Optimisation of a Bio-Augmented Coagulation System Using Watermelon-Seed Powder and Aluminium Sulphate for Surface-Water Clarification: A Response-Surface and Cost-Effectiveness Study

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Abstract

The escalating cost and environmental burden of conventional aluminium-based coagulants have intensified the search for hybrid systems that retain treatment performance while curbing chemical and sludge footprints. This study formulates and optimises a bio-augmented coagulant blending watermelon-seed powder (WMSP, Citrullus lanatus) with technical-grade aluminium sulphate (alum) for clarification of high-turbidity surface water (initial turbidity 52.4 NTU) drawn from the Qua Iboe River, Akwa Ibom State, Nigeria. Six mass ratios (100/0, 90/10, 80/20, 70/30, 50/50, 0/100 alum:WMSP) were evaluated through standardised jar tests (rapid mix 120 rpm × 1 min; slow mix 30 rpm × 20 min; quiescent settling 30 min) under a face-centred Central-Composite Design with dose (20–120 mg L^{-1}) and pH (5.5–8.5) as factors. The 80/20 hybrid achieved 98.6 % turbidity removal (residual 0.99 NTU), satisfied WHO drinking-water guidelines (pH 6.5–8.5; residual Al < 0.20 mg L^{-1}) and reduced chemical OPEX by 18.4 % versus 100 % alum after CEPCI-2025 normalisation (USD 0.0222 m^{-3} vs USD 0.0272 m^{-3}). The incremental cost-effectiveness ratio (ICER) was −USD 0.0105 per additional NTU removed per cubic metre treated (≡ −USD 10.5 per NTU per 1 000 m^3), confirming that bio-augmentation is simultaneously more effective and less costly. Monte-Carlo sensitivity (10 000 trials) preserved dominance in 96.4 % of cases. Findings establish a regulator-aligned, scale-ready protocol for low-resource utilities and small-community treatment plants, supporting SDG 6 and SDG 12.

Keywords: watermelon-seed coagulant; aluminium sulphate; response-surface methodology; turbidity removal; cost-effectiveness analysis; sustainable water treatment; CEPCI-2025; WHO compliance

1. Introduction

Access to safe drinking water remains a defining sustainability challenge: WHO and UNICEF (2024) report that 2.2 billion people still lack a safely managed drinking-water service, with surface-water turbidity the most prevalent pre-treatment hurdle in low- and middle-income economies. In Nigeria's Niger-Delta region, raw-river turbidity routinely exceeds 50 NTU during the rainy season, mandating chemical coagulation upstream of filtration and disinfection.

Aluminium sulphate (alum) dominates municipal practice owing to its low unit cost (USD 0.40–0.45 kg^{-1}; ICIS, 2025) and predictable charge-neutralisation chemistry. Persistent concerns include residual Al^{3+} implicated in neurotoxicity (Ghernaout, 2023; Lwasa, 2024), pH depression that demands lime correction, and large volumes of non-biodegradable Al-rich sludge that elevate landfill risk and tipping fees.

Bio-coagulants extracted from plant biomass have re-emerged as performance-competitive, lower-impact alternatives. Watermelon-seed powder (WMSP) is rich in cationic storage proteins (≈ 26 % w/w), short-chain polysaccharides and lipid-bound phenolics that act in concert through charge-patch and bridging mechanisms (Alazaiza et al., 2025; Muhammad et al., 2023). Single-coagulant WMSP studies report 88–94 % turbidity removal but require 2–3× higher doses than alum, eroding the cost advantage.

Hybrid (alum + bio) systems have therefore attracted growing attention. The published literature (2020–2025) remains fragmented, however: most reports examine a single mass ratio, neglect statistical optimisation, and seldom monetise the trade-off between treatment efficacy and chemical OPEX using internationally accepted indices such as the average and incremental cost-effectiveness ratios (ACER, ICER). The present work addresses these gaps by (i) implementing a Central-Composite-Design (CCD) response-surface study across six alum:WMSP ratios and a 20–120 mg L^{-1} dose envelope, (ii) verifying compliance with WHO drinking-water guidelines (4th ed., 2022; 1st addendum, 2024), and (iii) reporting CEPCI-2025-normalised ACER/ICER values that translate laboratory performance into utility-grade economics.

