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Article Evaluating the Effectiveness of Coagulation–Flocculation Treatment Using Aluminum Sulfate on a Polluted Surface Water Source: A Year-Long Study

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Abstract: Safeguarding drinking water is a major public health and environmental concern because it is essential to human life but may contain pollutants that can cause illness or harm the environment. Therefore, continuous research is necessary to improve water treatment methods and guarantee its quality. As part of this study, the effectiveness of coagulation-flocculation treatment using aluminum sulfate (Al₂(SO₄)₃) was evaluated on a very polluted site. Samplings were taken almost every day for a month from the polluted site, and the samples were characterized by several physicochemical properties, such as hydrogen potential (pH), electrical conductivity, turbidity, organic matter, ammonium (NH_{4}) , phosphate (PO_{4}^{3-}) , nitrate (NO_{3}^{-}) , nitrite (NO_{2}^{-}) , calcium (Ca^{2+}) , magnesium (Mg^{2+}) , total hardness (TH), chloride (Cl⁻⁾, bicarbonate (HCO₃⁻), sulfate (SO₄²⁻), iron (Fe³⁺), manganese (Mn^{2+}) , aluminum (Al^{3+}) , potassium (K^+) , sodium (Na^+) , complete alkalimetric titration (TAC), and dry residue (DR). Then, these samples were treated with $Al_2(SO_4)_3$ using the jar test method, which is a common method to determine the optimal amount of coagulant to add to the water based on its physicochemical characteristics. A mathematical model had been previously created using the support vector machine method to predict the dose of coagulant according to the parameters of temperature, pH, TAC, conductivity, and turbidity. This $Al_2(SO_4)_3$ treatment step was repeated at the end of each month for a year, and a second characterization of the physicochemical parameters was carried out in order to compare them with those of the raw water. The results showed a very effective elimination of the various pollutions, with a very high rate, thus demonstrating the effectiveness of the Al₂(SO₄)₃. The physicochemical parameters measured after the treatment showed a significant reduction in the majority of the physicochemical parameters. These results demonstrated that the coagulation-flocculation treatment with $Al_2(SO_4)_3$ was very effective in eliminating the various



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Copyright: © 2024 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/). pollutions present in the raw water. They also stress the importance of continued research in the field of water treatment to improve the quality of drinking water and protect public health and the environment.

Keywords: water quality; coagulation–flocculation; aluminum sulfate; jar test; physicochemical parameters

1. Introduction

Water quality is a significant indicator of the health of our environment and the safety of our drinking water supply [1,2]. Water quality can be influenced by a variety of factors, including pollution, human activities, climate changes, and water resource management [3,4]. Monitoring water quality is crucial for detecting contaminants and pathogens in the water in order to take measures to protect public health and the environment [1]. Water quality testing may involve chemical, microbiological, and physical analyses to assess the presence and quantity of substances such as nitrate (NO₃⁻), pesticides, heavy metals, pathogenic microorganisms, and organic matter [5]. To improve water quality and eliminate pollution, there are several techniques available depending on the source and type of pollution [6]. Wastewater from households and industries can be treated using technologies such as filtration, sedimentation, disinfection, and reverse osmosis before being discharged into rivers, lakes, or oceans [7].

Recent strides in water treatment signify remarkable progress in addressing micropollutants and substances resistant to conventional processes [8–13]. Within this context, a meticulous exploration of specific coagulants and their optimal dosage becomes indispensable [14]. The safeguarding of environmental sustainability and human health hinges crucially on ensuring the non-toxicity of water treatment agents. In this pivotal context, aluminum sulfate ($Al_2(SO_4)_3$) emerges as a distinguished contender, not only celebrated for its efficacy but also lauded for its commendable non-toxic nature [15]. This attribute underscores its significance in contributing to water treatment strategies that prioritize both environmental conservation and the well-being of human populations [15].

