of Civil Engineering, Lee Kong Chian Faculty of Engineering & Science, Universiti Tunku Abdul Rahman, Sungai Long Campus, Jalan Sungai Long, Bandar Sungai Long, Kajang 43200, Selangor, Malaysia
 - ⁴ HICoE-Centre for Biofuel and Biochemical Research, Institute of Self-Sustainable Building, Department of Fundamental and Applied Sciences, Universiti Teknologi PETRONAS, Seri Iskandar 32610, Perak Darul Ridzuan, Malaysia
 - ⁵ Department of Chemical Engineering, Khalifa University, Shakhbout Bin Sultan St—Zone 1, Abu Dhabi P.O. Box 127788, United Arab Emirates
 - ⁶ Zhejiang Provincial Key Laboratory for Subtropical Water Environment and Marine Biological Resources Protection, Wenzhou University, Wenzhou 325035, China
 - ⁷ Department of Chemical and Environmental Engineering, Faculty of Science and Engineering, University of Nottingham Malaysia, Jalan Broga, Semenyih 43500, Selangor Darul Ehsan, Malaysia
 - ⁸ Department of Sustainable Engineering, Saveetha School of Engineering, SIMATS, Chennai 602105, India
- * Correspondence: leefw@utar.edu.my (F.-W.L.); yeekchia.ho@utp.edu.my (Y.-C.H.)

Abstract: The ever-present environmental crises are current research hotspots. Nature-based solutions have been shown to have multiple co-benefits toward solving these crises. Plant-based coagulants are known to be a cost-effective and environmentally friendly approach for coagulation and flocculation processes for drinking-water treatment. In this study, a natural coagulant was extracted from *Annona diversifolia* seed, and its effectiveness was investigated for turbidity reduction using jar test in kaolin suspension, river water, and evaluation of factors for sludge dewatering. The characterisation studies of *Annona diversifolia* seed extract were carried out using techniques including Fourier-transform infrared spectroscopy (FTIR), zeta potential analyser, and transmission electron microscopy (TEM). Response surface methodology (RSM) was also performed for the optimisation study. The results from FTIR showed that *Annona diversifolia* seed extract contains carboxyl and hydroxyl functional groups. The charge density was found to be negative. A web-like structure surface morphology was observed from TEM. The optimum treatment settings were found to be at pH 3 and a dosage of 25 mg/L for water treatment, and 50 mg/L for sludge dewatering, which were comparable to the metal-salts coagulants. *Annona diversifolia* seed extract has been shown to be a good natural coagulant. Further research can be conducted to modify and enhance its performance.

Keywords: *Annona diversifolia* seed extract; natural coagulant-aid; water treatment; sludge dewatering



Citation: Tijjani Usman, I.M.; Lee, F.-W.; Ho, Y.-C.; Khaw, H.-P.; Chong, Q.-W.; Kee, Y.-M.; Lim, J.-W.; Show, P.-L. Evaluation of *Annona diversifolia* Seed Extract as A Natural Coagulant for Water Treatment. *Sustainability* **2023**, *15*, 6324. <https://doi.org/10.3390/su15076324>

Academic Editor: Luca Di Palma

Received: 28 February 2023

Revised: 27 March 2023

Accepted: 28 March 2023

Published: 6 April 2023



Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Nature-based solutions (NBS) are among the fundamental pillars of solving environmental crises and climate change [1]. They are regarded as low-cost solutions compared to conventional methods. The use of NBS is known to be beneficial in improving human, social, and economic health in a highly effective manner [2,3]. As diverse as NBS could be, within the context of this article, the use of NBS can be defined as a means of using nature-based materials in aiding water and wastewater treatment.

According to the European Union (EU) environment, multiple benefits can be achieved from the use of NBS. One of the promises of NBS is its promotion towards delivering multiple sustainability goals synergistically [4,5]. From the 2022 Sustainable Development Goals (SDG) report, one can observe that meeting clean water and environment to achieve 2030 goals will require a quadruple increase in the rate of progress. Moreover, the report indicated that despite the increase in safely managed clean water supply, only 81% of world coverage will be achieved by 2030 [6]. This means that approximately 1.6 billion people will be without a safely managed water supply. Furthermore, agricultural and untreated wastewater have been identified to be the two biggest threats to environmental water quality worldwide. Their discharge introduces excess pollutants into waterbodies including groundwater, which upsets the ecosystems' functionality. With an integrated monitoring system and the use of NBS, water quality issues will be minimised, and a clean water supply could be achieved worldwide, thus fulfilling SDG 6 [7,8].

The general assumption is that environmental pollution remediation, including that of water and wastewater, requires technological solutions stands; however, researchers have shown that NBS and technology can be powerful allies [4,9,10]. Notwithstanding, this needs science-based proof as evidence of NBS performance efficiencies. In view of sustainable water and wastewater treatment, the use of nature-based materials could contribute tremendously to aiding the coagulation and flocculation processes, thus minimising the use of chemicals and the production of toxic sludges from water and wastewater treatment plants.

The coagulation and flocculation process (CFP) has been identified as the most efficient and reliable method for water and wastewater treatment and it requires low energy and time consumption during operation [11,12]. The CFP is highly effective in the removal of suspended solids from wastewater and in sludge dewatering [13]. The process is achieved through the addition of coagulants into the wastewater during the treatment. These coagulants can be chemical-based or nature-based. The conventional chemicals commonly used alongside their aids include aluminium salts and ferric salts, and synthetic polymers, respectively [14]. Despite their wide applicability, they have exhibited some shortcomings such as ineffectiveness in low-temperature waters, production of large quantities of sludge, affecting the pH of treated water, high procurement costs, and also harmful effects on human health due to the presence of the chemical residuals in treated water [15]. The application of synthetic polymers such as polyacrylamide derivatives and polyethyleneimines has also been reported to pose some environmental problems. Many of these chemicals are expensive and non-biodegradable and their residual monomers are neurotoxic and carcinogenic [16]. Therefore, it is desirable to replace these chemical coagulants and their aids with natural alternatives which can be extracted from either plants, microorganisms, or animal waste [17].

Plant-based natural coagulants are found to be the most desirable of all-natural coagulants [18,19]. The use of plant-based natural coagulants dates back centuries in traditional wastewater treatment. Various plant species have been identified to be extracted for producing plant-based coagulants. Nirmali seeds, *Moringa oleifera* seeds, and opuntia ficus-indica cactus are considered the most common natural coagulants [20–23]. The seeds of *Feronia limonia*, *Carica papaya*, *Prunus armeniaca*, *Persea americana*, *Mangifera indica*, and the peels of *Citrus sinensis* have been applied for treating turbid synthetic water and wastewater. A turbidity reduction of more than 80% has been reported to be achieved using fruit waste coagulants [21,24–28]. The use of plant-based coagulants has been found to produce less voluminous sludge, which is the most important step to reduce the overall cost of a sewage treatment plant operation [29,30]. Another tropical fruit tree is *Annona diversifolia*: it has fruits of different shapes and contains a lot of seeds.

Annona diversifolia seed extract has been found to contain polysaccharides that are capable of reducing chemical oxygen demand (COD) and turbidity in wastewater [31]. The seed extracts have been shown to possess functional groups of carboxyl and hydroxyl groups that are highly associated with the flocculating ability of plant materials [18,19,23]. Since few studies have focused on the use of *Annona diversifolia* seed extract as a coagulant

in wastewater treatment, the objective of this study is to evaluate the capability of *Annona diversifolia* seed extract for water treatment and sludge dewatering.

