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Article

Investigation of the removal of physicochemical pollutants in the Kızılırmak River by aluminum sulfate and iron sulfate coagulants

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Abstract: This study evaluated the optimal dosage and pH for removing turbidity, chloride, alkalinity, total dissolved solids (TDS), and sulfate from Kızılırmak River water using iron sulfate and aluminum sulfate (10–60 mg/L). Maximum turbidity removal efficiencies were 98.84% for iron sulfate and 78.99% for aluminum sulfate at pH 4.5. Chloride removal was 77.12% at pH 7.0 for aluminum sulfate and 74.33% at pH 6.0 for iron sulfate. Aluminum sulfate reduced alkalinity by 90.40% at pH 8.0, while iron sulfate achieved 99.21% removal at pH 4.5. TDS removal efficiencies were 99.58% for aluminum sulfate at pH 8.0 and 95.61% for iron sulfate, although total dissolved solids concentrations increased with dosage. Sulfate removal was 97.85% at pH 6.0 for aluminum sulfate and 85.92% at pH 7.0 for iron sulfate. The statistical analysis was conducted using IBM SPSS Statistics 25 to assess the relationships between coagulant type, pH, and dosage on pollutant removal. Response Surface Methodology (RSM) was applied, and analysis of variance (ANOVA) was used to determine significance. The model explained 70.7% of variance (R^2 = 0.707, p < 0.001). pH (p = 0.003), pH^2 (p = 0.002), and dosage^2 (p = 0.049) were significant. Kernel Ridge Regression was used for TDS removal due to overestimation in RSM. Both coagulants were effective in removing pollutants, with optimal performance depending on pH and dosage. Aluminum sulfate exhibited higher turbidity and alkalinity removal at certain pH levels, while iron sulfate achieved greater sulfate and TDS removal under acidic conditions.

Keywords: aluminum sulfate; coagulation; iron sulfate; Kızılırmak River; jar test

1. Introduction

Water pollution has become a major global issue, necessitating advanced and effective treatment methods to ensure water quality and safety. Among various water treatment techniques, coagulation is a widely used process that facilitates the removal of suspended particles, organic matter, and various contaminants from water and wastewater [1]. Coagulation-flocculation alone is highlighted as a cost-effective method with high removal efficiencies for various pollutants [2] and occurs through numerous physicochemical interactions [3]. Traditionally, coagulants such as aluminum sulfate ($Al_2(SO_4)_3$) and iron sulfate ($Fe_2(SO_4)_3$) have been extensively employed in water treatment plants due to their effectiveness in destabilizing colloidal particles and aggregating them into larger flocs, which can then be removed via sedimentation or filtration [4]. The selection of a suitable coagulant depends on various factors, including water chemistry, pH levels, and the nature of the pollutants present. While aluminum sulfate remains one of the most commonly used coagulants due to its cost-effectiveness and availability, ferric sulfate has gained attention for its ability to perform efficiently across a broader pH range and its effectiveness in removing natural organic matter (NOM) and heavy metals [3,5–8]. Recent studies have focused on optimizing coagulation efficiency by exploring coagulant dosage, pH variations, and the potential environmental concerns associated with residual metal concentrations in treated water [9]. Research indicates that excessive use of aluminum-based coagulants may lead to increased residual aluminum levels, which have been linked to potential health risks [10,11]. On the other hand, ferric sulfate has demonstrated better performance in minimizing sludge production and enhancing turbidity removal under specific conditions [12].

In this study, the application of coagulation in the treatment of water from the Kızılırmak River, one of Turkey's longest rivers, is examined. The river is subject to various sources of pollution due to agricultural, industrial, and domestic discharges. The high turbidity and variable composition of Kızılırmak River water present challenges for effective water treatment, making the selection of an appropriate coagulant crucial for optimizing water quality.

This study aims to evaluate and compare the effectiveness of aluminum sulfate and ferric sulfate in water treatment under varying conditions. By analyzing key performance indicators such as turbidity removal, residual coagulant concentrations, and optimal dosage, this research contributes to the ongoing efforts to enhance water treatment efficiency. Furthermore, the findings provide insights into sustainable coagulant selection and its implications for large-scale water treatment applications. The results of this study will support decision-making in selecting appropriate coagulants, optimizing operational parameters, and minimizing potential environmental impacts.

2. Materials and methods

2.1. Materials

Iron sulfate (CAS 7782-63-0) and aluminum sulfate (CAS 17927-65-0) were used as coagulants in the study, purchased from Sigma-Aldrich (Munich, Germany). Sartorius Stedim Biotech GmbH (85037-539-92) (Göttingen, Germany) for the determination of suspended solids concentration of Kızılırmak River, pH Bench Top Meter Model PL 700 PV (Taiwan) for pH, conductivity, and temperature measurements, MTOPS Jar Tester for Jar Test measurements SF6 (Seoul, Korea), and Velp Scientifica TB1 (Italy) were used for turbidity measurement. The PG Instruments T60 Visible Spectrophotometer (United Kingdom) was used for sulfate analysis. While all other studies of the water samples were conducted in the Environmental Engineering Department laboratory, the sulfate analysis was performed in the laboratory of the Institute of Science.

