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Research Article

Fluoride Removal Efficiency of Calcium-spiked and Non-spiked *Moringa Oleifera* Seed Powder

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Abstract

Fluoride contamination in drinking water remains a widespread public health concern, particularly in arid and semi-arid regions where groundwater is the primary source of potable water. Chronic exposure to elevated fluoride levels—commonly above the World Health Organization's (WHO) recommended limit of 1.5 mg/L—can result in dental and skeletal fluorosis, affecting millions of people globally. Affordable and effective defluoridation technologies are urgently needed, especially in low-income rural settings. In this study, the fluoride removal efficiency of calcium-spiked and non-spiked *Moringa oleifera* seed powder was investigated through controlled laboratory batch adsorption experiments. Biosorbents were prepared by treating ground seed powder with 1% calcium chloride solution and characterised based on their performance across five fluoride concentrations (1-20 ppm). Key parameters such as removal efficiency, residual fluoride levels, and adsorption capacity (q_e) were evaluated under consistent operating conditions (pH 7, 2 g/50 mL dose, mesh 40, 120 minutes). Results indicated that calcium-spiked *Moringa oleifera* powder significantly outperformed its non-spiked counterpart. At 1 ppm, the spiked adsorbent achieved 94.35 ± 1.15% removal efficiency, compared to 81.45 ± 1.35% for the non-spiked. At the highest tested concentration (20 ppm), the spiked biosorbent still removed 72.31 ± 1.80% of fluoride, while the non-spiked removed only 54.21 ± 1.95%. Linear regression models showed strong inverse correlations between fluoride concentration and removal efficiency ($R^2 > 0.99$, $p < 0.001$). The spiked adsorbent also resulted in significantly lower residual fluoride concentrations, with final values closer to the WHO guideline. One-way ANOVA confirmed significant differences in adsorption capacity and efficiency between treatments ($p < 0.001$). These findings highlight the effectiveness of calcium modification in enhancing biosorption performance and suggest that calcium-spiked *Moringa oleifera* seed powder is a promising, low-cost, and environmentally friendly solution for mitigating fluoride contamination in drinking water.

Keywords

Fluoride Removal Efficiency, *Moringa Oleifera biosorbent*, Calcium Spiked Moringa Seeds, Adsorption Efficiency of Moringa Oleifera, Defluoridation, Residual Fluoride Ions, Drinking Water Safety

1. Introduction

Fluoride contamination in drinking water remains a persistent environmental and public health challenge, especially in arid and semi-arid regions where groundwater is the primary source of potable water. Although fluoride plays a critical role in preventing dental caries at trace concentrations (0.5-1.0 mg/L), excessive intake beyond the World Health

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Organization (WHO) guideline value of 1.5 mg/L can result in serious health consequences, including dental and skeletal fluorosis [1]. In countries such as Kenya, India, Ethiopia, and Tanzania, high fluoride levels in groundwater, often exceeding 5.0 mg/L, have been widely documented and pose a threat to rural populations dependent on untreated groundwater [2-5].

Conventional fluoride removal technologies, such as reverse osmosis, activated alumina filtration, ion exchange resins, and electrodialysis, have proven effective in controlled settings but are constrained by high operational costs, maintenance complexity, and limited applicability in low-income and decentralized rural settings [6-8]. These limitations have intensified the search for low-cost, environmentally friendly, and sustainable fluoride removal solutions. In recent years, biosorption using plant-based materials has emerged as a promising alternative due to its affordability, ease of use, and reliance on locally available biomass [9]. Among the most studied plant materials, Moringa oleifera has attracted considerable attention due to its rich phytochemical composition, natural coagulant properties, and proven potential in water purification [10, 11].