2. Materials and Methods

2.1. Materials

Annona diversifolia seeds were collected from a local market in Perak, Malaysia. River water samples were collected from the Parit Water Treatment plant in Perak, Malaysia, and stored in a cold room at 4 °C. A 500 mL BEHR Soxhlet Extractor was used for extraction at the Environmental Engineering laboratory Universiti Teknologi PETRONAS. An activated sludge sample was collected from the Sewage Treatment Plant (STP) located at Universiti Teknologi PETRONAS. Sigma Aldrich supplied analytical-reagent-grade sodium hydroxide (NaOH), hydrochloride acid (HCl) and kaolin powder with 100% purity, and pH 6–7.

2.2. Methods

2.2.1. Extraction and Characterisation of *Annona diversifolia* Seed

The seeds of *Annona diversifolia* were washed with tap water. The seeds were kept at room temperature to dry. The seed kernels were ground to a fine powder using an ordinary blender. The seed powder was then extracted in a Soxhlet extractor in the presence of hexane (60–80 °C for 6 h) to remove the fat. The defatted powder was dried in an oven overnight at 50 °C. The final products were stored in an air-tight container at room temperature until they were used.

Fourier-transform infrared spectroscopy (FTIR, Spectrum one, Perkin Elmer-Waltham, MA, USA) was used to determine the functional groups of *Annona diversifolia* seed extract (ADE). The operation scanning ranged from 4000 cm⁻¹ to 400 cm⁻¹. The charge density and particle size of the ADE were determined using a Zeta Potential Analyser (Zetasizer Nano, S90, Malvern, UK). Transmission electron microscopy (TEM) of the ADE and formed flocs were determined to identify the morphology, size, shape, and microstructure of the materials using Zeiss Libra 200 FE.

2.2.2. Coagulation and Flocculation Assay in Kaolin Suspension and River Water

Synthetic turbid water (kaolin suspension) for the jar test was prepared by adding kaolin to distilled water with a ratio of 0.15 g: 1 L. The measurement of pH and turbidity of the turbid water was performed using a portable pH meter (HACH Sension 1) and a turbidity meter (HACH 2100Q), respectively. The initial turbidity of kaolin suspension was at approximately 200 NTU. Analytical-reagent-grade sodium hydroxide (NaOH), and hydrochloride acid (HCl) were used to adjust the pH of the suspension.

Jar test apparatus was used to evaluate the efficiency of coagulants in kaolin suspension and river water according to the standard method jar test ASTM D2035-19 [32]. The jar test apparatus accommodates a series of six beakers with six spindle steel paddles. The working volume was 1 L. The pH and ADE coagulant dosage were adjusted for each beaker based on the experimental designs. The samples were then stirred with rapid mixing (150 rpm) for 3 min followed by slow mixing (25 rpm) for 15 min. Lastly, the samples were left standing still for 30 min. The supernatant was withdrawn using a pipette for final measurements. After the coagulation–flocculation process, the initial and final turbidities were measured, and the percentage of turbidity reduction was calculated according to Equation (1) [33].

$$\text{Turbidityreduction}(\%) = \frac{\text{InitialTurbidity} - \text{FinalTurbidity}}{\text{InitialTurbidity}} \times 100 \quad (1)$$

A factorial design was conducted to determine the influencing factors using Design Expert 7 software. The range for kaolin suspension was at pH 3 and 10, whilst the dosage was between 4 mg/L and 20 mg/L. On the other hand, the range for river water was between pH 3 and 9, whilst the dosage was between 0.1 mg/L and 20 mg/L.

2.2.3. Preparation and Characterisation of Activated Sludge

The pH, moisture content, total solids, total suspended solids, and volatile suspended solids of the bio-solid were measured following the APHA method. The preliminary analysis of biosolids showed a pH of 6.1, moisture content of 98.5%, total solid of 14,600 mg/L, total suspended solid of 31,240 mg/L, and volatile suspended solid of 10,813 mg/L.

2.2.4. Sludge Dewaterability Assay in Sewage Sludge

The time to filter (TTF) method (APHA method 2710H) was applied to measure sludge dewaterability as a simple, rapid, and inexpensive technique. In this method, a Buchner funnel with a Whatman filter paper was placed above a graduated cylinder. A total of 200 mL of sludge was poured into the funnel and vacuumed into the cylinder using a vacuum pump providing a constant vacuum of 51 kPa. The time required for collecting 100 mL of sample in the graduated cylinder was recorded using a stopwatch. The recorded time shows the efficiency of coagulants in the dewatering of sludge.

2.2.5. Response Surface Methodology

Response surface methodology (RSM) was employed to conduct optimisation [34–37]. Central composite design (CCD) was applied to build a second-order (quadratic) model to evaluate the combined impacts of independent variables on studied responses. The fitness of the quadratic model was expressed by the coefficient of determination (R^2). Analysis of variance (ANOVA) was used to analyse the statistical significance of the model at $p < 0.10$.

3. Results

3.1. Extraction and Characteristics of *Annona diversifolia* Seed Extract (ADE)

The results of FTIR analysis for *Annona diversifolia* seed extract (ADE) are presented in Figure 1.

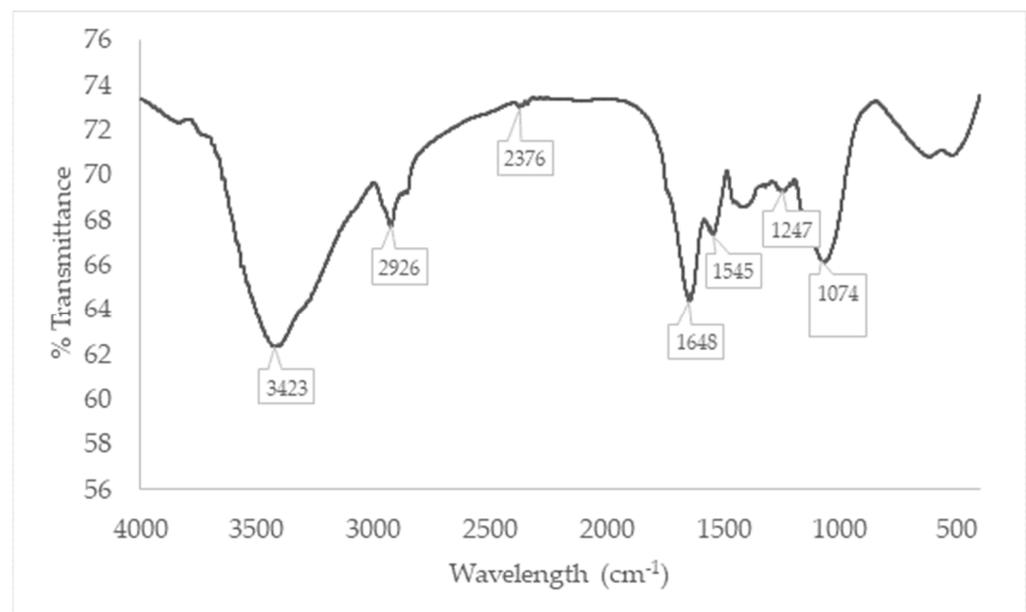


Figure 1. Fourier-transform infrared spectroscopy (FTIR) spectra of *Annona diversifolia* seed extract (ADE).

The spectrum shows the presence of hydroxyl (-OH) group at approximately 3423 cm^{-1} , medium peaks of amine group including N-H at 1648 cm^{-1} and C-N at 1074 cm^{-1} , and also a weaker symmetric stretching of carboxyl (COO^-) group at 1545 cm^{-1} . ADE can undergo negative charge-assisted H-bond interaction with their oxygen-containing functional groups. Table 1 presents the particle size and zeta potential of ADE. The charge was found to be negative.

Table 1. Particle size (nm) and zeta potential (mV) of ADE coagulant.