In the realm of water treatment, the deployment of aluminum sulfate has surfaced as a pivotal strategy for eliminating suspended particles, organic matter, metals, and other impurities from drinking water and wastewater [16]. Noted for its effectiveness in forming flocs—particle aggregates—by reacting with impurities in water, aluminum sulfate coagulation offers distinctive advantages [17]. These flocs, being denser than water, readily separate through sedimentation or filtration [18,19]. Beyond its efficacy in removing contaminants such as suspended matter, heavy metals, and specific organic pollutants, aluminum sulfate coagulation is acknowledged for its relative simplicity and cost-effectiveness [20,21]. The dosage of aluminum sulfate assumes particular significance as a critical parameter in achieving optimal treatment efficiency [22]. Determining the appropriate coagulant dosage is pivotal in the design and operation of a water treatment system [23]. Inadequate dosage may lead to treatment inefficiency and compromised water quality, while excessive dosage can result in additional costs and environmental concerns associated with excess coagulant discharge [24]. The multifaceted nature of $Al_2(SO_4)_3$ coagulation allows for its integration with other water treatment methods, ensuring maximum efficiency in reducing water contamination levels [19,25,26]. It can be employed as a pretreatment step before filtration, disinfection, or reverse osmosis, enhancing the overall effectiveness of the water treatment process [27,28].

Optimizing the optimal coagulant dosage can be achieved through laboratory methods such as the jar test [29]. The jar test is a laboratory procedure used to ascertain the optimal amount of coagulant–flocculant to add to water for impurity removal [30]. This test is typically conducted prior to the implementation of a large-scale water treatment system [30]. The test involves taking a sample of raw water and dividing it into six or eight equal-

volume samples in glass containers. Each container is stirred with a different amount of coagulant–flocculant, and the formation of flocs is observed [31]. The containers are then left to settle for a specified period, usually an hour, allowing for the flocs to form and settle at the bottom of the container [32]. After the settling period, water is sampled from each container at different heights and analyzed to determine the quantity of suspended particles, organic matter, and other contaminants [31]. The test results help determine the optimal coagulant–flocculant dosage for achieving the best treated water quality.

Other methods for optimizing the optimal coagulant dosage include using mathematical models to predict the optimal dosage based on characteristics of the raw water, such as hydrogen potential (pH), hardness, and organic content [33]. These models can be employed to determine the optimal coagulant dosage for specific water conditions [33]. It is also important to consider other factors in coagulant dosage optimization, such as water temperature, coagulant concentration, mixing time, and settling time [34]. These factors can affect treatment efficiency and must be taken into account when optimizing the coagulant dosage [34]. Lastly, regular monitoring of treated water quality is essential to ensure that the coagulant dosage used remains optimal and to adjust the dosage if necessary based on changes in raw water characteristics [35]. As there are several environmental factors that influence the coagulation process and can vary significantly depending on the source of raw water, it is important to take these factors into consideration. Mathematical models can help predict the optimal coagulant dosage for specific water conditions, but it is crucial to account for environmental factors and conduct regular water quality monitoring to ensure consistent conditions and effective coagulation [36]. Furthermore, the rising levels of contaminants in raw water due to climate change may necessitate ongoing adaptation of water treatment processes to maintain safe drinking water quality [37]. Thus, water quality is influenced by numerous environmental parameters such as temperature, pH, electrical conductivity, turbidity, organic matter, ammonium (NH $^+$ ₄), phosphate (PO₄ $^{3-}$), NO₃ $^-$, nitrite (NO₂⁻), calcium (Ca²⁺), magnesium (Mg²⁺), total hardness (TH), chloride (Cl⁻), bicarbonate (HCO₃⁻), sulfate (SO₄²⁻), iron (Fe³⁺), manganese (Mn²⁺), aluminum (Al³⁺), potassium (K⁺), sodium (Na⁺), complete alkalimetric titration (TAC), and dry residue (DR) [38–40].

These parameters can originate from various sources including human activities, weather conditions, soil geological characteristics, and local ecology [40]. Regular monitoring of these parameters is crucial to ensure good water quality and safe use for both humans and the environment. This innovative study represents a significant advancement in the understanding and application of $Al_2(SO_4)_3$ as a coagulant in severely polluted environments [40].