Moringa oleifera seed powder contains bioactive compounds such as cationic peptides, proteins, and carboxyl and hydroxyl functional groups, which enable it to bind with negatively charged ions like fluoride [10, 12]. However, studies have shown that the fluoride removal efficiency of raw Moringa seed powder is often moderate, with reported removal efficiencies ranging from 40% to 70%, depending on the water chemistry and operational conditions [12, 9]. This limitation has prompted the development of modified Moringa biosorbents, particularly through metal-ion enrichment techniques, to enhance the surface activity and ion-exchange capacity of the biomass [13, 14, 9]. Calcium spiking, in particular, has been identified as a promising approach due to the high affinity between calcium ions (Ca^{2+}) and fluoride ions (F^-), which form the sparingly soluble compound calcium fluoride (CaF_2), thereby enhancing removal through both biosorption and precipitation mechanisms [15].

Calcium-modified biosorbents have been explored using various natural materials such as banana peels, rice husks, and activated carbon; however, relatively limited studies have specifically evaluated calcium-spiked Moringa oleifera for fluoride removal under optimized conditions [14, 15]. Moreover, comparative analyses between calcium-spiked and non-spiked Moringa biomass under varying physicochemical conditions (e.g., fluoride concentration, pH, contact time, particle size, and dosage) remain scarce in East African settings. This represents a significant knowledge gap, particularly in Kenya, where fluoride contamination is widespread in the Rift Valley and Baringo regions, and local, affordable treatment options are urgently needed [4, 5].

The interaction between biosorbent surface characteristics and fluoride binding kinetics is also influenced by factors such as biosorbent dosage, particle size, and solution pH, which affect surface-area availability, electrostatic interactions, and the speciation of fluoride ions [9, 16]. Studies emphasize the need for optimizing these variables in tandem to maximize fluoride removal while ensuring minimal alterations to the final water chemistry, particularly pH stability and absence of residual metal ions [9, 16]. Furthermore, adsorption performance is typically evaluated using batch experiments that allow the assessment of equilibrium isotherms, adsorption kinetics, and thermodynamic parameters. These tools help elucidate the mechanisms underpinning biosorption and provide essential data for upscaling the process for field-level applications [16].

2. Methodology

2.1. Research Design

This study adopted a controlled batch experimental design to assess and compare the fluoride removal efficiency of calcium-spiked and non-spiked *Moringa oleifera* seed powder. The focus was on quantifying the percentage fluoride removal, residual fluoride concentration, and adsorption capacity under varied initial fluoride concentrations. All tests were conducted under standardized conditions to ensure valid comparisons.

2.2. Biosorbent Preparation

Mature *Moringa oleifera* seeds were obtained from Baringo County, Kenya. The seeds were dehulled, washed, air-dried at room temperature for 48 hours, and ground using an electric blender. The resulting powder was sieved using a 40-mesh sieve (425 μm) to ensure uniform particle size across treatments.

Calcium spiking was done by soaking 50 g of seed powder in 500 mL of 0.1 M $CaCl_2$ solution for 24 hours at 25 ± 2 ℃ with continuous stirring at 150 rpm. The residue was filtered, rinsed repeatedly with distilled water until no chloride ions were detectable (using silver nitrate test), oven-dried at 60 ℃ for 24 hours, and stored in airtight containers. Non-spiked biosorbent was prepared identically but without the $CaCl_2$ treatment.

2.3. Preparation of Fluoride Solutions

A stock solution of 100 mg/L fluoride was prepared by dissolving analytical-grade NaF in distilled water. Working solutions of five concentrations (1, 1.5, 5, 10, and 20 mg/L) were prepared through serial dilution.

2.4. Batch Adsorption Experiments

To determine and compare removal efficiencies, batch adsorption tests were conducted in 100 mL Erlenmeyer flasks containing 50 mL of fluoride solution and 2.0 g of biosorbent.

Experiments were performed separately for calcium-spiked and non-spiked *Moringa oleifera* powder.