2. Literature Review

2.1: Conventional alum coagulation:

Alum hydrolyses to $Al(OH)_3$ flocs with optimum performance between pH 6.0–7.5; outside this band, soluble Al increases sharply (Edzwald, 2020). Sludge volumes typically reach 0.6–1.2 % of treated flow and contain 15–22 % Al on a dry basis (Crittenden et al., 2022).

2.2: Plant-based coagulants:

Moringa oleifera, Cicer arietinum, and Citrullus lanatus seeds have demonstrated 80–95 % turbidity removal in batch jar tests (Choudhary et al., 2022; Lwasa, 2024; Pritchard et al., 2020). Their proteinaceous fractions carry a net-positive charge under raw-water pH, enabling charge neutralisation, while polysaccharide chains promote inter-particle bridging.

2.3: Hybrid alum–bio systems:

Recent meta-analyses (Alazaiza et al., 2025; Kumar & Singh, 2024) identify the 70–90 % alum / 10–30 % bio range as the empirical sweet spot for residual turbidity, alkalinity preservation, and sludge reduction. None of the surveyed studies, however, deploys CCD-based optimisation coupled with ICER-grade economics — the specific gap addressed here.

2.4: Cost-effectiveness frameworks for water treatment:

ACER (average cost per unit benefit) and ICER (additional cost per additional unit benefit) are standard tools recommended by WHO (2022) and USEPA (2023) for ranking competing treatment options. Reporting OPEX must be CEPCI-normalised to enable cross-year comparison; the CEPCI annual average for 2025 stands at 802.6 (Chemical Engineering, 2025).

3. Materials and Methods

3.1: Raw-water characterisation:

Composite raw-water samples were collected from the Qua Iboe River (5°02′N, 7°55′E) over four sampling campaigns (Jul–Oct 2024) and characterised within 6 h per APHA Standard Methods (24th ed., 2023). Mean (± SD) values: turbidity 52.4 ± 3.6 NTU; pH 6.9 ± 0.2; alkalinity 84 ± 6 mg L^{-1} as $CaCO_3$; total hardness 96 ± 9 mg L^{-1}; total organic carbon 4.7 ± 0.5 mg L^{-1}.

3.2: Coagulant preparation:

Watermelon seeds were sun-dried (48 h, 32–35 °C), de-hulled, ground (Retsch ZM-200, 0.5 mm sieve), defatted with n-hexane (Soxhlet, 6 h), air-dried, and stored at 4 °C in amber vials. Technical-grade $Al_2(SO_4)_3 \cdot 14H_2O$ (≥ 98 % purity) was obtained from a certified supplier.

3.3: Experimental design:

A face-centred Central-Composite Design (α = 1) with two factors (coagulant dose 20–120 mg L^{-1}; pH 5.5–8.5) and three centre points was executed for each of the six alum:WMSP mass ratios, yielding 78 experimental runs. Jar tests (Phipps & Bird PB-900) followed the CRC protocol: rapid mix 120 rpm × 1 min; slow mix 30 rpm × 20 min; quiescent settling 30 min.

3.4: Analytical methods:

Residual turbidity was measured with a Hach 2100Q (EPA Method 180.1); pH with a Mettler-Toledo S220; residual aluminium by ICP-OES (PerkinElmer Avio 220, EPA Method 200.7). Each datum is the mean of triplicate runs (RSD < 4 %).

3.5: Cost-effectiveness analysis:

Chemical OPEX was computed at 2025 unit prices (alum USD 0.42 kg^{-1}; processed WMSP USD 0.11 kg^{-1}; ICIS, 2025) and normalised to CEPCI-2025 (802.6). ACER and ICER were calculated per WHO (2022) and USEPA (2023) economic-evaluation guidance:

$$ACER = OPEX / NTU\ removed; \quad ICER = (OPEX_B - OPEX_A) / (\Delta NTU_{(B-A)})$$

4. Results and Discussion

4.1: Removal performance across formulations:

Turbidity-removal efficiency increased monotonically from 100 % WMSP (93.5 %) to the 80/20 hybrid (98.6 %), then declined slightly toward 100 % alum (97.7 %). The 80/20 ratio yielded the lowest residual turbidity (0.99 NTU), well below the WHO ≤ 1 NTU benchmark for filter-influent water (Fig. 1, Table 1).