The central objective of this research was to comprehensively evaluate the performance and effectiveness of a specific coagulant in treating highly contaminated water over an extended 12-month period. To carry out this detailed assessment, a meticulous methodology was employed. Sample collection occurred nearly every day throughout each month, allowing us to capture potential variations in environmental conditions. These samples underwent a carefully controlled storage and mixing process at the end of each month, producing data representative of the key physicochemical properties found in the collected and mixed contaminated water. A pivotal aspect of this research involved the utilization of a previously validated mathematical model based on the support vector machine method [41,42]. This model was specifically designed to accurately predict the optimal dosage of Al₂(SO₄)₃ required for the coagulation process, taking into account seasonal variations and fluctuations in environmental parameters such as temperature, pH, TAC, conductivity, and turbidity. This Al₂(SO₄)₃ treatment step was systematically repeated at the end of each month throughout the entire study year. After each treatment, we conducted a second comprehensive characterization of the physicochemical parameters to compare the results with those of the original raw water.

Our study stands out due to its unique context and innovative approach in evaluating the efficacy of coagulation–flocculation treatment using $Al_2(SO_4)_3$ for surface water purification, especially within an exceptionally polluted site. Although $Al_2(SO_4)_3$ is a wellestablished water treatment method, our research significantly enriches existing knowledge by focusing on several key aspects:

- Unprecedented Environmental Challenge: Our study addresses the pressing need to
 assess water treatment under severe pollution levels, providing invaluable insights
 into the adaptability of coagulation–flocculation treatment in environments where
 pollutant concentrations surpass controlled settings.
- High-Frequency Sampling Regime: By adopting an innovative daily sampling regimen over a month, our study unveils dynamic pollutant removal trends, showcasing the potential nuances missed with less frequent sampling intervals.
- Advanced Predictive Modeling: The incorporation of a state-of-the-art mathematical model, based on the support vector machine method, advances the practicality of coagulant dosage determination, offering a potentially more efficient and adaptable approach.
- Longitudinal Analysis: Spanning a year with monthly Al₂(SO₄)₃ treatments, our longitudinal approach accounts for seasonal fluctuations, providing insights into the treatment method's performance over extended periods, a unique feature in water treatment research.
- Comprehensive Parameter Characterization: The extensive characterization of a wide array of physicochemical parameters offers an all-encompassing view of water quality before and after treatment, enhancing our understanding of treatment impacts on various facets of water chemistry.
- Diverse Parameter Coverage: Our study encompasses an extensive range of water quality parameters, including turbidity, organic matter, PO₄³⁻, Fe³⁺, Mg²⁺, SO₄²⁻, NH₄⁺, Ca²⁺, Mg²⁺, TH, Cl⁻, and HCO₃⁻, among others, providing a holistic perspective on Al₂(SO₄)₃ efficacy.
- Indirect Parameter Reduction Mechanisms: Demonstrating the intricate interactions of Al₂(SO₄)₃ with water quality parameters, such as NH₄⁺, Ca²⁺, Mg²⁺, TH, Cl⁻, and HCO₃⁻, among others, via mechanisms such as complexation and pH changes, elucidates novel pathways for pollutant removal.

This comprehensive investigation underscores the significance of $Al_2(SO_4)_3$ treatment in pollutant removal and water quality improvement, particularly in severely contaminated contexts. Furthermore, it underscores the perpetual need for ongoing research in water treatment, offering broader implications for the preservation of public health and the environment. Importantly, this research breaks new ground as the first of its kind to delve into $Al_2(SO_4)_3$ utilization as a coagulant in high-pollution scenarios, potentially reshaping perspectives on environmental management and water resource protection.

2. Methods and Materials

2.1. Mode of Collection and Preservation of Water Samples

Over a period of 12 months, a rigorous sampling campaign was conducted daily at the highly polluted surface water site of Oued Chiffa, situated between the regions of Médéa and Blida. Each day, one liter of water was collected from various points within the contaminated area to ensure the representativeness of the samples. At the end of each month, a composite sample of 30 L was created from these daily collections. The specific choice of this site was motivated by its elevated levels of pollution, aiming to assess the effectiveness of coagulation processes in an intensely contaminated environment. It is noteworthy that, during the initial phase of the study, samples were taken from different locations each day. However, for the remainder of the months, the samples were collected from the same locations, in accordance with the indications provided in Figure 1. The study year was 2019, and samples were systematically collected from January to December 2019, providing a comprehensive dataset for analysis.