Record No.	Size (nm)	Zeta Potential (mV)
Average	193.8	−27.0

The images from the transmission electron microscope (TEM) analysis of ADE and formed flocs are shown in Figure 2.

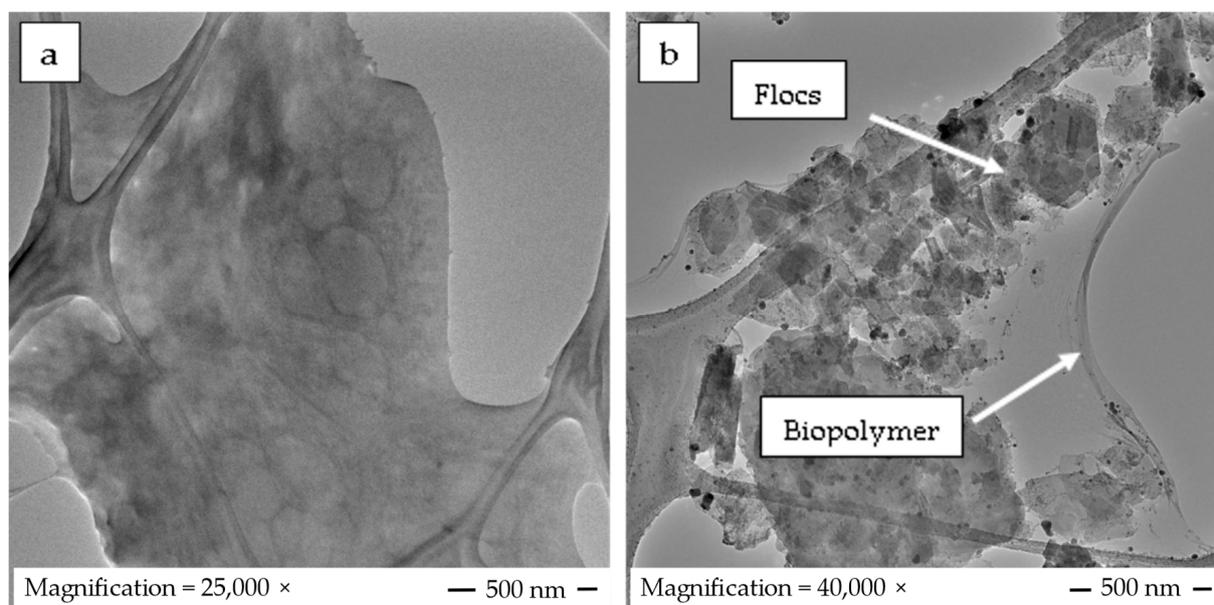


Figure 2. Transmission electron microscope (TEM) analysis of (a) *Annona diversifolia* seed extract (ADE), and (b) formed flocs.

The coagulant image showed a web-like structure at a magnification of 25,000, while the flocs aggregate was shown under a magnification of 40,000. ADE may function as a web network to trap the particles by adsorbing them on its surface.

3.2. Coagulation and Flocculation Performance of *Annona diversifolia* Seed Extract (ADE)

From the jar test, results for the performance of *Annona Diversifolia* seed extract (ADE) and FeCl_3 using kaolin suspension and river water are presented in Figures 3 and 4, respectively.

3.3. Sludge Dewatering at Varying pH and *Annona diversifolia* Seed Extract (ADE)

The variation in the time to filter (TTF) with different pH values and *Annona diversifolia* seed extract (ADE) coagulant dosages is presented in Figure 5. The results revealed that TTF significantly decreased with decreasing pH. Rapid dewaterability occurred (6 min) with an application of 50 mg/L of the coagulant at pH 3. In acidic pH, the concentration of positive charges increases in the sludge, which attracts negatively charged molecules, i.e., sewage sludge particles and ADE coagulant. A polymer bridge is formed when two or more particles are adsorbed on the surface of the polymer. The formed polymer bridge adsorbs particles from sludge and increases the floc sizes, which improves the sludge dewatering [38,39].

3.4. Screening and Optimisation of Process Factors Using Factorial Design and Response Surface Methodology

3.4.1. Screening Process

The outcomes of the screening process of the obtained results from the coagulation tests with kaolin synthetic wastewater using ADE or FeCl_3 , and with the river water using

ADE or alum were stored in the software. After storing the range of factors in the software, nine conditions were developed by the software, and the experiments were conducted based on the factorial design. In Table 2, ANOVA shows that pH, coagulant dosage and their interactions are significant ($p < 0.10$) in the coagulation and flocculation processes.

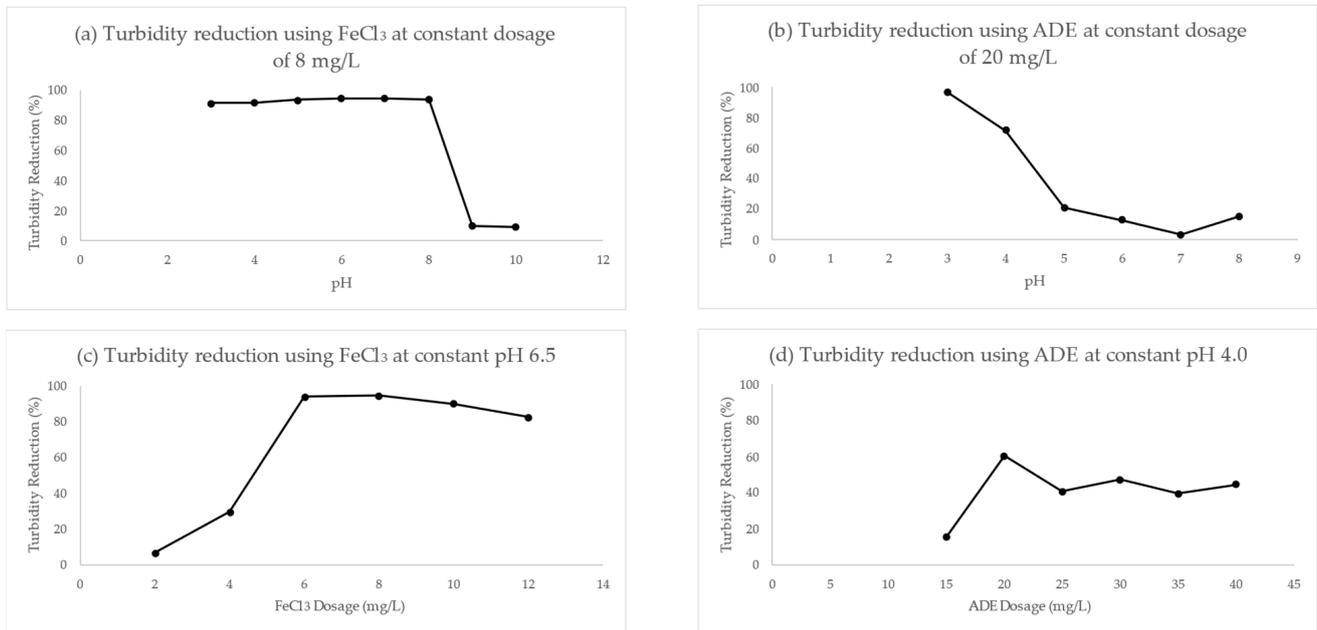


Figure 3. The amount of turbidity reduction percentage in kaolin synthetic wastewater supplied with varying pH and fixed coagulant dosage of FeCl₃ (8 mg/L) and *Annona diversifolia* seed extract (ADE) (20 mg/L) (a,b), and in the presence of a varying dosages of FeCl₃ and ADE at fixed pH of 6.5 and 4.0, respectively (c,d).