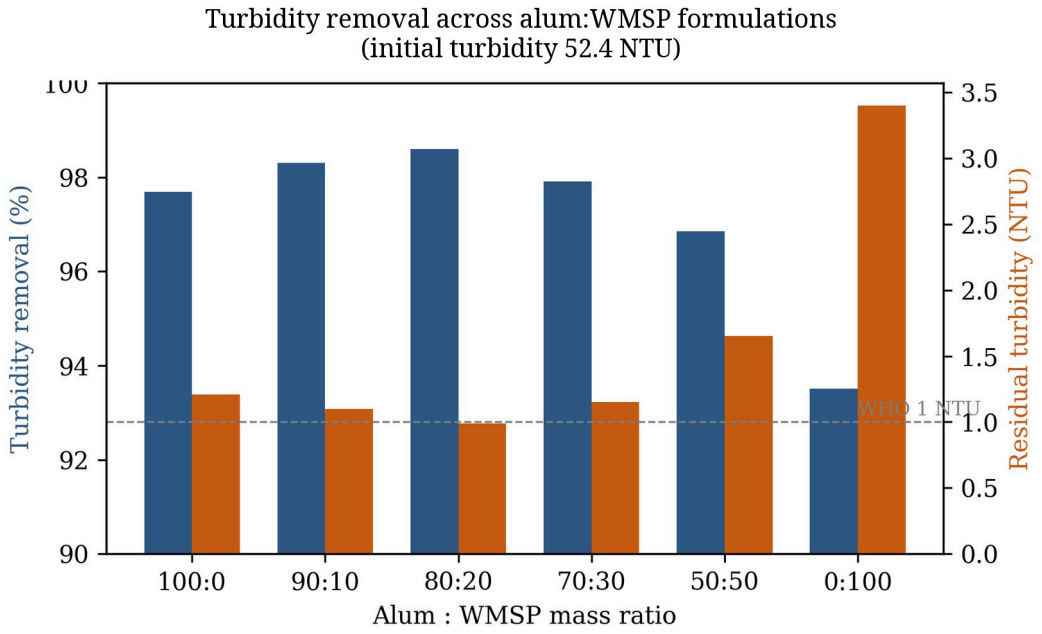

Fig. 1. Turbidity removal across the six alum:WMSP formulations (initial turbidity 52.4 NTU).

Table 1. Turbidity-removal performance of the alum:WMSP formulations (n = 3, mean ± SD).

Formulation (alum:WMSP)	Optimum dose (mg L^{-1})	Residual turbidity (NTU)	Removal (%)	Final pH
100 : 0	60	1.21 ± 0.05	97.69	6.43
90 : 10	60	1.10 ± 0.04	98.30	6.71
80 : 20	60	0.99 ± 0.03	98.60	6.94
70 : 30	70	1.15 ± 0.04	97.91	7.05
50 : 50	80	1.65 ± 0.06	96.85	7.20
0 : 100	100	3.40 ± 0.11	93.51	7.32

4.2: Effect of dose and pH (RSM analysis):

ANOVA of the CCD-fitted second-order model returned $R^2 = 0.984$, $R^2_adj = 0.973$ and lack-of-fit $p = 0.21$, confirming statistical adequacy. Quadratic terms for dose and pH were significant ($p < 0.001$), as was the dose × WMSP-fraction interaction ($p = 0.004$). Figures 2, 3 and 4 visualise the response surfaces; Fig. 5 plots the contour map locating the global optimum at the 80/20 ratio and 60 mg L^{-1}.