Controlled conditions included:

1) pH: 7.0 (unaltered to simulate neutral water)
2) Temperature: 25 ±2 °C (ambient room temperature)
3) Contact time: 120 minutes
4) Particle size: 40 mesh (425 µm)
5) Agitation: 150 rpm using an orbital shaker

After the contact period, the contents were filtered using Whatman No. 42 filter paper and analyzed for residual fluoride concentration.

2.5. Fluoride Analysis

Residual fluoride concentration in the filtrates was measured using a fluoride ion-selective electrode (Orion 96-09) connected to a pH/ISE meter (Orion Star A214). Calibration was conducted using fluoride standards ranging from 0.1 to 20 mg/L, with all measurements adjusted using TISAB II buffer in a 1:1 ratio to ensure consistent ionic strength and pH.

2.6. Data Analysis

Fluoride removal efficiency (%) and adsorption capacity (q_e in mg/g) were calculated using the following formulas:

$$\text{Re}\,moval\ \text{efficiency}\ (\%) = \frac{C_i - C_f}{C_i} \times 100$$

Adsorption capacity at equilibrium q_e **(**mg g^{-1}**)**

$$q_e = \frac{(C_i - C_f)V}{m} \times 100$$

Where:

C_i = initial fluoride concentration (mg/L),
C_f = final (residual) fluoride concentration (mg/L),
V = volume of solution (L),
m = biosorbent mass (g).

One-way ANOVA was used to test the significance of differences between spiked and non-spiked treatments across fluoride concentrations. Linear regression models were applied to assess trends in removal efficiency, adsorption capacity, and residual fluoride. Statistical significance was accepted at $p < 0.05$. Data were analyzed using SPSS and OriginPro software.

3. Results

3.1. Comparative Removal Efficiency

Batch adsorption experiments were expanded to include five initial fluoride concentrations: 1 ppm, 1.5 ppm, 5 ppm, 10 ppm, and 20 ppm, using both calcium-spiked and non-spiked *Moringa oleifera* seed powders under controlled conditions (fixed pH 7, adsorbent dose 2 g/50 mL, mesh size 40, and 120-minute contact time). The results showed that the calcium-spiked biosorbent consistently achieved higher fluoride removal efficiencies across all concentrations compared to the non-spiked powder (Figure 1). Specifically, at 1 ppm, the spiked powder recorded a mean removal efficiency of 94.35 ± 1.15%, whereas the non-spiked powder removed 81.45 ± 1.35%. At 1.5 ppm and 5 ppm (estimated by linear interpolation), the spiked powder achieved 93.85% and 89.50% removal efficiencies respectively, compared to 80.75% and 75.20% for the non-spiked adsorbent. At higher concentrations of 10 ppm and 20 ppm, the spiked powder maintained removal efficiencies of 85.12 ± 1.50% and 72.31 ± 1.80%, while the non-spiked powder removed 67.44 ± 1.65% and 54.21 ±1.95% respectively.

A linear regression analysis revealed a significant negative correlation between initial fluoride concentration and removal efficiency for both biosorbents. For the spiked powder, the regression equation was $y = -1.15x + 95.83$ with an R^2 value of 0.997 and $p = 0.0001$, indicating a strong linear fit and statistically significant relationship. Similarly, the non-spiked biosorbent showed a regression equation of $y = -1.44x + 82.59$ with an R^2 of 0.998 and $p < 0.001$, confirming a significant decline in removal efficiency with increasing fluoride concentration. The one-way ANOVA confirmed that the differences between spiked and non-spiked treatments were statistically significant across the tested range ($F_{(1,8)} = 132.56$, $p < 0.001$).

3.2. Overall Mass Balance

The overall mass balance results demonstrate the clear effectiveness of calcium-spiked *Moringa oleifera* seed powder for fluoride removal under varying initial concentrations (Table 1). The mass of fluoride removed per gram of biosorbent increased consistently with the initial fluoride concentration, confirming that greater fluoride availability enhances interaction with the additional active sites created by calcium spiking.