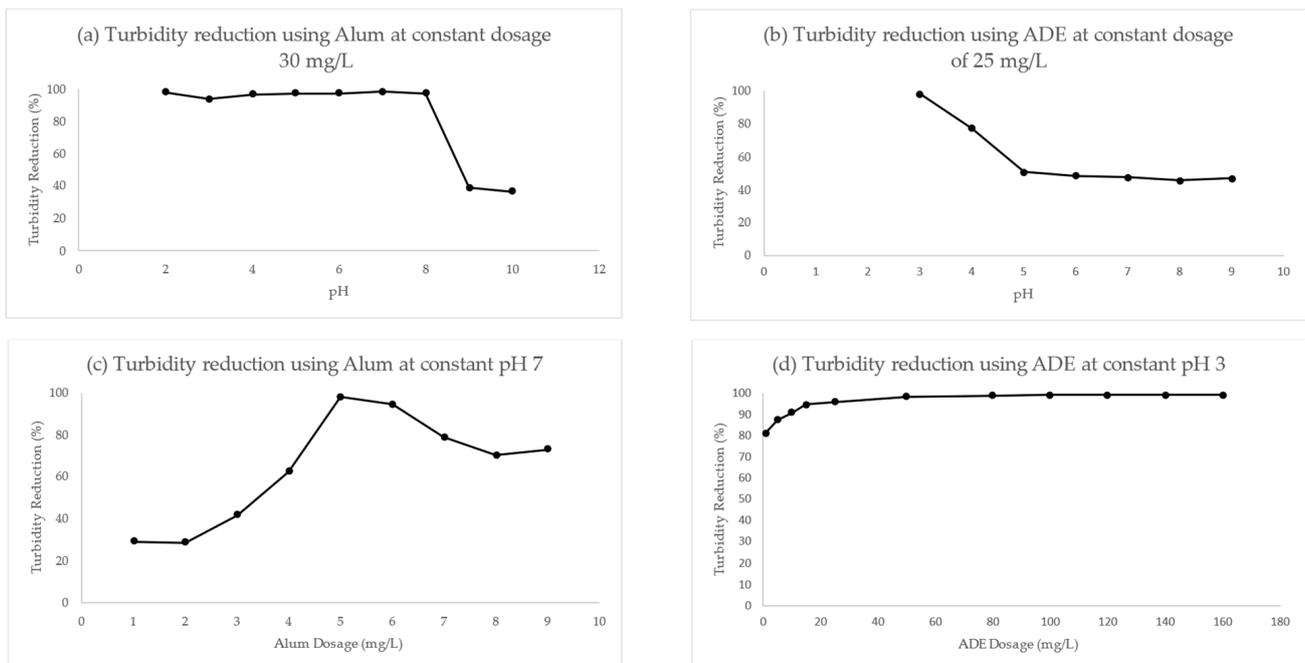


Figure 4. The amount of turbidity reduction percentage in river water supplied with varying pH and fixed coagulant dosage of Alum (30 mg/L) and *Annona diversifolia* seed extract (ADE) (25 mg/L) (a,b), and in the presence of a varying dosages of Alum and ADE at fixed pH of 7 and 3, respectively (c,d).

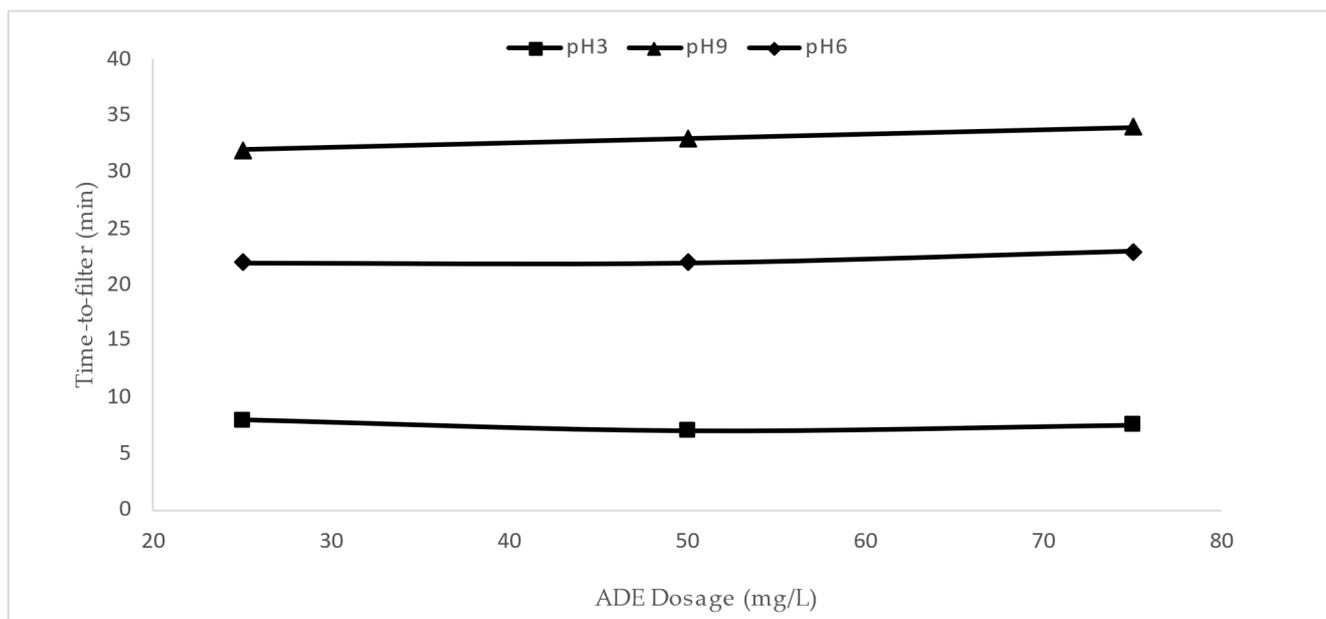


Figure 5. Time to filter that is required for sludge dewaterability with the application of different *Annona diversifolia* seed extract (ADE) coagulant dosages and pH.

Table 2. Analysis of variance (ANOVA) of data obtained from the screening process related to kaolin synthetic wastewater, and river water experiments.

Wastewater	Coagulants	Source	Sum of Squares	Mean Square	F Value	p-Value
Kaolin synthetic wastewater	<i>Annona diversifolia</i> seed extract (ADE) coagulant	pH	6821.93	6821.93	723.42	<0.0001
		Dosage	80.91	80.91	8.58	0.0428
		pH × Dosage	56.18	56.18	5.96	0.0712
	FeCl ₃	pH	4823.3	4823.3	14,483.52	<0.0001
		Dosage	11.22	11.22	33.7	0.0044
		pH × Dosage	22.56	22.56	67.75	0.0012
River water	ADE coagulant	pH	5061.61	5061.61	2532.05	<0.0001
		Dosage	33.35	33.35	16.68	0.0150
		pH × Dosage	29.43	29.43	14.72	0.0185
	Alum	pH	1274.13	1274.13	7395.71	<0.0001
		Dosage	1370.85	1370.85	7957.11	<0.0001
		pH × Dosage	1284.86	1284.86	7458.00	<0.0001

3.4.2. Optimisation Process

The relationship between the independent variables and the responses was analysed using RSM. The central composite design (CCD) matrix and turbidity reduction percentage related to the application of the experimental coagulants were determined and the ANOVA results for the model terms and predicted models are presented in Table 3.

The results of coagulant application in kaolin synthetic water showed that pH and coagulant dosage are significant factors ($p < 0.10$) in the coagulation process with the application of ADE; however, pH was found to be the only significantly effective factor when FeCl₃ was applied. This can be due to the chosen experimental range for the FeCl₃ dosage. It is presumed that a wider range of FeCl₃ coagulant dosages can lead to more reliable results. On the other hand, the data related to river water showed that pH and

coagulant dosage are significant factors in coagulation and flocculation processes using either ADE or alum as coagulants.