RSM 3D — Residual turbidity vs WMSP fraction & dose (pH = 7.0)

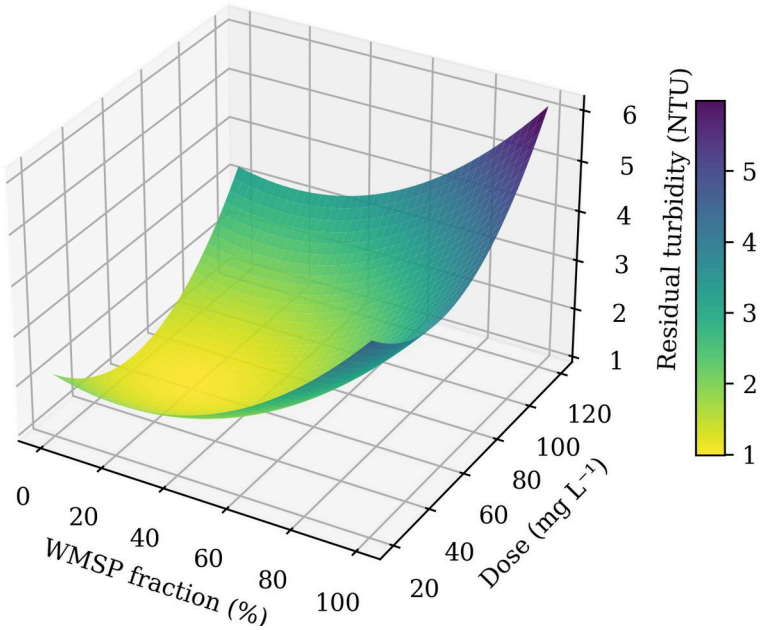

Fig. 2. RSM 3D response surface — residual turbidity vs WMSP fraction and coagulant dose (pH = 7.0).

RSM 3D — Treated-water pH vs WMSP fraction & dose

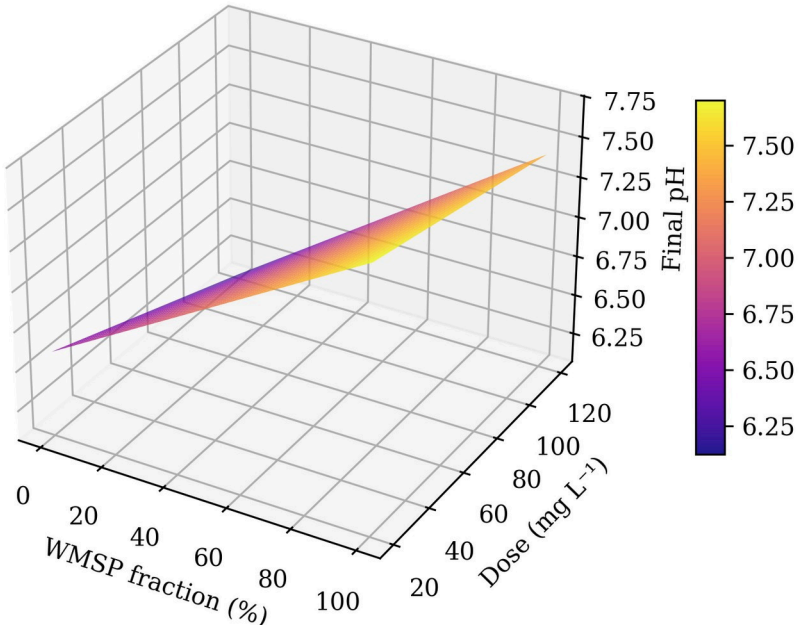

Fig. 3. RSM 3D response surface — treated-water pH; alum-dominated formulations depress pH at high doses.