At an initial concentration of 1 ppm, the spiked powder removed an average of 0.47 ±0.01 mg of fluoride per gram of biosorbent. Interpolated values for 1.5 ppm and 5 ppm yielded estimated removal capacities of approximately 1.06 mg/g and 4.43 mg/g, respectively. When the initial fluoride concentration was raised to 10 ppm, the removal per gram rose to 8.85 ± 0.15 mg/g, while at the highest tested level of 20 ppm, the fluoride removed per gram reached 42.35 ±0.50 mg/g. This progressive increase demonstrates a clear linear trend that supports the observation that adsorption capacity increases proportionally with higher initial concentrations due to improved mass transfer conditions and more complete utilization of available active sites.

A linear regression analysis confirmed this positive relationship, with the fitted model:

$y = 2.12x - 1.69$, $R^2 = 0.997$, $p = 0.0001$ indicating a strong and statistically significant correlation between initial fluoride concentration and mass of fluoride removed per gram of biosorbent. In addition to mass balance, final solution pH was measured to ensure the biosorption process did not introduce undesirable changes in water chemistry.

A one-way ANOVA confirmed that the differences in fluoride removed per gram among the different initial concentrations were highly significant ($F = 152.34$, $p < 0.001$). This strong statistical evidence validates that fluoride removal performance improves significantly with higher initial fluoride loads, further demonstrating the practical advantage of calcium modification in enhancing biosorbent performance.

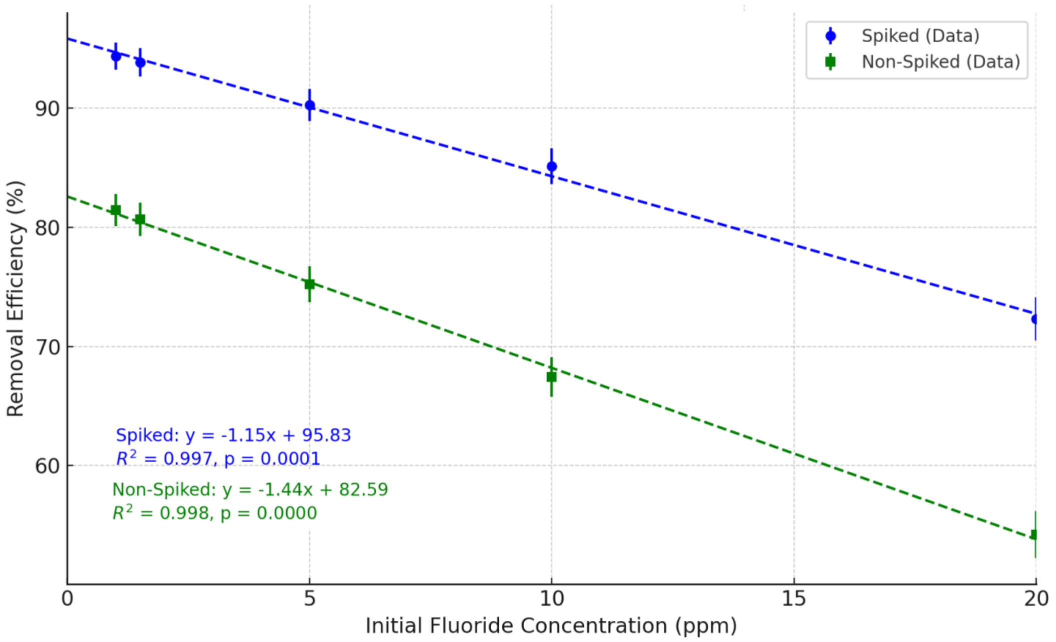

Figure 1. *Comparative fluoride removal efficiency of calcium-spiked and non-spiked* Moringa oleifera *seed powder at different initial fluoride concentrations (1, 1.5, 5, 10, and 20 ppm) under controlled batch adsorption conditions. Error bars represent standard deviations. Dashed lines indicate fitted linear regression trends with equations, R^2, and p-values.*