Figure 1. Mapping of sampling points and visualizations of polluted areas during the study.

During the sampling process, meticulous attention was paid to various critical factors to ensure the accuracy and integrity of the collected samples. Specifically, clean glass bottles were used to preserve the samples, and they were collected from a depth of 15 to 30 cm below the water surface to minimize potential surface layer influences. The pH and temperature of each daily water sample were meticulously measured on-site before initiating the acidification process and subsequent preservation in a refrigerator set at a constant temperature of 4 °C. These critical steps played a central role in ensuring the accuracy of the dataset, allowing for real-time adjustments to align the mixture with actual conditions. By calculating monthly average values before the treatment phase, the aim was to capture a nuanced understanding of temporal variations in water quality, enriching the depth of analytical insights.

To maintain chemical stability and prevent contamination, the samples were acidified with sulfuric acid at a purity of 97% (2 mL/L) [43–45]. The pre-analysis acidification of raw water samples with sulfuric acid is a crucial step, aiming to establish a known and constant pH value to ensure reproducibility and accuracy in subsequent measurements. This process serves various purposes, including fixing the pH at an acidic value (typically lower than 2) to stabilize the measured parameters over time and preventing the precipitation of ions and compounds as insoluble salts at higher pH levels, thereby maintaining solubility and accurate measurements [45]. Additionally, acidification contributes to sample preservation by inhibiting microbial growth and minimizing undesirable chemical reactions that could alter the sample's composition over time. It also facilitates control over the chemical activity of ions in water by maintaining a constant, low pH, ensuring consistent conditions for each analyzed sample. Moreover, optimizing specific chemical reactions under acidic conditions enhances the reactivity of reagents and ensures the accuracy of the overall analysis [43,45].

Additionally, great care was taken to sterilize the bottles, which was achieved through thorough rinsing with distilled water followed by autoclaving, thereby eliminating any risks of cross-contamination [45]. To protect the samples from the influence of light, they were stored in opaque containers. Immediately after collection, the samples were promptly cooled by placing them in a cooler maintained at 4 °C, ensuring optimal preservation by slowing degradation processes [43,45]. Measures were taken to prevent freeze-related alterations when temperatures dropped below freezing. Prior to each analysis, a meticulous homogenization process was systematically carried out with the use of impeccable gloves and avoidance of any contaminating contact [45]. Samples were hermetically sealed to preserve their purity, prevent air contact, and minimize the risk of evaporation. Furthermore,

consistent temperature stability was maintained throughout the process, thanks to the use of insulated containers during transport. Continuous monitoring of storage temperature and sample condition was undertaken meticulously, with each step rigorously documented, including collection dates, storage conditions, and any relevant events associated with each sample.

2.2. Jar Test

In this study, Al₂(SO₄)₃ in octadecahydrate form (Al₂(SO₄)₃•18H₂O) was chosen for its crucial role in water treatment, leveraging its unique properties. With a molecular weight of approximately 666.429 g/mol, this form readily dissolves in water, promoting uniform distribution and efficient interaction with contaminants, crucial for removing impurities from water samples. Prior to the jar test, a comprehensive analysis, including pH evaluations during daily sampling (without acidifications), was conducted. The pH of the mixture was adjusted to the monthly average value (at the end of the month) by adding NaOH (1 M) on the treatment day, reflecting the actual pH values after acidification operations and preserving the sample integrity. Additionally, monthly average temperature values were calculated for water samples, adjusting each sample to this temperature before treatment with Al₂(SO₄)₃ for realistic conditions.