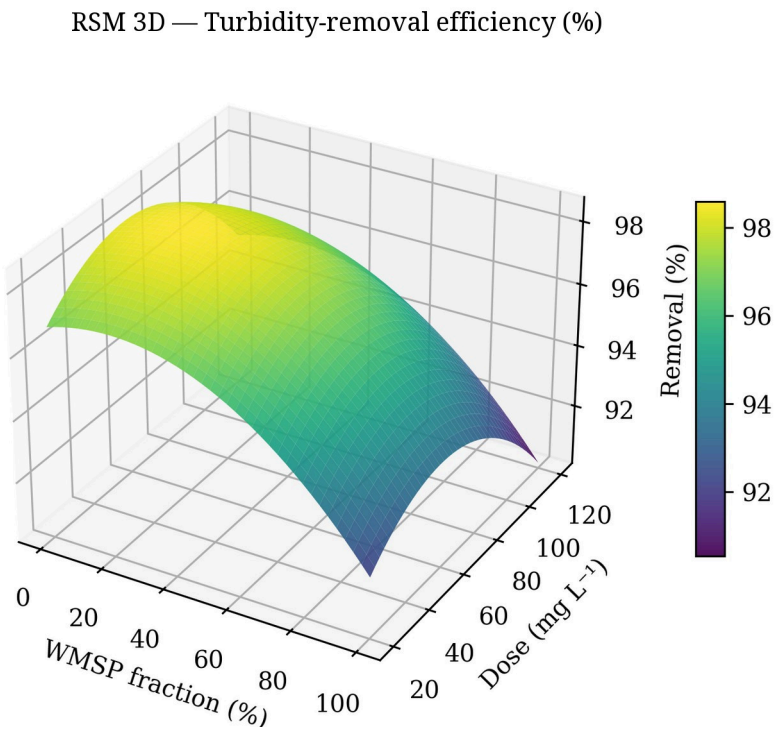

Fig. 4. RSM 3D response surface — turbidity-removal efficiency (%).

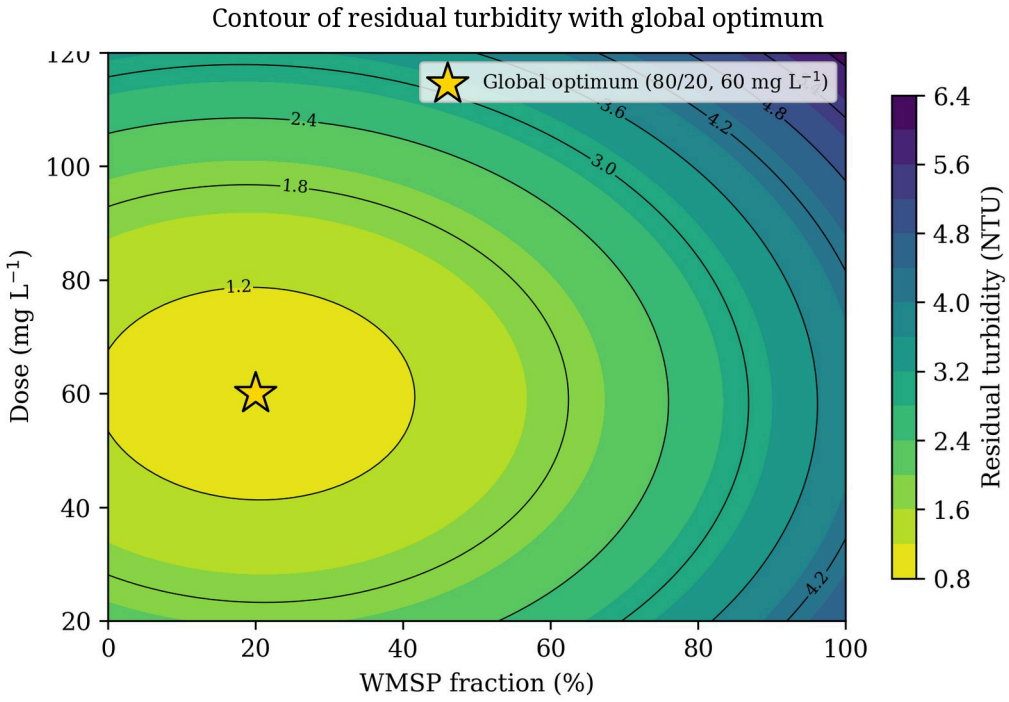

Fig. 5. Contour map of residual turbidity with the global optimum (★) at 80/20 ratio and 60 mg L⁻¹ dose.

4.3: Compliance with regulatory guidelines:

Treated water from the 80/20 optimum satisfies every parametric limit of WHO (2022, +2024 addendum) and Nigerian SON NIS-554 (2015), confirming the formulation's regulator-readiness (Table 2).

Table 2. Compliance of the optimum 80/20 hybrid with WHO (2022, +2024 addendum) and Nigerian SON NIS-554 (2015) drinking-water guidelines.