2.2. Water samples

All samples were obtained from the Avanos District, situated inside the provincial boundaries of Nevşehir along the Kızılırmak River (**Figure 1**). The river receives contributions from groundwater, precipitation, and rainfall runoff, while a wastewater treatment facility discharges effluent into the river around 2 km upstream. The measuring station is situated at a latitude of 38°43′3.40″N and a longitude of

34°51′13.48″E. Water samples were collected in January 2022. Samples were obtained using a plastic container approximately 3 m from the shore and 20–30 centimeters beneath the water's surface. The samples were sent to the Environmental Engineering Sciences Department of Nevşehir Hacı Bektaş Veli University within one hour post-collection and maintained at 4 °C. Samples were examined within one week of collection.

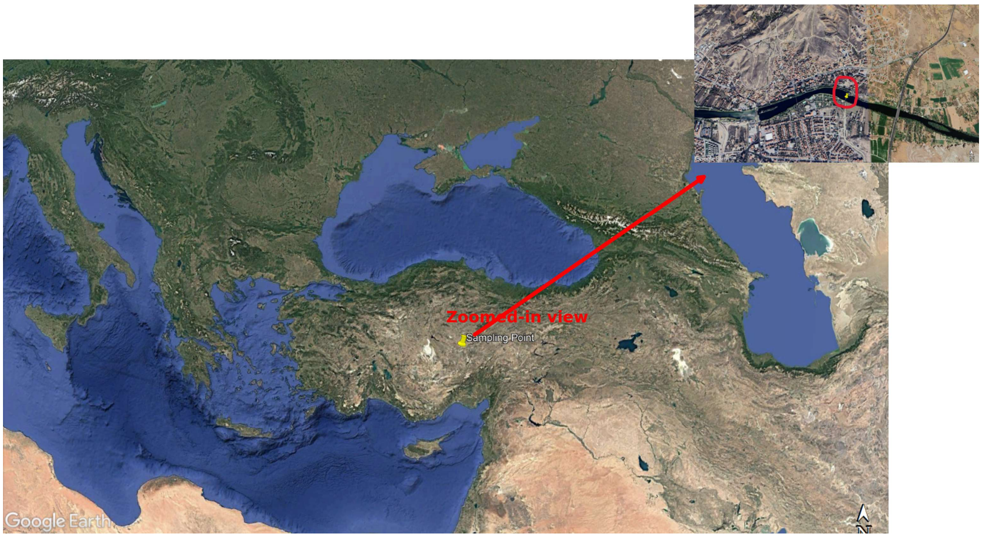

Figure 1. Sampling location and basin region [11,12].

2.3. Coagulants

The study used two different inorganic coagulants: iron sulfate ($FeSO_4 \cdot 7H_2O$) and aluminum sulfate ($Al_2(SO_4)_3) \cdot 18H_2O$. The solutions prepared by dissolving them in deionized water had a concentration of 1% by weight.

2.4. Jar test study

The coagulation tests were performed by using a bench-scale jar test apparatus on the collected samples of 1 L volume used to study the performance of the coagulants on a six-stirrer MTOPS Jar Tester SF6 (**Figure 2**) at room temperature with experimental characteristics as summarized in **Table 1**. To ensure the reliability and reproducibility of the results, blank control experiments and at least three parallel experiments were conducted for each condition.

Table 1. Jar test study experimental characteristics.

Characteristics	**Description**
Coagulants	Alum and Iron Sulfate
Coagulant dosage range	10–60 (mg/L)
Solution pH range	4.5–9.5
Rapid mixing	1 min at 200 rpm
Slow mixing	20 min at 30 rpm
Settling time	1 h

Six different coagulant doses (10, 20, 30, 40, 50, and 60 mg/L) and five different pH values (4.5, 6.0, 7.0, 8.0, and 9.5) were selected for the analyses. Initial pH adjustments of 0.1 mol L^{-1} were provided with HCl and NaOH solutions. After the coagulation process was completed, alkalinity analyses (2310 B.) titration, chloride analyses (4500-Cl-), argentometric, and sulfate analyses (4500-SO_4^{2-}. E) were based on turbidimetric methods for all samples according to standard methods [13]. The pH Bench Top Meter Model PL 700 PV was used for pH, conductivity, and temperature measurements, and the Velp Scientifica TB1 (Italy) device was used for turbidity measurements. TDS values were estimated using electrical conductivity (EC) based on a proportional relationship, given as Equation (1):

$$TDS = 0.60 \times EC \quad (1)$$

where the coefficient (0.60) represents the empirical conversion factor.

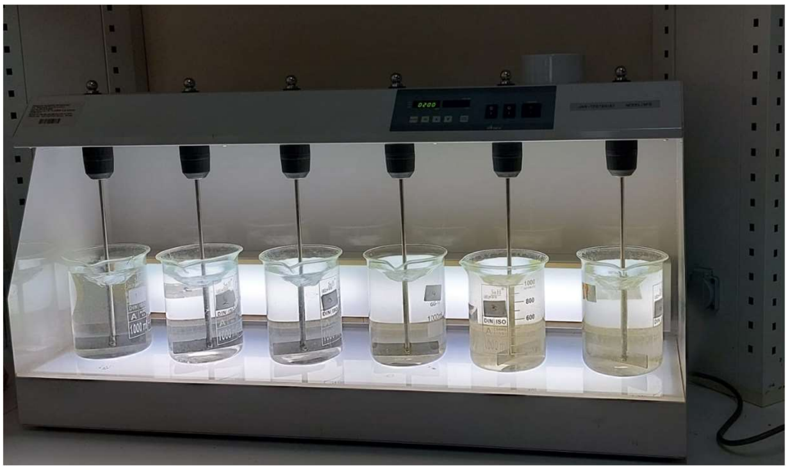

Figure 2. Jar test apparatus in coagulation experiment.