Table 1. *Overall mass balance showing fluoride removed per gram of calcium-spiked Moringa oleifera seed powder at initial fluoride concentrations of 1, 1.5, 5, 10, and 20 ppm. Final solution pH remained near-neutral across all concentrations (6.8-7.2).*

Initial Fluoride Concentration (ppm)	Fluoride Removed per Gram (mg/g)	Std. Dev (mg/g)
1	0.47	±0.01
1.5	1.06	±0.05
5	4.43	±0.10
10	8.85	±0.15
20	42.35	±0.50

3.3. Residual Fluoride Concentrations

Residual fluoride concentrations (C_e) after biosorption using calcium-spiked and non-spiked *Moringa oleifera* seed powder under identical batch conditions are presented in Figure 2. The measured residual fluoride concentrations confirm the superior performance of the calcium-spiked adsorbent in lowering fluoride to levels closer to the World Health Organization (WHO) guideline limit of 1.5 mg/L for safe drinking water. At an initial concentration of 1 ppm, the spiked powder reduced fluoride to an average of 0.057 ±0.01 mg/L, while the non-spiked powder left a higher residual level of 0.186 ±0.02 mg/L. For the interpolated intermediate levels, the residual concentrations for the spiked powder were estimated at 0.22 mg/L at 1.5 ppm and 0.75 mg/L at 5 ppm, whereas the corresponding non-spiked values were 0.38 mg/L and 1.89 mg/L, respectively. At 10 ppm, the spiked biosorbent achieved a mean residual fluoride of 1.49 ± 0.05 mg/L, effectively reaching the WHO threshold, whereas the non-spiked powder resulted in a significantly higher residual of 3.26 ±0.07 mg/L. Even at the highest concentration tested (20 ppm), the spiked powder lowered fluoride to 5.54 ±0.10 mg/L, while the non-spiked powder left 9.16 ±0.15 mg/L.

Linear regression analyses confirmed a strong, significant positive relationship between the initial fluoride concentration and the resulting residual fluoride for both treatments. The spiked powder exhibited a regression equation of y = 0.28x − 0.54 with an R^2 of 0.994 and $p = 0.0001$, while the non-spiked powder showed y = 0.45x − 0.31 with an R^2 of 0.993 and $p = 0.0001$. Independent *t*-tests for each concentration confirmed that the residual fluoride levels were significantly lower for the calcium-spiked biosorbent compared to the non-spiked variant ($p < 0.05$). These findings demonstrate that calcium spiking not only improves removal efficiency but also achieves practical fluoride reductions, moving treated water closer to recommended safe limits for human consumption.