Table 3. Analysis of variance (ANOVA) of data obtained from optimisation process related to kaolin synthetic wastewater, and river water experiments.

Wastewater	Coagulants	Source	Sum of Squares	Mean Square	F Value	p-Value
Kaolin synthetic wastewater	<i>Annona diversifolia</i> seed extract (ADE) coagulant	pH	8317.18	8317.18	458.05	<0.0001
		Dosage	93.54	93.54	5.15	0.0575
		pH × Dosage	30.20	30.20	1.66	0.0382
		pH × pH	4360.46	4360.46	240.14	<0.0001
		Dosage × Dosage	33.52	33.52	1.85	0.2164
	FeCl ₃	pH	2950.38	2950.38	21.46	0.0012
		Dosage	40.75	40.75	0.30	0.5994
		pH × Dosage	1051.06	1051.06	7.65	0.0219
		pH × pH	832.55	832.55	1.66	0.2386
		Dosage × Dosage	1046.13	1046.13	2.09	0.1920
River water	ADE coagulant	pH	7702.73	7702.73	5269.33	<0.0001
		Dosage	46.26	46.26	31.65	0.0008
		pH × Dosage	29.43	29.43	20.13	0.0028
		pH × pH	1933.70	1933.70	1355.60	<0.0001
		Dosage × Dosage	0.43	0.43	0.30	0.6032
	Alum	pH	3202.12	3202.12	50.34	0.0002
		Dosage	657.52	657.52	10.34	0.0148
		pH × Dosage	871.43	871.43	13.7	0.0076
		pH × pH	884.52	884.52	13.91	0.0074
		Dosage × Dosage	357.32	357.32	5.62	0.0496

Significant model terms contributed to developing well-fitted predicted models where the turbidity reduction percentage as the responses (y) was a function of pH (x_1) and coagulant dosage (x_2). Each model was calculated as the sum of the intercept coefficient, two linear (x_1 and x_2), two quadratic (x_1^2 and x_2^2), and one interaction effect (x_1x_2), as shown in Table 4. The high values of R^2 indicated that the data fitted adequately to the second-order polynomial models for all the responses in all experiments.

Table 4. The predicted models for the dependent variables.

Wastewater	Coagulants	Quadratic Equation	R ²
Kaolin synthetic wastewater	ADE coagulant	$y = 6.04 - 37.23x_1 - 3.95x_2 + 2.75x_1x_2 + 39.73x_1^2 + 3.48x_2^2$	0.9910
	FeCl ₃	$y = 75.80 - 36.37x_1 + 9.93x_2 + 0.73x_1x_2 - 17.36x_1^2 - 19.46x_2^2$	0.8658
River water	ADE coagulant	$y = 36.36 - 35.83x_1 - 2.78x_2 - 2.71x_1x_2 + 26.79x_1^2 + 0.40x_2^2$	0.9990
	Alum	$y = 85.41 - 23.10x_1 + 10.47x_2 - 14.76x_1x_2 - 17.90x_1^2 + 11.37x_2^2$	0.9723

The influence of independent variables on the turbidity reduction percentage can be found based on the ANOVA results for model terms (see Table 3) and the response surface plots. Figure 6a,b presents the contour plots obtained from the application of ADE and FeCl₃, respectively.

The response surface contour plots detected optimal areas where maximum responses are achieved by proper values of the tested experimental factors. According to the graphs in Figure 6a,b, ADE was more effective at pH 3 and a dosage of 15 mg/L, while FeCl₃ performed best at pH 4 and a dosage of 12 mg/L.

The pH has an important role in the coagulation process: it affects the electrical potential gradient on the surface of the particle. During the coagulation and flocculation process, the added dosage of *Annona diversifolia* seed extract (ADE) coagulant in the water destabilizes the negative surface of particles and promotes the London–Van der Waals force of attraction between the particles, which leads to aggregation [11]. At a high pH, the concentration of H⁺ decreases. Negatively charged functional groups of ADE (hydroxyl (OH⁻) and carboxyl (COO⁻)), and the negatively charged kaolin surface create a repulsive force that allows the coagulant molecules to be extended and to produce loops and tails

to promote bridging mechanisms and large open-structure flocs [40]. Figure 7 shows the schematic diagram of how adsorption and bridging mechanisms occur.

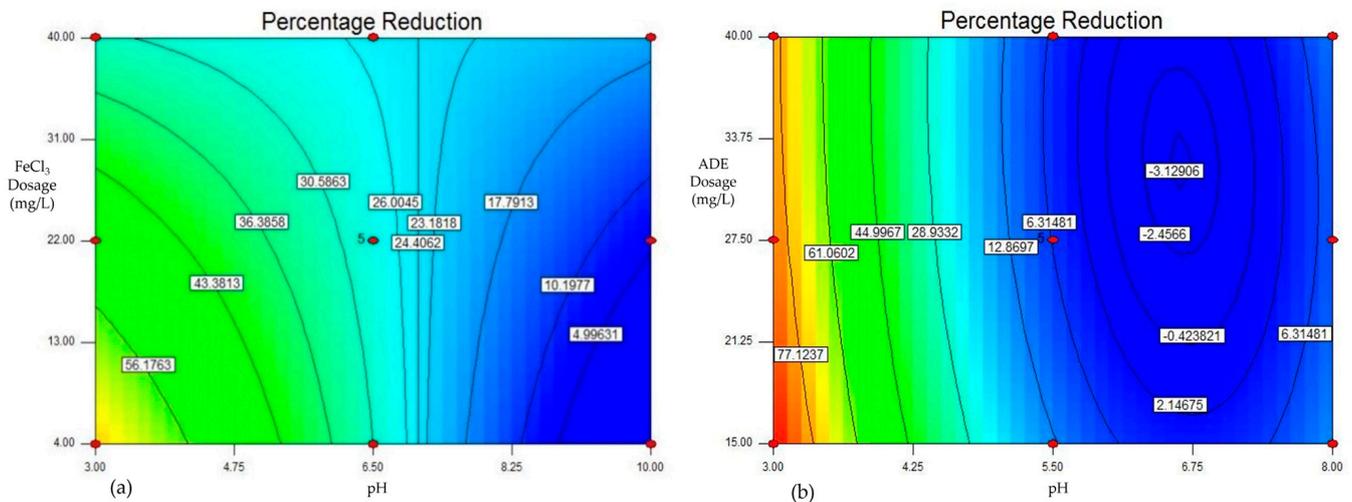


Figure 6. (a) Contour plot for turbidity reduction percentage affected by *Annona diversifolia* seed extract (ADE) coagulant; (b) contour plot for turbidity reduction percentage affected by FeCl_3 .

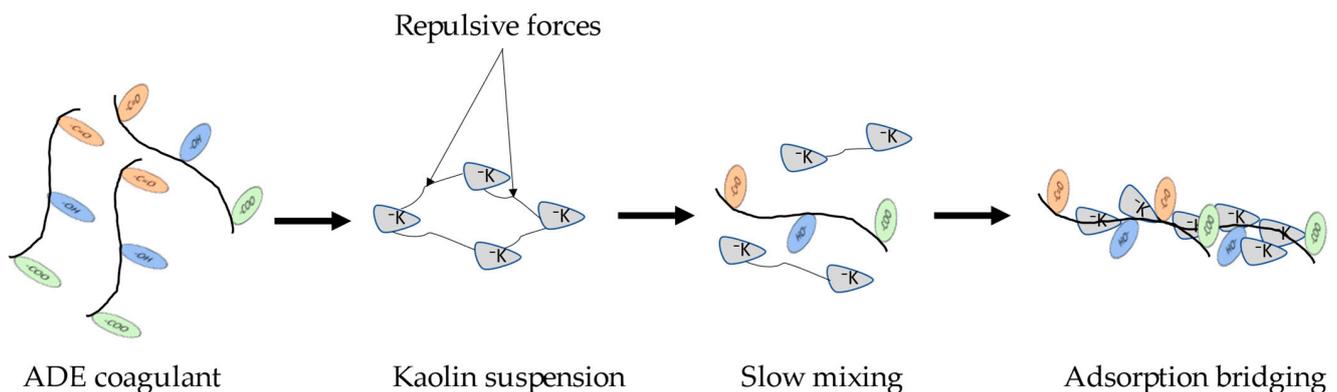


Figure 7. Schematic diagram of coagulant adsorption and bridging mechanisms.