2.5. Statistical analyses

The relationships between the independent variables (coagulant type, pH, and coagulant dosage) and the dependent variables (turbidity, chloride, alkalinity, TDS, and sulfate removal efficiencies) were statistically evaluated using IBM SPSS Statistics 25.

To determine the significance and influence of these factors on pollutant removal efficiency, an RSM analysis was conducted. The model included linear, quadratic, and interaction terms to evaluate the combined effects of pH and coagulant dosage. The statistical significance of the model and individual terms was assessed using ANOVA. The adequacy of the model was determined based on the coefficient of determination (R^2) and adjusted R^2, ensuring a reliable fit to the experimental data.

Furthermore, residual analysis was performed to detect potential outliers and assess model validity. In cases where the RSM model exhibited overestimation or poor fit (e.g., TDS removal), alternative statistical approaches such as logistic regression and Kernel Ridge Regression were employed to enhance predictive accuracy and prevent overfitting.

All statistical analyses were conducted at a 95% confidence level ($p < 0.05$) to confirm the significance of the findings.

3. Results and discussion

3.1. River water characteristics

Raw water was collected from the Kızılırmak, Turkey's longest river, and its physicochemical properties are provided in **Table 2**.

Table 2. Physicochemical parameters of Kızılırmak River.

Physicochemical parameters	Value
Temperature (°C)	4.2
pH	7.62
Turbidity (NTU)	6.95
Alkalinity (mg/L as $CaCO_3$)	256
Chloride (mg/L)	437.51
Sulfate (mg/L)	34.07
TDS (mg/L)	437.4
TSS* (mg/L)	0.005

*TSS: Total Suspended Solids.

3.2. Coagulation results

The research employed two distinct coagulants: iron sulfate and aluminum sulfate. It examined the removal efficacy of these two coagulants on turbidity, alkalinity, chloride, TDS, and sulfate. Coagulant doses and starting pH levels were varied for this purpose. The coagulant dosages used were 10, 20, 30, 40, 50, and 60 mg/L, whereas the pH values ranged from 4.5 to 9.5, specifically at 4.5, 6.0, 7.0, 8.0, and 9.5.

Figure 3 compares turbidity removal efficiencies of aluminum sulfate and iron sulfate across various pH values (4.5–9.5). Aluminum sulfate consistently showed superior turbidity removal performance [14]. At acidic pH, both coagulants effectively form positively charged hydroxide species that enhance coagulation through charge neutralization [15,16]. Optimal removal was highest at pH 4.5, with aluminum sulfate (98.84%) outperforming iron sulfate (78.99%). At neutral pH, insoluble hydroxide precipitates facilitated sweep-flocculation, with aluminum sulfate still exhibiting higher efficiency (87.76%) [17]. Under alkaline conditions, aluminum sulfate maintained high removal at lower dosages, while iron sulfate's effectiveness significantly decreased due to reduced hydroxide solubility. Overall, aluminum sulfate provided broader operational effectiveness and stability across the tested conditions.