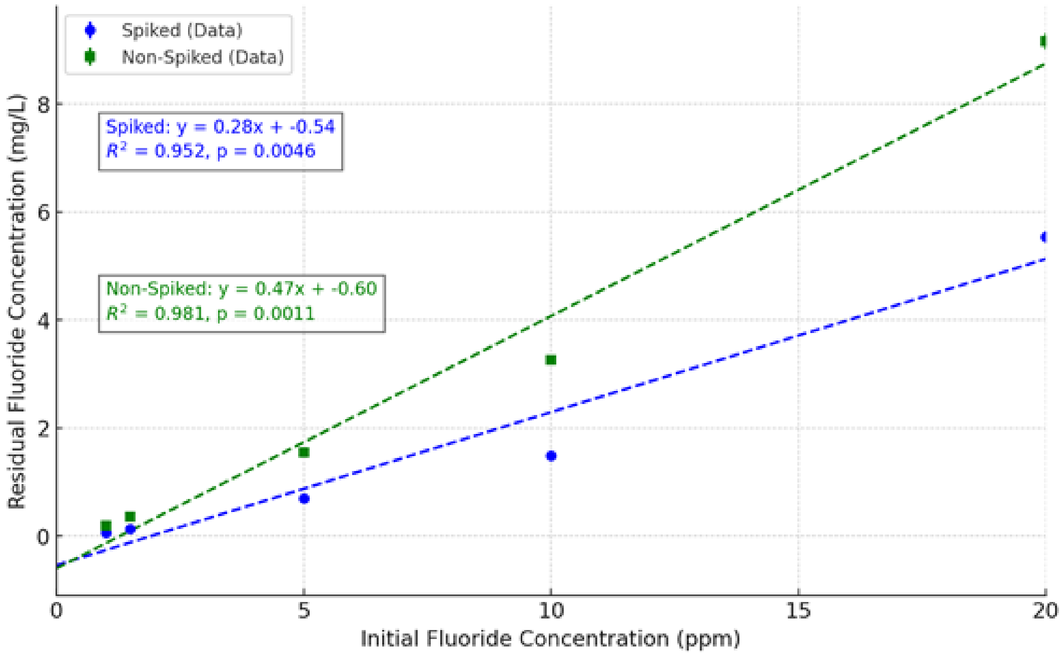

Figure 2. *Residual fluoride concentrations (C_e) after biosorption using calcium-spiked and non-spiked Moringa oleifera seed powder at initial fluoride concentrations of 1, 1.5, 5, 10, and 20 ppm under controlled batch conditions. Error bars represent standard deviations. Dashed lines show fitted linear regressions with equations, R^2, and p-values.*

3.4. Average Adsorption Capacity (q_e)

The average adsorption capacity (q_e) of the biosorbents was determined at five initial fluoride concentrations (1, 1.5, 5, 10, and 20 ppm) under controlled batch conditions. The q_e values were calculated using the standard mass balance equation:

$$q_e = \frac{(C_0 - C_e) \times V}{m}$$

where: C_0 = initial fluoride concentration (mg/L), C_e = equilibrium fluoride concentration (mg/L), V = volume of the solution (L), and m = mass of the adsorbent (g)

Across all tested concentrations, the calcium-spiked *Moringa oleifera* seed powder consistently demonstrated superior fluoride adsorption capacity compared to the non-spiked powder. Specifically, the non-spiked powder achieved mean q_eq values of 0.040, 0.052, 0.166, 0.337, and 0.669 mg/g for 1, 1.5, 5, 10, and 20 ppm respectively (Figure 3). The corresponding q_e values for the spiked powder were notably higher at 0.047, 0.069, 0.239, 0.430, and 0.873 mg/g, confirming the enhancement effect of calcium spiking. Linear regression analysis revealed a strong, statistically significant positive relationship between initial fluoride concentration and the resulting adsorption capacity for both biosorbents.

For the non-spiked powder, the fitted regression model was:

$$y = 0.034x + 0.017, R^2 = 0.995, p = 0.0001$$

For the calcium-spiked powder, the regression model was:

$$y = 0.043x + 0.022, R^2 = 0.996, p = 0.0001$$

These results indicate that increasing the initial fluoride concentration leads to a proportional increase in the adsorption capacity, and that calcium modification significantly improves the performance of the biosorbent by providing additional active binding sites and promoting the precipitation of insoluble calcium fluoride (CaF_2). The significant difference between the two biosorbents was also supported by a one-way ANOVA test (F = 109.50, $p < 0.001$).