The anionic coagulant polymer is then attracted to the positive side of destabilized particles and forms flocs [26,38]. The numerical optimisation tool of design expert software predicted the optimum values of the independent variables to maximize the coagulants' turbidity removal ability. The actual and predicted values of turbidity reduction percentage are listed in Table 5. Based on the data, the actual amounts were found to be in close similarity and strong agreement with the predicted values for all percentages of turbidity reduction.

Nature-based coagulants are highlighted as good alternatives to conventional chemical coagulants for water and wastewater treatments. However, several limitations have been associated with their implementation on a larger scale. In light of plant-based natural coagulants, major disadvantages have been identified including thermo-sensitivity, pH sensitivity, a loss of viscosity during storage, and their vulnerability to microbial contamination [41]. A lack of rigorous investigation into their resilience to the biological environment has been outlined [42,43]. Even though plant-based coagulants are often associated with multiple benefits, only the benefits within the focus of the research are considered, resulting in the side-lining of a co-benefits investigation. However, this may be due to the difficulty in monetizing the co-benefits and to the high uncertainty of market values [44–46].

Additionally, large knowledge gaps in terms of the long-term effectiveness of plant-based coagulants as regards environmental impact have been reported [43]. This was incorporated with inadequate evidence on how their usage will positively affect human

health due to long-term consumption. Another main problem with the present research is the lack of investment in nature-based solutions (NBS). Despite research evidence that proves the benefits of NBS to both humans and the environment, insufficient investments are being made toward their implementation [47–51]. For example, researchers have shown how modified plant-based natural coagulants could serve as good alternatives to chemical-based coagulants [52]. They are capable of working 70% more than conventional coagulants. Sludges produced from the use of plant-based natural coagulants are regarded as less voluminous, biodegradable and biocompatible; therefore, they reduce water waste as sludge and can be used for agricultural soil amendments [24]. Notwithstanding, research on nature-based solutions is strongly underfunded, and this lack of financing has been identified as an obstacle to the application of nature-based solutions worldwide [49,53–55].

Table 5. The actual and predicted values of turbidity reduction percentage for kaolin synthetic wastewater and river water.

Wastewater	Coagulants	pH	Dosage (mg/L)		Predicted Percentage of Turbidity Reduction (%)	Actual Percentage of Turbidity Reduction (%)
Kaolin synthetic wastewater	<i>Annona diversifolia</i> seed extract (ADE)	1	3.89	33.79	44.48	45.34
		2	4.11	27.38	38.89	37.87
		3	4.38	25.67	31.52	32.89
	FeCl ₃	1	4.34	13.50	83.16	83.90
		2	3.18	11.21	92.27	91.54
		3	4.37	12.56	81.79	80.98
River water	ADE	1	3.09	16.61	95.62	98.40
		2	3.47	16.52	84.94	85.81
		3	3.19	11.95	92.72	92.88
	Alum	1	3.12	21.63	93.63	97.95
		2	3.57	21.63	94.18	98.40
		3	4.24	29.87	96.37	98.83

The future perspectives on the use of nature-based solutions will first require a systemic state-of-the-art change in the way we conduct research and run academic and industrial institutions. Research to enhance the efficiency of promising plant-based materials should be encouraged. Modification using grafting copolymerisation for example has shown to be highly effective, particularly in altering and enhancing the coagulation and flocculation potentials of natural materials. More investigations need to be conducted on other co-benefits of plant-based coagulants, including their potential as disinfectants, their biodegradability and biocompatibility, and their positive benefits to human health. Pilot studies need to be designed and run to enable the scaling up of positive research, thus actualising plant-based natural coagulants as an alternative to chemical coagulants.

4. Conclusions

In this study, the use of nature-based solutions and their benefits were introduced, and a natural coagulant was extracted from the *Annona diversifolia* seed extract (ADE), with its effectiveness being evaluated for water treatment and sludge dewatering. Three experiments were conducted to determine the optimum coagulant dosage and pH for turbidity reduction in kaolin synthetic wastewater and river water, and for time-to-filter values in sludge dewatering. FeCl₃ and alum coagulants were used with similar synthetic wastewater and river water for comparison purposes. The optimisation of independent variables was conducted to determine the optimum conditions for the best treatment performance.

It was concluded that the *Annona diversifolia* seed extract (ADE) coagulant works well in acidic conditions (pH 3) with a concentration of 25 mg/L for water treatment and 50 mg/L for sludge dewatering. The coagulation capability of ADE was comparable to the metal-salts coagulants.

Further research can be conducted to determine ADE coagulant ability for heavy metal removal from industrial effluents and sludges. Interaction between the surface charge and the pH of a coagulant could be investigated in future studies. Further modifications can be conducted to enhance the coagulation and flocculation properties of *Annona diversifolia* seed extract. Similarly, research on different plant waste can be conducted to maximize the possibility of establishing nature-based coagulants readily available for use. This could be more useful in remote locations where no water treatment plants are in existence. This research could contribute towards achieving Sustainable Development Goal 6 (SDG 6).

Author Contributions: I.M.T.U.: Research writing and revision. F.-W.L.: Editing, revision and supervision. Y.-C.H.: Supervision and revision. H.-P.K.: Analysis and review Q.-W.C.: Analysis and review. Y.-M.K.: Analysis and review. J.-W.L.: Supervision. and P.-L.S.: Analysis and review. All authors have read and agreed to the published version of the manuscript.

Funding: This research was supported in part by the Kurita Asia Research Grant (22Pmy103) provided by the Kurita Water and Environment Foundation.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Publicly available datasets were analysed in this study. These data can be found here: <https://www.whitehouse.gov/wp-content/uploads/2022/11/Nature-Based-Solutions-Roadmap.pdf>, accessed on 20 January 2023; <http://www.un.org/publications>, accessed on 20 January 2023; <https://data.europa.eu/doi/10.2777/236007>, accessed on 30 January 2023.