To enhance operational efficiency and reduce chemical waste, a support vector machine (SVM) mathematical model was employed to predict the optimal coagulant dosage. This model integrates pH, electrical conductivity, temperature, turbidity, and TAC values from combined samples (the mixture) [41,42]. This technique, validated and previously published in our earlier article [41,42], adapts to changing conditions, aligning with goals of profitability and environmental sustainability to minimize operational costs and chemical waste and addressing complexities related to climate change and fluctuating parameters [41,42].

Following the determination of the optimal dosage, the study proceeded to the jar test. Over a period of more than 12 months, jar tests (Floculateur Fisher 1198) were carried out with only three stirrers out of the six available and one-liter beakers containing 500 mL of mixed monthly samples. Controlled agitation at 180 rpm for 5 min [43,46], followed by intentional reduction to 40 rpm over the next 30 min, facilitated optimal interaction and floc development [42]. After a settling period of 45 min, the supernatant was extracted and filtered through specialized paper, enabling a comprehensive comparison between treated and untreated water samples [41–43]. This systematic approach allowed for a thorough evaluation and precise quantification of the effectiveness of Al₂(SO₄)₃.

2.3. PhysicoChemical Analysis

After daily samplings and meticulous adherence to precautions, a monthly physicochemical analysis of combined samples was conducted in two stages: an initial assessment followed by a second analysis after coagulation–flocculation treatment [41,45]. This comprehensive analysis measures 21 key parameters, as previously mentioned. This process is repeated at the end of each month for a year, strictly following established protocols and guidelines [47]. Detailed protocols for each parameter are available in the supplementary data.

2.4. Statistical Analysis

The measurements were conducted three times for each data point, and the results of the experimental data were presented as the average along with the standard deviation (SD). The statistical assessment (performed using ANOVA) was carried out using OriginPro 8 software with a 95% confidence level (where p < 0.05). Tukey's one-way comparison test was applied for post hoc analysis.

Table 1, which compiles the minimum ('min'), median ('mean'), maximum ('max'), and standard deviation ('STD') values of the data, provides a comprehensive statistical analysis. Additionally, these minimum, median, and maximum values for each parameter in polluted water (raw water) were derived from a thorough analysis conducted throughout the year. Its inclusion aims to highlight the elevated level of pollution at the specified site, thereby underscoring its failure to meet global standards for treated water.

Key Parameters	Unit	Min	Mean	Max	STD	
Potential hydrogen	-	7.65	7.79	8.19	0.14	
Electrical conductivity	$\mu \mathrm{S}\mathrm{cm}^{-1}$	2870.00	3315.00	3410.00	207.82	
Turbidity	NTU	3.01	4.94	9.26	2.03	
Organic matter	mg/L	20.50	25.30	29.2	2.55	
Ammonium	mg/L	0.64	0.79	0.88	0.08	
Phosphate	mg/L	0.53	0.75	0.98	0.13	
Nitrate	mg/L	55.00	66.05	96.977	10.81	
Nitrite	mg/L	0.24	0.32	0.38	0.03	
Calcium	mg/L	240.28	254.30	276.35	10.73	
Magnesium	mg/L	168.04	197.20	202.10	10.35	
Total hardness	°F	130.08	145.74	153.29	6.99	
Chloride	mg/L	510.00	554.70	575.10	19.96	
Bicarbonate	mg/L	400.00	448.27	496.42	32.39	
Sulfate	mg/L	540.00	652	900	119.50	
Iron	mg/L	Negligible	Negligible	Negligible	/	
Manganese	mg/L	Negligible	Negligible	Negligible	/	
Aluminum	mg/L	Negligible	Negligible	Negligible	/	
Potassium	mg/L	21.50	23.20	25.20	1.41	
Sodium	mg/L	2150.00	270.00	380.00	55.03	
Complete alkalimetric	mg/L	212 54	415 47	428 75	29.82	
titration	of CaCO ₃	515.54	413.47	420.75	27.02	
Dry residue	mg/L	2100.00	2494.00	2980.00	297.69	

Table 1. Comprehensive statistical analysis of minimum, median, maximum, and standard deviation values for polluted water parameters throughout the year.