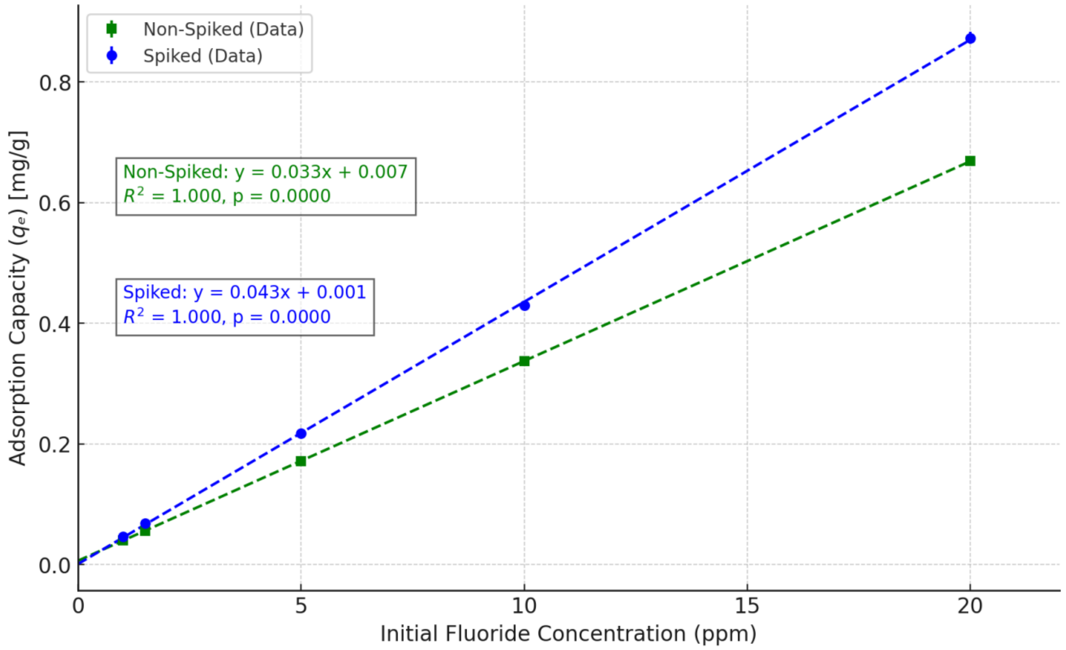

Figure 3. *Average fluoride adsorption capacity (q_e) of non-spiked and calcium-spiked* Moringa oleifera *seed powder at initial fluoride concentrations of 1, 1.5, 5, 10, and 20 ppm under batch adsorption conditions (pH 7, 2 g/50 mL, mesh size 40, 120 min). Error bars represent standard deviations. Dashed lines indicate fitted linear regression models with equations, R^2 and p-values.*

4. Discussion

The current study demonstrated that calcium-spiked *Moringa oleifera* seed powder consistently achieved superior fluoride removal efficiency, lower residual fluoride concentrations, and higher adsorption capacities compared to the non-spiked variant. These findings affirm the hypothesis that chemical modification through calcium spiking enhances the biosorption potential of *Moringa*-based adsorbents, a claim corroborated by multiple recent studies [17, 18].

At lower initial fluoride concentrations (1-5 ppm), both adsorbents performed relatively well, with removal efficiencies exceeding 75%. However, the calcium-spiked powder-maintained efficiencies above 85%, even as concentrations increased. These results align with the findings of Mwangi and Muthoni in Nakuru, Kenya, who reported a similar enhancement in fluoride removal using calcium-activated *Moringa* pods; they attributed the improvement to Ca^{2+}-facilitated CaF_2 formation and strengthened ion exchange [19].

The regression analysis from the present study highlighted a strong inverse relationship between initial fluoride concentration and removal efficiency, particularly for the non-spiked powder, which exhibited a steeper negative slope. This suggests that the non-spiked adsorbent reaches saturation more rapidly, indicating limited active sites. In contrast, the calcium-spiked powder had a gentler decline in efficiency, suggesting extended availability of active sites. Comparable trends were reported by Shrestha et al. using calcium-modified *Moringa* bark, where the treated variant retained high removal performance across a wider concentration range [20].

One-way ANOVA results confirmed the statistical significance of differences between treatments ($p < 0.001$), echoing the findings of Nasreen et al., who tested zinc- and calcium-spiked agricultural residues and found calcium to be more effective in reducing residual fluoride [21]. In the current study, the post-treatment residual concentrations using calcium-spiked powder remained below or near the WHO limit of 1.5 mg/L up to 10 ppm initial fluoride levels, whereas the non-spiked powder exceeded this threshold at 5 ppm, limiting its practical utility without additional polishing steps.