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

References

1. CEQ; OSTP; CPO. Opportunities for Accelerating Nature-Based Solutions: A Roadmap for Climate Progress, Thriving Nature, Equity, and Prosperity; Washington, D.C. 2022. Available online: <https://www.whitehouse.gov/wp-content/uploads/2022/11/Nature-Based-Solutions-Roadmap.pdf> (accessed on 20 January 2023).
2. Carvalho, P.N.; Finger, D.C.; Masi, F.; Cipolletta, G.; Oral, H.V.; Tóth, A.; Regelsberger, M.; Exposito, A. Nature-based solutions addressing the water-energy-food nexus: Review of theoretical concepts and urban case studies. *J. Clean. Prod.* **2022**, *338*, 130652. [CrossRef]
3. Xie, L.; Bulkeley, H.; Tozer, L. Mainstreaming sustainable innovation: Unlocking the potential of nature-based solutions for climate change and biodiversity. *Environ. Sci. Policy* **2022**, *132*, 119–130. [CrossRef]
4. Castellar, J.A.C.; Torrens, A.; Buttiglieri, G.; Monclús, H.; Arias, C.A.; Carvalho, P.N.; Galvao, A.; Comas, J. Nature-based solutions coupled with advanced technologies: An opportunity for decentralized water reuse in cities. *J. Clean. Prod.* **2022**, *340*, 130660. [CrossRef]
5. Rizzo, A.; Sarti, C.; Nardini, A.; Conte, G.; Masi, F.; Pistocchi, A. Nature-based solutions for nutrient pollution control in European agricultural regions: A literature review. *Ecol. Eng.* **2023**, *186*, 106772. [CrossRef]
6. Nations, U. The Sustainable Development Goals Report 2022; 22-04175; United Nations, United States of America. 2022. Available online: <http://www.un.org/publications> (accessed on 20 January 2023).
7. Obaideen, K.; Shehata, N.; Sayed, E.T.; Abdelkareem, M.A.; Mahmoud, M.S.; Olabi, A.G. The role of wastewater treatment in achieving sustainable development goals (SDGs) and sustainability guideline. *Energy Nexus* **2022**, *7*, 100112. [CrossRef]
8. Sørup, H.J.D.; Brudler, S.; Godskesen, B.; Dong, Y.; Lerer, S.M.; Rygaard, M.; Arnbjerg-Nielsen, K. Urban water management: Can UN SDG 6 be met within the Planetary Boundaries? *Environ. Sci. Policy* **2020**, *106*, 36–39. [CrossRef]
9. Jarosiewicz, P.; Fazi, S.; Zalewski, M. How to boost Ecohydrological Nature-Based Solutions in water quality management. *Ecohydrol. Hydrobiol.* **2022**, *22*, 226–233. [CrossRef]
10. Serra, T.; Barcelona, A.; Pous, N.; Salvadó, V.; Colomer, J. Disinfection and particle removal by a nature-based *Daphnia* filtration system for wastewater treatment. *J. Water Process Eng.* **2022**, *50*, 103238. [CrossRef]
11. Bratby, J. Coagulation and flocculation in water and wastewater treatment. In *Coagulation and Flocculation in Water and Wastewater Treatment*; IWA Publishing: London, UK, 2016. [CrossRef]
12. Carpinteyro-Urban, S.; Torres, L.G. Use of Response Surface Methodology in the Optimization of Coagulation-Flocculation of Wastewaters Employing Biopolymers. *Int. J. Environ. Res.* **2013**, *7*, 717–726. [CrossRef]
13. He, D.-Q.; Wang, L.-F.; Jiang, H.; Yu, H.-Q. A Fenton-like process for the enhanced activated sludge dewatering. *J. Chem. Eng.* **2015**, *272*, 128–134. [CrossRef]
14. Choy, S.Y.; Prasad, K.M.; Wu, T.Y.; Raghunandan, M.E.; Ramanan, R.N. Utilization of plant-based natural coagulants as future alternatives towards sustainable water clarification. *J. Environ. Sci.* **2014**, *26*, 2178–2189. [CrossRef] [PubMed]
15. Oladoja, N.A. Headway on natural polymeric coagulants in water and wastewater treatment operations. *J. Water Process Eng.* **2015**, *6*, 174–192. [CrossRef]

16. Asrafuzzaman, M.; Fakhruddin, A.N.; Hossain, M.A. Reduction of turbidity of water using locally available natural coagulants. *ISRN Microbiol.* **2011**, *2011*, 632189. [[CrossRef](#)]
17. Vijayaraghavan, G.; Sivakumar, T.; Kumar, A.V. Application of plant-based coagulants for wastewater treatment. *Int. J. Adv. Eng. Res. Stud.* **2011**, *1*, 88–92.
18. Nayak, A.K.; Bera, H.; Hasnain, M.S.; Pal, D. Synthesis and Characterization of Graft Copolymers of Plant Polysaccharides. In *Biopolymer Grafting: Synthesis and Properties*; Elsevier: Amsterdam, The Netherlands, 2018; pp. 1–62. [[CrossRef](#)]
19. Setia, A. Applications of graft copolymerization. In *Biopolymer Grafting: Applications*; Elsevier: Amsterdam, The Netherlands, 2018; pp. 1–44. [[CrossRef](#)]
20. Kurniawan, S.B.; Abdullah, S.R.S.; Imron, M.F.; Said, N.S.M.; Ismail, N.; Hasan, H.A.; Othman, A.R.; Purwanti, I.F. Challenges and Opportunities of Biocoagulant/Biofloculant Application for Drinking Water and Wastewater Treatment and Its Potential for Sludge Recovery. *Int. J. Environ. Res. Public Health* **2020**, *17*, 9312. [[CrossRef](#)]
21. Othmani, B.; Rasteiro, M.G.; Khadhraoui, M. Toward green technology: A review on some efficient model plant-based coagulants/flocculants for freshwater and wastewater remediation. *Clean Technol. Environ. Policy* **2020**, *22*, 1025–1040. [[CrossRef](#)]
22. Yin, C.-Y. Emerging usage of plant-based coagulants for water and wastewater treatment. *Process Biochem.* **2010**, *45*, 1437–1444. [[CrossRef](#)]
23. Kumar, V.; Othman, N.; Asharuddin, S. Applications of Natural Coagulants to Treat Wastewater—A Review. *MATEC Web Conf.* **2017**, *103*, 06016. [[CrossRef](#)]
24. Chua, S.-C.; Show, P.L.; Chong, F.K.; Ho, Y.-C. Lentil waste as a novel natural coagulant for agricultural wastewater treatment. *Water Sci. Technol.* **2020**, *82*, 1833–1847. [[CrossRef](#)]
25. Chua, S.C.; Malek, F.; Mustafa, M.A.; Ismail, M.R.; Sujarwo, N.W.; Wawan, Lim, J.; Ho, Y. Optimized Use of Ferric Chloride and Sesbania Seed Gum (SSG) as Sustainable Coagulant Aid for Turbidity Reduction in Drinking Water Treatment. *Sustainability* **2020**, *12*, 2273. [[CrossRef](#)]
26. El Bouaidi, W.; Libralato, G.; Douma, M.; Ounas, A.; Yaacoubi, A.; Lofrano, G.; Albarano, L.; Guida, M.; Loudiki, M. A review of plant-based coagulants for turbidity and cyanobacteria blooms removal. *Environ. Sci. Pollut. Res. Int.* **2022**, *29*, 42601–42615. [[CrossRef](#)] [[PubMed](#)]
27. Ho, Y.C.; Norli, I.; Alkarkhi, A.F.; Morad, N. Extraction, characterization and application of malva nut gum in water treatment. *J. Water Health* **2015**, *13*, 489–499. [[CrossRef](#)] [[PubMed](#)]
28. Nayak, A.K.; Pal, D. Plant-derived polymers: Ionically gelled sustained drug release systems. In *Encyclopedia of Biomedical Polymers and Polymeric Biomaterials*, 1st ed.; Taylor & Francis Group: Abingdon, UK, 2015; Volume 11, p. 16. [[CrossRef](#)]
29. Saxena, K.; Brighu, U. Comparison of floc properties of coagulation systems: Effect of particle concentration, scale and mode of flocculation. *J. Environ. Chem. Eng.* **2020**, *8*, 104311. [[CrossRef](#)]
30. Wang, D.; Wu, R.; Jiang, Y.; Chow, C.W.K. Characterization of floc structure and strength: Role of changing shear rates under various coagulation mechanisms. *Colloids Surf. A Physicochem. Eng. Asp.* **2011**, *379*, 36–42. [[CrossRef](#)]
31. Torres, L.G.; Carpinteyro-Urban, S.; Corzo-Rios, L.J. Use of Annona Diversifolia and A. Muricata Seeds as Source of Natural Coagulant-Flocculant Aids for the Treatment of Wastewaters. *Eur. J. Biotechnol. Biosci.* **2013**, *1*, 16–22.
32. ASTM. *Coagulation-Flocculation Jar Test of Water (ASTM D2035-19)*; ASTM International: West Conshohocken, PA, USA, 2019. [[CrossRef](#)]
33. APHA; AWWA; WEF. *Standard Methods for the Examination of Water and Wastewater*; American Public Health Association (APHA): Washington, DC, USA, 1999.
34. Breig, S.J.M.; Luti, K.J.K. Response surface methodology: A review on its applications and challenges in microbial cultures. *Mater. Today Proc.* **2021**, *42*, 2277–2284. [[CrossRef](#)]
35. Choi, H.J.; Naznin, M.; Alam, M.B.; Javed, A.; Alshammari, F.H.; Kim, S.; Lee, S.H. Optimization of the extraction conditions of Nypa fruticans Wurmb. using response surface methodology and artificial neural network. *Food Chem.* **2022**, *381*, 132086. [[CrossRef](#)]
36. Desta, W.M.; Bote, M.E. Wastewater treatment using a natural coagulant (Moringa oleifera seeds): Optimization through response surface methodology. *Heliyon* **2021**, *7*, e08451. [[CrossRef](#)]
37. Montgomery, D.C. Response Surface Methods and Designs. In *Design and Analysis of Experiments*; John Wiley & Sons, Inc.: Hoboken, NJ, USA, 2017; pp. 489–558.
38. Koul, B.; Bhat, N.; Abubakar, M.; Mishra, M.; Arukha, A.P.; Yadav, D. Application of Natural Coagulants in Water Treatment: A Sustainable Alternative to Chemicals. *Water* **2022**, *14*, 3751. [[CrossRef](#)]
39. Tchobanoglous, G.; Burton, F.L.; Stensel, H.D. Adsorption. In *Wastewater Engineering: Treatment and Reuse*; Metcalf & Eddy Inc., Ed.; McGraw-Hill: New York, NY, USA, 2003.
40. Ho, Y.C.; Norli, I.; Alkarkhi, A.F.M.; Morad, N. Characterization of biopolymeric flocculant (pectin) and organic synthetic flocculant (PAM): A comparative study on treatment and optimization in kaolin suspension. *Bioresour. Technol.* **2010**, *101*, 1166–1174. [[CrossRef](#)]
41. Gupta, S.; Sharma, P.; Soni, P.L. Carboxymethylation of Cassia occidentalis seed gum. *J. Appl. Polym. Sci.* **2004**, *94*, 1606–1611. [[CrossRef](#)]
42. Seddon, N.; Chausson, A.; Berry, P.; Girardin, C.A.J.; Smith, A.; Turner, B. Understanding the value and limits of nature-based solutions to climate change and other global challenges. *Philos. Trans. R. Soc. B* **2020**, *375*, 20190120. [[CrossRef](#)] [[PubMed](#)]