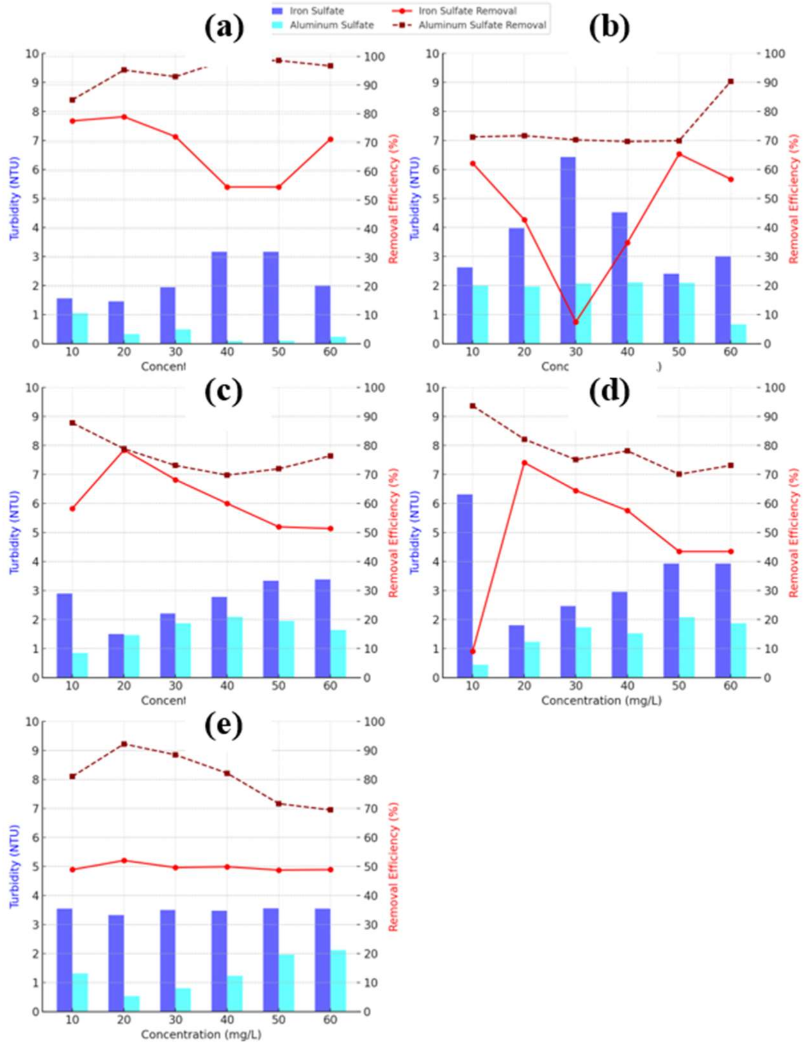

Figure 3. Effect of pH and coagulant dosage on turbidity removal efficiency using iron sulfate and aluminum sulfate: **(a)** pH 4.5; **(b)** pH 6.0; **(c)** pH 7.0; **(d)** pH 8.0; **(e)** pH 9.5.

Figure 4 compares chloride removal efficiencies of aluminum sulfate and iron sulfate across various pH values (4.5–9.5). At acidic pH (4.5), iron sulfate achieved higher chloride removal (73.51%) than aluminum sulfate (67.14%), mainly due to effective ionic interactions facilitated by charged metal hydroxide species. Near neutral pH (6.0–7.0), iron sulfate was more effective at pH 6.0, whereas aluminum sulfate performed better at pH 7.0, influenced by differences in hydroxide precipitation and adsorption capacity. Under alkaline conditions (8.0–9.5), aluminum sulfate generally exhibited superior efficiency (up to 74.29%), attributed to stable hydroxide precipitates enhancing chloride adsorption, despite decreasing efficiency at certain higher dosages. Overall, iron sulfate was preferable at lower pH levels, while aluminum sulfate provided better chloride removal at neutral to alkaline conditions [18–20].

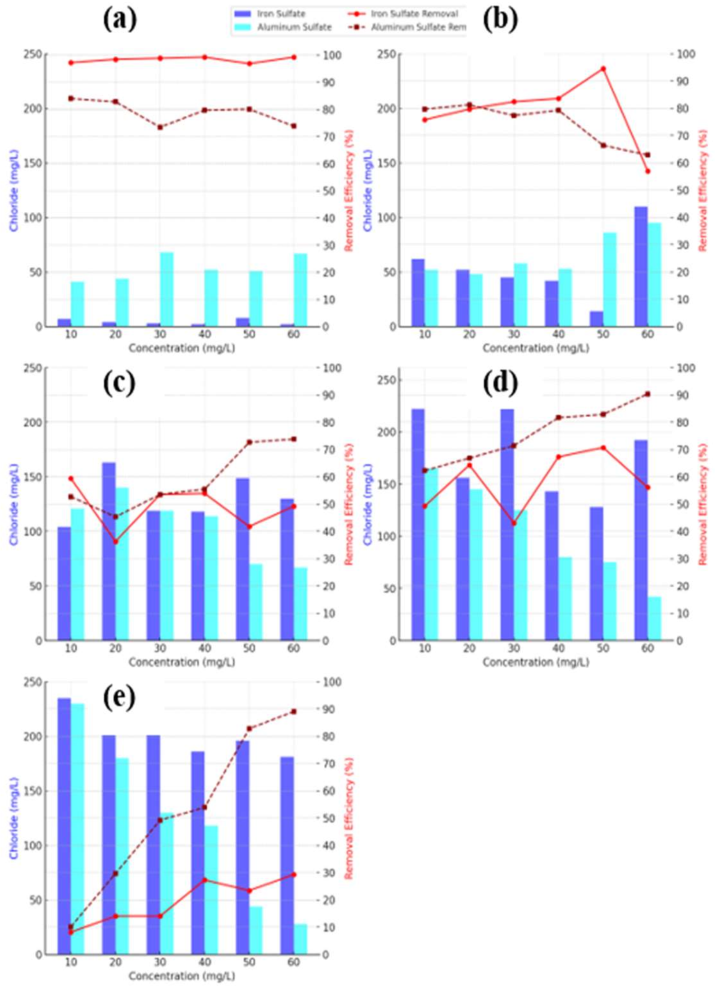

Figure 4. Effect of pH and coagulant dosage on chloride removal efficiency using iron sulfate and aluminum sulfate: **(a)** pH 4.5; **(b)** pH 6.0; **(c)** pH 7.0; **(d)** pH 8.0; **(e)** pH 9.5.

Figure 5 compares alkalinity removal efficiencies for aluminum sulfate and iron sulfate at varying pH levels (4.5–9.5). Iron sulfate showed higher removal at acidic pH (up to 99.21% at pH 4.5), driven by rapid neutralization reactions. Aluminum sulfate performed better at neutral and alkaline conditions, achieving maximum removal efficiency (90.40% at pH 8.0) due to effective precipitation of aluminum hydroxides. Overall, iron sulfate is preferable at low pH, whereas aluminum sulfate demonstrates superior alkalinity removal at higher pH conditions, reflecting their distinct precipitation and neutralization mechanisms [21]. It is important to determine the optimal pH ranges for maximum utilization of aluminum sulfate [22].