3. Results and Discussion

As previously highlighted, the SVM model developed and published in our previous article was employed in this study to predict the coagulant doses required for treating contaminated water [41,42]. This prediction was achieved by incorporating the pH, electrical conductivity, temperature, turbidity, and TAC parameters of the raw water [41,42]. The coagulant doses obtained through this method are documented in Table 2. This predictive approach based on SVM demonstrated significant accuracy, thereby validating the utility of this model in optimizing the coagulation–flocculation process.

Table 2. Coagulant-flocculant doses (mg/L) by month.

January	February	March	April	May	June	July	August	September	October	November	December
427.94	414.64	427.35	427.03	427.62	427.69	426.87	427.19	427.14	427.08	427.25	427.91

Table 2 illustrates the variation in coagulant–flocculant doses used each month in the treatment of contaminated water. The values exhibit slight fluctuations from one month to another, suggesting that the quantity of coagulant–flocculant required may depend on environmental conditions or the characteristics of the raw water throughout the months. These data are crucial for ensuring effective water treatment throughout the year. Indeed, the coagulant–flocculant dose values exhibit minor month-to-month variations due to the similarity in environmental conditions and characteristics of the raw water throughout the year. In other words, the minimal variations in coagulant–flocculant doses reflect the fact that the 21 physicochemical parameters of the raw water remained relatively constant and highly polluted throughout the year (Table 1). This indicates that the raw water characteristics did not change significantly from one month to another, which explains the close proximity of the coagulant–flocculant doses.

To initiate the analysis, histograms for each physicochemical parameter in raw and treated water were plotted for easy evaluation of the effectiveness of $Al_2(SO_4)_3$. It thus contributed to highlighting the effectiveness of this specific process.

3.1. Potential Hydrogen Activity (pH) of Water

Before adding $Al_2(SO_4)_3$ to raw water samples, a meticulous readjustment of their initial pH was carried out using a NaOH solution. This step was necessary as the samples had been initially acidified for preservation purposes. As convincingly highlighted by Figure 2 (raw waters), the pH values ranged from 7.65 to 8.19, demonstrating the strict adherence to the standards set for drinking water by the Official Journal of the Algerian Republic. As perspicuously illustrated by Figure 2, depicting the characteristics of raw waters, the pH values exhibited a varied range, ranging between 7.65 and 8.19. This meticulous approach ensures the reliability of the results and underscores the commitment to practices in compliance with rigorous standards in force.



Figure 2. Variation in monthly means of pH as a function of time (months) of raw and treated waters.

The introduction of $Al_2(SO_4)_3$ resulted in a careful transformation of the raw water samples, leading to a slight decrease in pH. The pH values converged towards slightly acidic ranges, ranging from pH 4.8 to 5.7 (as depicted in Figure 2). This pH adjustment can be attributed to the coagulation process, during which $Al_2(SO_4)_3$ neutralized negative charges within the liquid, generating hydroxide ions (OH⁻) and, consequently, causing a modest decrease in pH. It is essential to note that even though the pH of the water reached as low as pH 4.8 following $Al_2(SO_4)_3$ treatment, it is generally accepted that this variation does not result in substantial issues.

Furthermore, it is worth highlighting that in conventional drinking water treatment facilities pH correction is typically performed at the end of the treatment process, just before distribution. It is also pertinent to mention that there is a documented decrease in the effectiveness of chlorine against microbial contamination beyond a pH value of 8.19 [47–49]. Nevertheless, the pH adjustments resulting from $Al_2(SO_4)_3$ treatment in this study remained well within acceptable ranges, thereby ensuring that water quality met the relevant standards without compromising safety. This detailed analysis enhances our understanding of the pH reaction when using the coagulant $Al_2(SO_4)_3$.

3.2. Electrical Conductivity

Variation in the electrical conductivity of water samples has been considered a critical indicator of mineralization and dissolved ion content. In this section, we have examined

in detail the chemical mechanisms that were responsible for the observed reduction in electrical conductivity following the introduction of $Al_2(SO_4)_3$ as a treatment.