43. Bourguignon, D. Nature-Based Solutions Concept, Opportunities, and Challenges; EPRS | European Parliamentary Research Service, European Parliament. 2017. Available online: <http://www.europarl.europa.eu/thinktank/> (accessed on 23 January 2023).
44. Czembrowski, P.; Kronenberg, J.; Czepkiewicz, M. Integrating non-monetary and monetary valuation methods—SoftGIS and hedonic pricing. *Ecol. Econ.* **2016**, *130*, 166–175. [[CrossRef](#)]
45. Mukherjee, N.; Sutherland, W.J.; Dicks, L.; Hoge, J.; Koedam, N.; Dahdouh-Guebas, F. Ecosystem service valuations of mangrove ecosystems to inform decision making and future valuation exercises. *PLoS ONE* **2014**, *9*, e107706. [[CrossRef](#)]
46. Nesshover, C.; Assmuth, T.; Irvine, K.N.; Rusch, G.M.; Waylen, K.A.; Delbaere, B.; Haase, D.; Jones-Walters, L.; Keune, H.; Kovacs, E.; et al. The science, policy, and practice of nature-based solutions: An interdisciplinary perspective. *Sci. Total Environ.* **2017**, *579*, 1215–1227. [[CrossRef](#)]
47. Faivre, N.; Fritz, M.; Freitas, T.; de Boissezon, B.; Vandewoestijne, S. Nature-Based Solutions in the EU: Innovating with nature to address social, economic and environmental challenges. *Environ. Res.* **2017**, *159*, 509–518. [[CrossRef](#)]
48. Narayan, S.; Beck, M.W.; Reguero, B.G.; Losada, I.J.; van Wesenbeeck, B.; Pontee, N.; Sanchirico, J.N.; Ingram, J.C.; Lange, G.M.; Burks-Copes, K.A. The Effectiveness, Costs and Coastal Protection Benefits of Natural and Nature-Based Defences. *PLoS ONE* **2016**, *11*, e0154735. [[CrossRef](#)]
49. Liu, H.-Y.; Jay, M.; Chen, X. The Role of Nature-Based Solutions for Improving Environmental Quality, Health, and Well-Being. *Sustainability* **2021**, *13*, 10950. [[CrossRef](#)]
50. Ferreira, C.S.S.; Potočki, K.; Kapović-Solomon, M.; Kalantari, Z. Nature-Based Solutions for Flood Mitigation and Resilience in Urban Areas. In *Nature-Based Solutions for Flood Mitigation: Environmental and Socio-Economic Aspects*; Ferreira, C.S.S., Kalantari, Z., Hartmann, T., Pereira, P., Eds.; Springer International Publishing: New York, NY, USA, 2022; pp. 59–78. [[CrossRef](#)]
51. Kumar, P.; Debele, S.E.; Sahani, J.; Rawat, N.; Marti-Cardona, B.; Alfieri, S.M.; Basu, B.; Basu, A.S.; Bowyer, P.; Charizopoulos, N.; et al. Nature-based solutions efficiency evaluation against natural hazards: Modelling methods, advantages, and limitations. *Sci. Total Environ.* **2021**, *784*, 147058. [[CrossRef](#)]
52. Mensah-Akutteh, H.; Buamah, R.; Wiafe, S.; Nyarko, K.B. Optimising coagulation-flocculation processes with aluminium coagulation using response surface methods. *Appl. Water Sci.* **2022**, *12*, 188. [[CrossRef](#)]
53. Brink, E.; Aalders, T.; Adam, D.; Feller, R.; Henselek, Y.; Hoffmann, A.; Ibe, K.; Matthey-Doret, A.; Meyer, M.; Negrut, N.L.; et al. Cascades of green: A review of ecosystem-based adaptation in urban areas. *Glob. Environ. Chang.* **2016**, *36*, 111–123. [[CrossRef](#)]
54. McVittie, A.; Cole, L.; Wreford, A.; Sgobbi, A.; Yordi, B. Ecosystem-based solutions for disaster risk reduction: Lessons from European applications of ecosystem-based adaptation measures. *Int. J. Disaster Risk Reduct.* **2018**, *32*, 42–54. [[CrossRef](#)]
55. Bulkeley, H.; Naumann, S.; Vojinovic, Z.; Calfapietra, C.; Whiteoak, K.; Freitas, T.; Vandewoestijne, S.; Wild, T. European Commission, Directorate-General for Research and Innovation. In *Nature-Based Solutions: State of the Art in EU-Funded Projects*; Freitas, T., Vandewoestijne, S., Wild, T., Eds.; Publications Office of the European Union: Luxembourg, 2020; Available online: <https://data.europa.eu/doi/10.2777/236007> (accessed on 30 January 2023).

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.