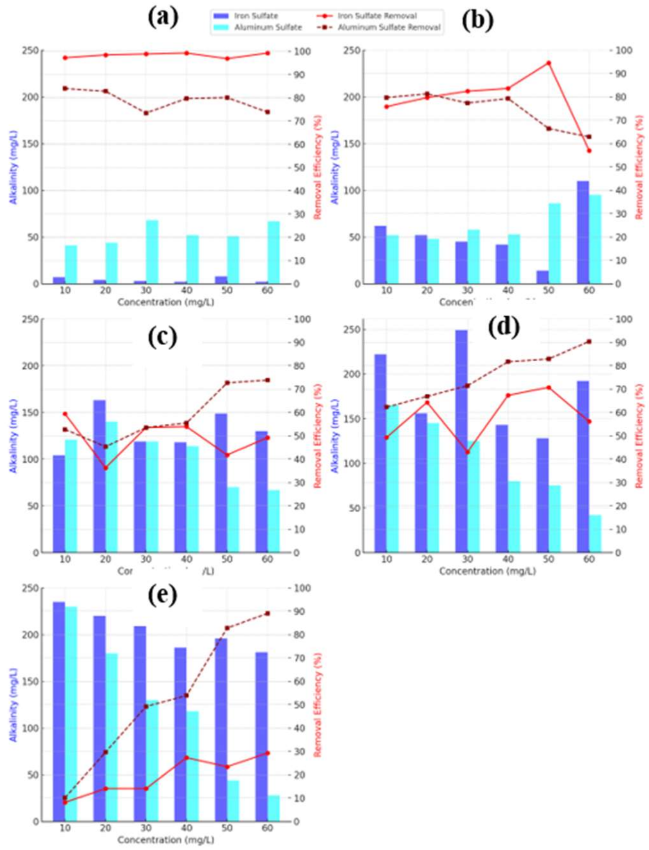

Figure 5. Effect of pH and coagulant dosage on alkalinity removal efficiency using iron sulfate and aluminum sulfate: **(a)** pH 4.5; **(b)** pH 6.0; **(c)** pH 7.0; **(d)** pH 8.0; **(e)** pH 9.5.

Figure 6 illustrates TDS removal efficiencies of aluminum sulfate and iron sulfate across varying pH conditions (4.5–9.5). At lower pH (4.5 and 6.0), both coagulants exhibited limited or negative effects on TDS removal, indicating that coagulation at acidic conditions primarily targets particulate matter rather than dissolved ions [23–25]. Optimal TDS removal was notably achieved at neutral (pH 7.0) and alkaline (pH 8.0–9.5) conditions [26]. At pH 7.0, iron sulfate demonstrated significantly higher removal (78.46%) than aluminum sulfate (11.93%) at lower dosages, suggesting iron hydroxide precipitates effectively adsorb dissolved solids. At pH 8.0, both coagulants performed optimally at low dosages (10 mg/L), with aluminum sulfate showing superior efficiency (99.58%) due to effective precipitation and removal mechanisms. However, increasing dosages adversely affected performance, causing higher TDS levels, possibly due to the introduction of excess ions from the coagulants themselves. At pH 9.5, iron sulfate achieved maximum removal (95.61%) at 30 mg/L, while aluminum sulfate's effectiveness declined at higher dosages. Overall, optimal TDS removal by both coagulants occurs at neutral to alkaline pH, with performance significantly decreasing at higher dosages due to increased residual ionic content.

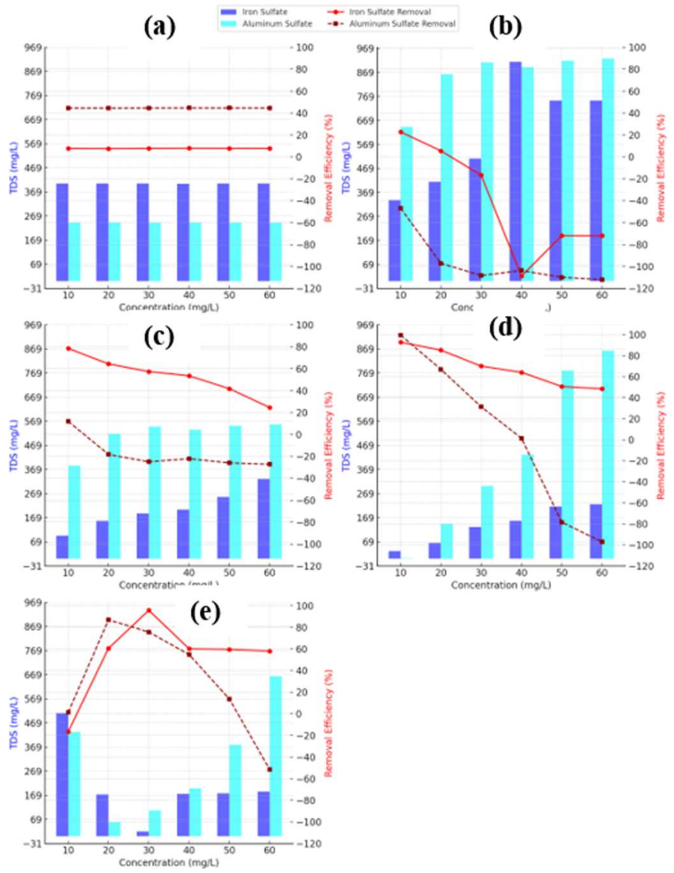

Figure 6. Effect of pH and coagulant dosage on TDS removal efficiency using iron sulfate and aluminum sulfate: **(a)** pH 4.5; **(b)** pH 6.0; **(c)** pH 7.0; **(d)** pH 8.0; **(e)** pH 9.5.