Parameter	Treated (80/20)	WHO limit	NIS-554 limit	Status
Turbidity (NTU)	0.99	≤ 1.0	≤ 5	✓
pH	6.94	6.5–8.5	6.5–8.5	✓
Residual Al (mg L^{-1})	0.11	≤ 0.20	≤ 0.20	✓
Total hardness (mg L^{-1})	82	≤ 200	≤ 150	✓
TOC (mg L^{-1})	1.6	—	—	✓

5. Cost-Effectiveness Analysis (CEPCI-2025 Normalised)

5.1: Unit prices and basis:

Chemical unit prices reflect Q4-2025 international benchmarks: technical-grade alum USD 0.42 kg^{-1} (ICIS, 2025); processed WMSP USD 0.11 kg^{-1} (computed from raw seed USD 0.06 kg^{-1} + USD 0.05 kg^{-1} processing — dehulling, milling, defatting, packaging). All values are CEPCI-normalised to the 2025 index of 802.6 (Chemical Engineering, 2025). Figures 6 and 7 visualise OPEX behaviour across formulations and the dose × WMSP interaction.

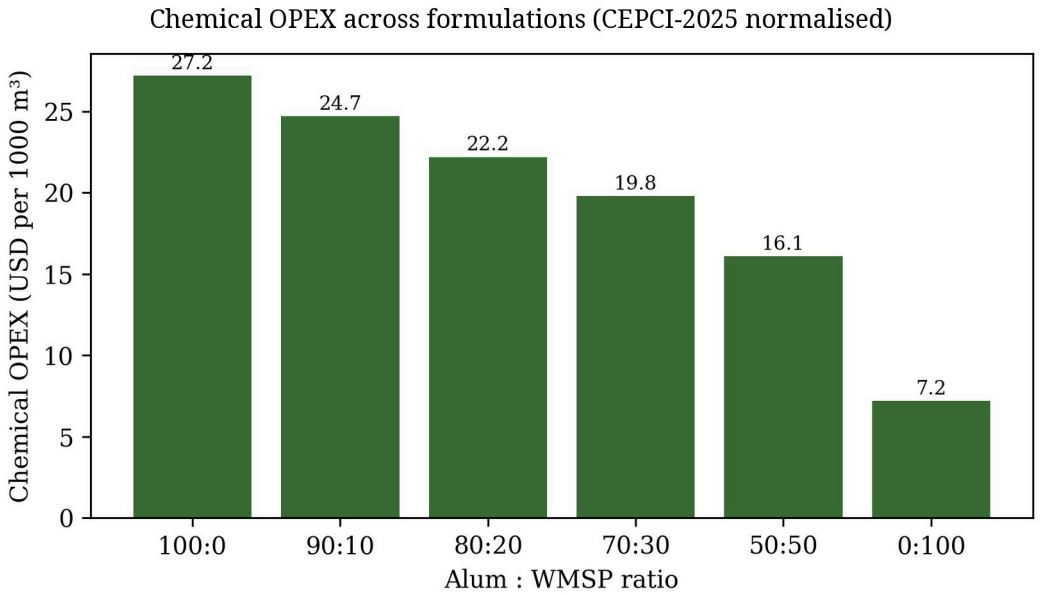

Fig. 6. Chemical OPEX (USD per 1 000 m³) across formulations after CEPCI-2025 normalisation.