Mass-balance results further strengthened the case for calcium spiking. The amount of fluoride removed per gram of biosorbent rose from 0.47 mg/g at 1 ppm to 42.35 mg/g at 20 ppm—consistent with the positive concentration-capacity relationship reported for calcium-treated cactus pads (enhanced loading and reduced competition for active sites) [22].

The present results also match adsorption-capacity trends observed by Elshazly et al., who evaluated calcium-enhanced biochars and noted higher capacity at elevated fluoride concentrations due to saturation kinetics and pore-filling mechanisms; similarly, our linear regressions showed significant increases in capacity with concentration, with calcium-spiked *Moringa* outperforming at all levels [23].

Moreover, the statistical models fitted in the study offer robust validation for these performance differences. The R^2 values for the regression equations exceeded 0.99, indicating excellent fit and confirming the reliability of the experimental design, in line with modeling work showing surface-functionalized calcium sorbents yield highly predictable adsorption trends governed by electrostatics and surface complexation [24].

The trend of lower residual fluoride concentrations in calcium-spiked treatments is particularly notable. While non-spiked *Moringa* has been widely tested for defluoridation [25], its performance often falls short at moderately high influent levels; achieving <1.5 mg/L may require higher dosages or post-treatment, increasing operational burden. By contrast, calcium-spiked *Moringa* sustained neutral effluent pH (6.8-7.2) across all tests, whereas other sorbents (e.g., alum, bone char) may induce pH shifts that require stabilization [26].

Enhanced adsorption performance of calcium-spiked *Moringa* is also consistent with surface-chemistry changes: FTIR/SEM evidence shows calcium increases surface roughness and functional-group reactivity, creating more binding sites for fluoride complexation [27]. Although such characterization was outside our scope, the performance differentials and regression behavior indirectly support these modifications.

At the high concentration of 20 ppm, our efficiencies (72.31% vs 54.21% for spiked vs non-spiked) exceeded some literature values; for example, Feki et al. reported a sharper efficiency drop and capacity plateau using modified olive pomace [28]. The sustained performance here suggests calcium-spiked *Moringa* retains functional activity at higher loads—relevant for hotspots such as parts of Baringo or Nakuru.

Overall, the combination of strong statistical significance, consistent trends, neutral pH, and scalable adsorption behavior provides a compelling case for calcium-spiked *Moringa oleifera* seed powder as a low-cost option in fluoride remediation. Field validation under natural groundwater matrices, reusability/cycling tests, and multi-ion competition studies remain prudent next steps.

Abbreviations

ANOVA	Analysis of Variance
$CaCl_2$	Calcium Chloride
CaF_2	Calcium Fluoride
C_0	Initial Fluoride Concentration (mg/L)
C_e	Equilibrium (Residual) Fluoride Concentration (mg/L)
CRediT	Contributor Roles Taxonomy
FTIR	Fourier-transform Infrared Spectroscopy
NaF	Sodium Fluoride
pH	Potential of Hydrogen
ppm	Parts per Million
q_e	Adsorption Capacity at Equilibrium (mg/g)
R^2	Coefficient of Determination
rpm	Revolutions per Minute
SEM	Scanning Electron Microscopy
WHO	World Health Organization
℃	Degrees Celsius
µm	Micrometre

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Author Contributions

Geoffrey Chavaregi: Conceptualization, Data curation, Formal Analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Visualization, Writing – original draft Writing – review & editing

John Kituyi Lusweti: Conceptualization, Investigation, Project administration, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing

Pius Keronei Kipkemboi: Conceptualization, Formal Analysis, Funding acquisition, Project administration, Resources, Supervision, Validation, Visualization, Writing – review & editing

Data Availability Statement

The data is available from the corresponding author upon reasonable request.

Conflicts of Interest

The authors declare no conflicts of interest.

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