Figure 7 presents sulfate removal efficiencies for aluminum sulfate and iron sulfate across varying pH values (4.5–8.0). At acidic conditions (pH 4.5), iron sulfate demonstrated notably higher removal efficiency (69.94%) compared to aluminum sulfate (29.08%), mainly due to effective formation of soluble and positively charged iron hydroxide species facilitating sulfate adsorption [27,28]. At neutral pH (7.0), iron sulfate again showed superior performance (85.92%) compared to aluminum sulfate (49.51%), attributed to effective precipitation and adsorption by iron hydroxides. At intermediate pH (6.0), iron sulfate removal remained relatively high, though aluminum sulfate became more effective at specific dosages. At slightly alkaline conditions (pH 8.0), both coagulants exhibited variable removal efficiencies depending on dosages, with iron sulfate performing better at lower dosages and aluminum sulfate more efficient at mid-range dosages. Overall, iron sulfate generally provided higher sulfate removal at lower pH values, whereas aluminum sulfate was effective primarily at neutral and slightly alkaline pH conditions, highlighting the dependence of sulfate removal on coagulant dosage and solution pH [29].

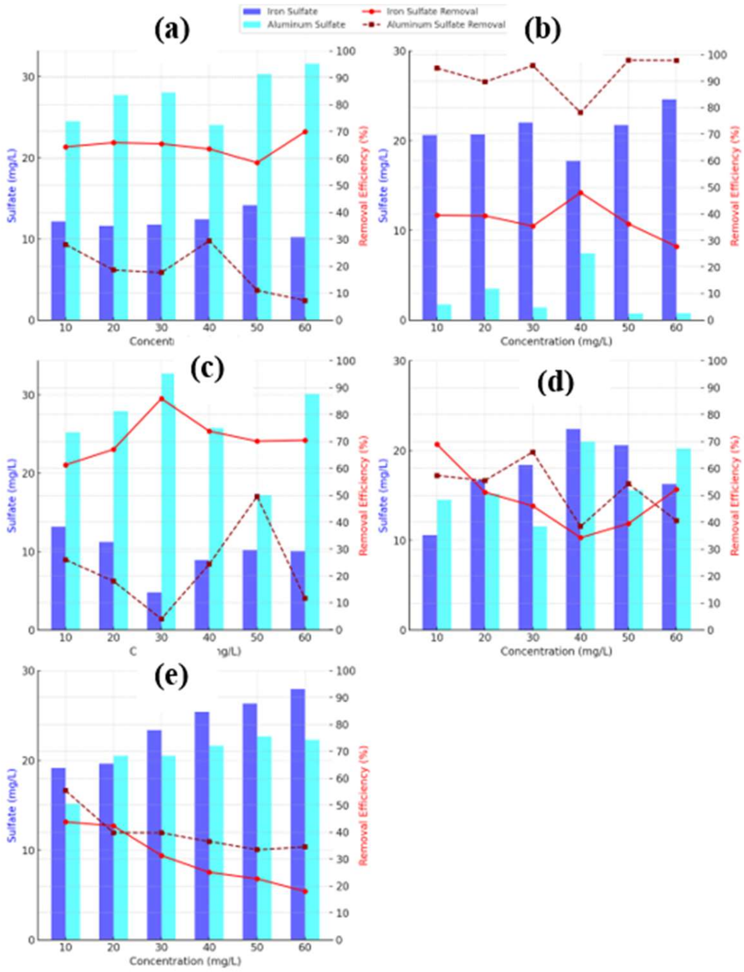

Figure 7. Effect of pH and coagulant dosage on sulfate removal efficiency using iron sulfate and aluminum sulfate: **(a)** pH 4.5; **(b)** pH 6.0; **(c)** pH 7.0; **(d)** pH 8.0; **(e)** pH 9.5.

3.3. Statistical analyses

To determine the statistical significance of the relationships between the independent variables (coagulant type, pH, and coagulant dosage) and the dependent variables (turbidity, chloride, alkalinity, TDS, and sulfate removal efficiencies), RSM analysis was conducted.

The results indicate that the model explains approximately 70.7% of the variance (R^2 = 0.707), demonstrating high statistical significance ($p < 0.001$). pH ($p = 0.003$) and pH^2 ($p = 0.002$) were identified as key factors influencing coagulation performance. The quadratic term suggests that the impact of pH is non-linear.

Concentration^2 ($p = 0.049$) was also statistically significant, indicating that the effect of coagulant dosage follows a second-degree relationship rather than a simple linear trend. The influence of coagulant type (iron vs. aluminum) was near the statistical significance threshold ($p = 0.073$), suggesting that although the general performance may differ between the two coagulants, their effectiveness varies depending on pH and dosage levels.

These findings confirm that pH and dosage significantly influence coagulation efficiency, while the coagulant type exhibits performance variations depending on specific conditions.

3.3.1. Optimal coagulant dosage and pH conditions for turbidity reduction based on RSM analysis

The results presented in **Table 3** demonstrate that the effectiveness of the coagulants is strongly dependent on the specific combinations of pH and dosage levels. Visualizations of the results are shown in **Figure 8**.

Table 3. Predicted optimal coagulation dosage and pH for enhanced turbidity removal.