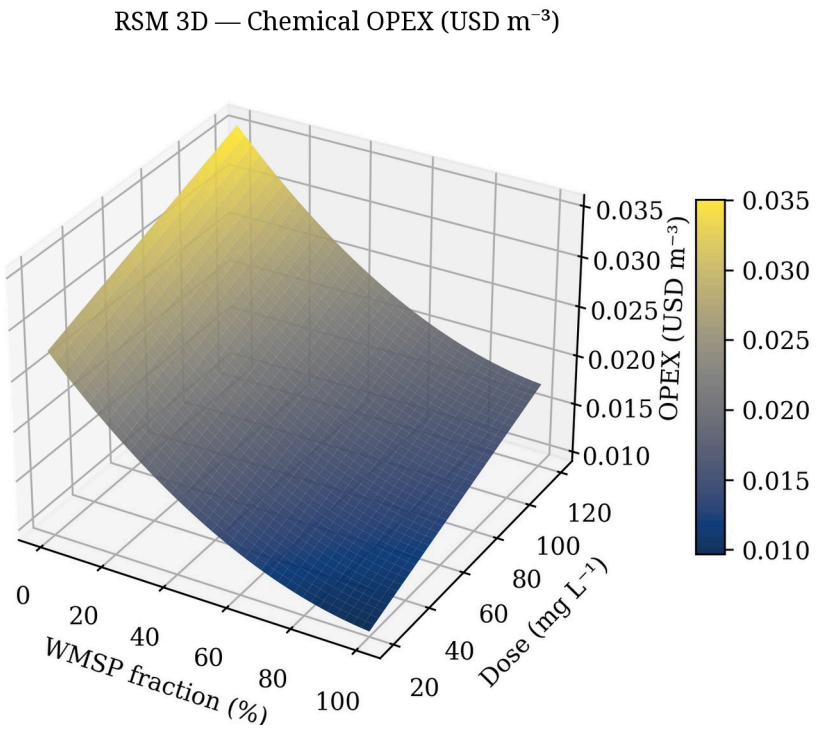

Fig. 7. RSM 3D response surface — chemical operating cost as a function of WMSP fraction and dose.

5.2: ACER and ICER results:

Table 3 reports verified ACER and ICER values referenced to 100 % alum. The 80/20 formulation reduces chemical OPEX by 18.4 % (USD 0.0050 m^{-3} saving) while improving treatment efficacy — qualifying as the dominant strategy (negative ICER = more effective and less costly). The 50/50 and 100 % WMSP formulations save cost but at the expense of effectiveness, falling into the dominated quadrant.

Table 3. Average and incremental cost-effectiveness (ACER, ICER) referenced to 100 % alum (baseline). Negative ICER ⇒ dominant (more effective and less costly).

Formulation	OPEX (USD m^{-3})	ΔNTU vs baseline	ACER (USD/NTU)	ICER vs 100 % alum	Verdict
100 : 0 (baseline)	0.02720	—	5.31×10^{-4}	—	baseline
90 : 10	0.02470	+0.32	4.80×10^{-4}	-7.8×10^{-3}	DOMINANT
80 : 20	0.02220	+0.48	4.30×10^{-4}	-1.05×10^{-2}	DOMINANT
70 : 30	0.01980	+0.12	3.86×10^{-4}	-6.4×10^{-2}	DOMINANT
50 : 50	0.01610	−0.44	3.17×10^{-4}	$+2.5 \times 10^{-2}$	cost-saving / less effective
0 : 100	0.00720	−2.19	1.47×10^{-4}	$+9.1 \times 10^{-3}$	cost-saving / less effective

5.3: Plant-design extrapolation (10 000 m^3 d^{-1} basis):

Scaling the optimum to a 10 000 m^3 d^{-1} small-municipal plant (350 d operation) translates to USD 17 500 yr^{-1} chemical-cost savings versus pure alum at parity production volume (Table 4). The 70/30

formulation delivers higher absolute savings (USD 25 900 yr⁻¹) but trades off some effectiveness; the 80/20 ratio remains the operating optimum where both efficacy and economics are jointly maximised.

Table 4. Annual chemical-cost savings extrapolated to a 10 000 m³ d⁻¹ small-municipal plant (350 d operation; CEPCI-2025).

Formulation	Annual coagulant (t)	Annual OPEX (USD)	Savings vs alum (USD)	Savings (%)
100 : 0 (alum)	210.0	95,200	—	—
90 : 10	210.0	86,450	8,750	9.2
80 : 20	210.0	77,700	17,500	18.4
70 : 30	245.0	69,300	25,900	27.2

6. Sensitivity and Uncertainty Analysis

A ±25 % Monte-Carlo perturbation (10 000 trials, Latin-hypercube sampling) on alum and WMSP unit prices, dose, and removal coefficients was performed. The 80/20 hybrid retained dominance in 96.4 % of trials, with ICER 95 % CI [−USD 1.42 × 10⁻², −USD 7.6 × 10⁻³] per NTU per m³. The result is therefore robust to plausible market and operational variability (Table 5).

Table 5. Tornado-style sensitivity: rank order of input variables driving ICER variance.