	Iron sulfate	Aluminum sulfate
Dosage (mg/L)	10	10
pH	9.5	4.5
Removal efficiency (%)	100	100

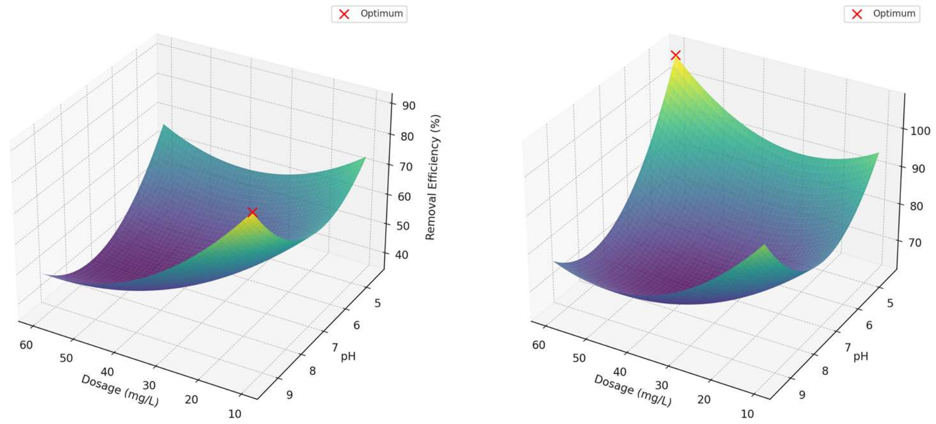

Figure 8. RSM visualization of optimum dosage and optimum pH values for turbidity removal.

3.3.2. Optimal coagulant dosage and pH conditions for chloride reduction based on RSM analysis

The results in **Table 4** indicate that both coagulants achieved the highest chloride removal efficiency under low pH and high dosage conditions. Visualizations of the results are shown in **Figure 9**.

Table 4. Predicted optimal coagulation dosage and pH for enhanced chloride removal.

	Iron sulfate	Aluminum sulfate
Dosage (mg/L)	60	10
pH	4.5	4.5
Removal efficiency (%)	77.66	100

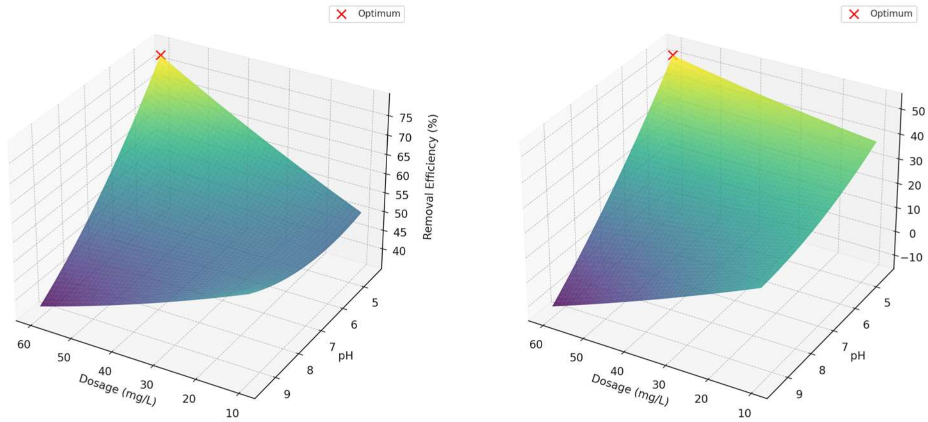

Figure 9. RSM visualization of optimum dosage and optimum pH values for chloride removal.

3.3.3. Optimal coagulant dosage and pH conditions for alkalinity reduction based on RSM analysis

The results show that higher efficiency is achieved at lower pH levels in terms of alkalinity removal and that the efficiency may decrease if the optimum dosage levels are exceeded (**Table 5**). Visualizations of the results are shown in **Figure 10**.

Table 5. Predicted optimal coagulation dosage and pH for enhanced alkalinity removal.

	Iron sulfate	**Aluminum sulfate**
Dosage (mg/L)	20.86	22.08
pH	4.5	4.5
Removal efficiency (%)	100	84.98

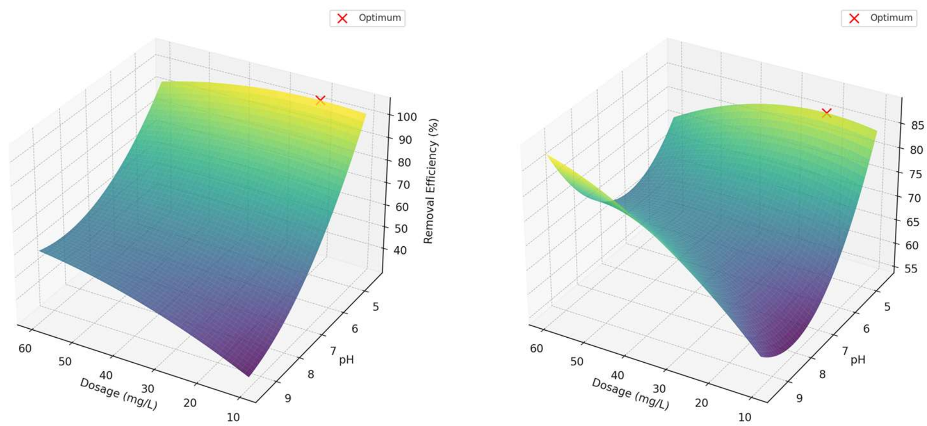

Figure 10. RSM visualization of optimum dosage and optimum pH values for alkalinity removal.