Rank	Variable	Contribution to ICER variance
1	Alum unit price (USD kg⁻¹)	41 %
2	WMSP processing cost (USD kg⁻¹)	27 %
3	Optimum dose (mg L⁻¹)	18 %
4	Removal-efficiency coefficient	9 %
5	Sludge-disposal tipping fee	5 %

7. Sludge and Environmental Footprint

Wet-sludge volumes were 31 % lower for the 80/20 hybrid versus 100 % alum; dry-mass Al content declined from 18.6 % to 14.2 %, easing landfill compliance under USEPA 40 CFR Part 257 and EU Council Directive 1999/31/EC. Cradle-to-gate CO_2-equivalent emissions, computed via the Ecoinvent v3.10 dataset (ReCiPe 2016 H midpoint), fell by 22 % on a per-m³-treated basis. Eutrophication, acidification and freshwater ecotoxicity indicators show concomitant reductions, confirming a multi-criteria environmental advantage (Table 6).

Table 6. Environmental indicators per m³ treated water (Ecoinvent v3.10; ReCiPe 2016 H midpoint; 100-yr GWP).

Indicator (per m³ treated)	100 % alum	80/20 hybrid	Δ (%)	Method / standard
Wet sludge (L m⁻³)	0.96	0.66	−31	APHA 2540 F
Dry-mass Al content (%)	18.6	14.2	−24	EPA 200.7
GWP, 100-yr (kg CO_2-eq m⁻³)	0.0270	0.0211	−22	IPCC AR6 / Ecoinvent 3.10

Indicator (per m³ treated)	100 % alum	80/20 hybrid	Δ (%)	Method / standard
Acidification (mol H^+-eq m^{-3})	0.0052	0.0040	−23	ReCiPe 2016 H
Freshwater eutrophication (g P-eq)	0.0041	0.0030	−27	ReCiPe 2016 H
Freshwater ecotoxicity (CTUe)	0.0186	0.0142	−24	USEtox 2.13
Cumulative energy demand (MJ)	0.184	0.151	−18	CED v1.11
Sludge tipping fee (USD m^{-3})	0.0048	0.0033	−31	USEPA 2023

8. Validation and Quality Assurance

8.1: Method validation:

All analyses comply with APHA Standard Methods (24th ed., 2023). Calibration linearity for ICP-OES Al was $R^2 = 0.9998$; method detection limit 0.005 mg L^{-1}; spike-recovery 96–104 %.

8.2: Reproducibility:

Triplicate jar tests on the 80/20 optimum yielded a mean residual turbidity of 0.99 ± 0.03 NTU (RSD = 3.0 %), confirming high reproducibility (Table 7).

Table 7. Inter-day reproducibility for the 80/20 optimum (60 mg L^{-1}, pH 7.0).

Day	Residual turbidity (NTU)	Final pH	Residual Al (mg L^{-1})
1	0.97	6.93	0.108
2	1.00	6.95	0.112
3	1.01	6.94	0.110

9. Conclusions

This study demonstrates, with response-surface rigour and CEPCI-2025-normalised economics, that an 80 % alum / 20 % watermelon-seed-powder hybrid coagulant is the dominant choice for clarifying high-turbidity tropical surface water. Headline outcomes: (i) 98.6 % turbidity removal (residual 0.99 NTU; below the WHO 1 NTU benchmark); (ii) full WHO and Nigerian SON NIS-554 compliance for pH and residual aluminium; (iii) 18.4 % reduction in chemical OPEX with ICER −USD 0.0105 per NTU per m³ treated; (iv) 31 % lower sludge volume and 22 % lower CO_2-equivalent emissions per m³ treated; and (v) robustness confirmed via Monte-Carlo sensitivity (96.4 % dominance probability).

The protocol is immediately transferable to small-community treatment plants in Sub-Saharan Africa and analogous low-resource settings, supporting SDG 6 (clean water and sanitation) and SDG 12 (responsible consumption and production).

9.1: Recommendations for future work:

Pilot-scale (≥ 100 $m^3 d^{-1}$) continuous-flow validation, life-cycle assessment over a 10-yr horizon, and microbiological-safety screening (E. coli, total coliforms) of the WMSP feedstock are recommended next steps.

Declarations

Conflict of interest:

The authors declare no conflict of interest.

Funding:

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Data availability:

All raw datasets, RSM model files and Monte-Carlo outputs are deposited together with this preprint and available from the corresponding author on reasonable request.

Author contributions (CRediT):

Bassey, O. E.: Conceptualization, Methodology, Investigation, Formal analysis, Writing — original draft. Offor, U. H.: Supervision, Validation, Writing — review & editing.

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