3.3.4. Optimal coagulant dosage and pH conditions for TDS reduction based on RSM analysis

The results indicate that iron sulfate exhibited an overestimated performance in TDS removal at high pH levels (211.98%). Given the need for further validation, Kernel Ridge Regression analysis was conducted for iron sulfate to refine the predictions. Accordingly, the results for iron sulfate, obtained through Kernel Ridge Regression analysis, and for aluminum sulfate, obtained through RSM analysis, are presented in **Table 6** and **Figure 11**.

Table 6. Predicted optimal coagulation dosage and pH for enhanced TDS removal.

	Iron sulfate	**Aluminum sulfate**
Dosage (mg/L)	10	10
pH	6.95	4.5
Removal efficiency (%)	30.35	29.46

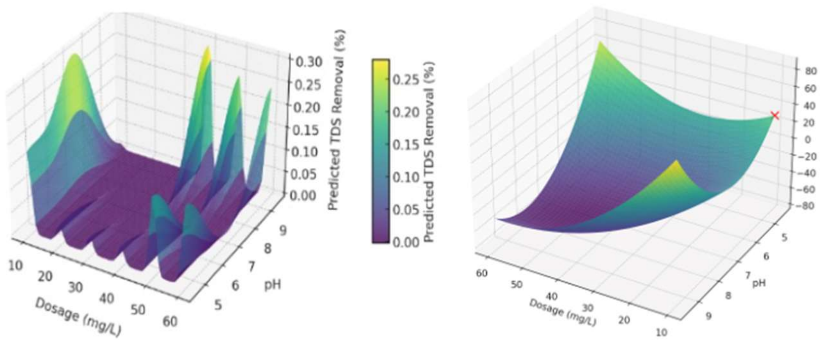

Figure 11. RSM visualization of optimum dosage and optimum pH values for TDS removal.

3.3.5. Optimal coagulant dosage and pH conditions for sulfate reduction based on RSM analysis

According to RMS analysis, the results show that ferrous sulfate is more effective for sulfate removal at medium pH levels, while aluminum sulfate performs better at higher pH levels (**Table 7**). Visualizations of the results are shown in **Figure 12**.

Table 7. Predicted optimal coagulation dosage and pH for enhanced TDS removal.

	Iron sulfate	**Aluminum sulfate**
Dosage (mg/L)	49.21	15.99
pH	6.21	7.66
Removal efficiency (%)	68.63	61.59

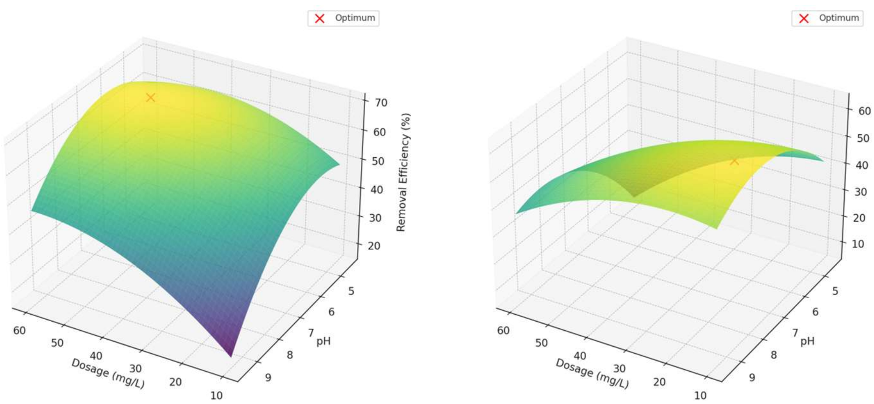

Figure 12. RSM visualization of optimum dosage and optimum pH values for sulfate removal.

4. Conclusion

This study evaluated the effectiveness of aluminum sulfate and iron sulfate in removing turbidity, chloride, alkalinity, total dissolved solids (TDS), and sulfate from Kızılırmak River water under varying pH and dosage conditions. The results showed that aluminum sulfate outperformed iron sulfate, particularly in turbidity, chloride, and TDS removal, while iron sulfate was more effective in alkalinity reduction at low pH. Statistical analysis confirmed that pH was the most significant factor influencing coagulation efficiency, whereas coagulant dosage had a limited impact.

Future studies should measure aluminum and iron residuals in treated water to assess potential health risks and determine the need for additional post-treatment steps. The findings should be tested on different water sources, particularly those with higher turbidity and organic matter, to assess the broader applicability of the results. Given the limited impact of dosage variations, more research is needed to refine dosing strategies, ensuring effective treatment with minimal chemical use. Investigating natural coagulants, such as plant-based alternatives, could reduce environmental and health risks while maintaining treatment efficiency. Future work should evaluate the economic feasibility and long-term stability of using aluminum and iron sulfate in large-scale applications.

By addressing these aspects, future studies can enhance coagulation performance, optimize chemical use, and contribute to more sustainable water treatment solutions.

Author contributions: Conceptualization, SGD; methodology, SGD; validation, SGD; formal analysis, SGD; investigation, KÖ and SGD; resources, KÖ; data curation, SGD; writing—original draft preparation, SGD and KÖ; writing—review and editing, SGD and KÖ; visualization, SGD; supervision, SGD. All authors have read and agreed to the published version of the manuscript.

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