As depicted significantly in Figure 3, the electrical conductivity values exhibited noteworthy variations between the raw and treated water samples. Raw water samples showed a wide range of conductivity, ranging from 2870 to 3410 µS/cm each month (Figure 3). It should be emphasized that these values exceeded the maximum limit set by the World Health Organization (WHO) standards, which was established at 2800 μ S/cm. However, a detailed examination of the data in Figure 3 reveals a distinct tendency of obtained values for treated water to remain below those of raw water, especially concerning the maximum value, which slightly exceeds 1000 μ S/cm. The action of Al₂(SO₄)₃ has thus significantly contributed to the reduction in electrical conductivity levels for treated water. In harmony with the findings of Rodier et al., it is established that a conductivity exceeding $600 \ \mu\text{S/cm}$ indicates substantial mineralization of waters, a feature clearly evident in the case of treated water [47]. Guidelines set by USEPA highlight that electrical conductivity tends to maintain relative stability within a distribution network, provided the balance with pipe materials is upheld. Substantial conductivity variations, as clearly illustrated by the comprehensive comparison between raw water and treated water conductivity levels (Figure 3), could potentially signal issues related to corrosion [41,50].



Figure 3. Variation in monthly mean conductivity $(\mu S/cm)$ over time (months) for raw and treated waters.

It is important to note that an 85.60% reduction in electrical conductivity was achieved through the coagulation process. Indeed, $Al_2(SO_4)_3$ is commonly used in water treatment for the coagulation and flocculation of suspended particles and impurities. Its primary role is to group these particles into larger flocs, making it easier to remove them from water through settling or filtration. This can lead to improved water clarity and a reduction in turbidity. However, concerning water conductivity, the addition of $Al_2(SO_4)_3$ generally does not directly reduce conductivity. Water conductivity is mainly influenced by the presence of dissociated ions such as Na⁺, K⁺, Ca²⁺, Mg²⁺, Cl⁻, SO₄²⁻, HCO₃⁻, and others [51]. $Al_2(SO_4)_3$ itself dissolves into Al^{3+} ions and SO_4^{2-} ions in water, but this does not significantly alter water conductivity. However, $Al_2(SO_4)_3$ can indirectly affect water conductivity through other parameters, such as [47,52–54]:

(1) pH: The water's pH can influence the effectiveness of Al₂(SO₄)₃ in reducing conductivity. Generally, Al₂(SO₄)₃ is more effective as a coagulant at slightly acidic or neutral pH levels, typically in the pH range of 5 to 8. When the water's pH is maintained within this range, Al₂(SO₄)₃ can form aluminum hydroxide (Al(OH)₃) precipitates that adsorb suspended particles and impurities. These precipitates contribute to the

formation of larger and heavier flocs, which are easier to remove from water. However, if the water's pH is too low (acidic, pH \leq 2) or too high (alkaline, pH \geq 12), the formation of Al(OH)₃ precipitates can be hindered, thus reducing coagulation efficiency. Additionally, water conductivity can be influenced by the presence of dissolved ions, including OH⁻ and hydronium ions (H₃O⁺), which depend on the water's pH. A significant pH change can indirectly affect conductivity by altering the concentration of these ions in the water.

- (2) Complexation with Other Physicochemical Parameters: Al₂(SO₄)₃ complexation with other physicochemical parameters can potentially contribute to reducing water conductivity, especially if these parameters are associated with suspended particles. For example, if certain physicochemical parameters (such as dissolved ions) are bound to suspended particles, Al₂(SO₄)₃ can play a role in their coagulation and flocculation, leading to their removal from water. This could result in a reduction in conductivity, as conductive dissolved ions would be removed along with the suspended particles. In general, Al₂(SO₄)₃ is commonly used in water treatment to remove various parameters and impurities. The main parameters that Al₂(SO₄)₃ can easily eliminate include suspended particles, turbidity, organic matter, PO₄³⁻, heavy metals, and certain inorganic impurities. However, these parameters (such as suspended particles, turbidity, organic matter, PO₄³⁻, and heavy metals) can influence water conductivity in different ways. Here is how these parameters interact with conductivity.
- Suspended Particles and Turbidity: Suspended particles, responsible for water turbidity, may contain adsorbed dissolved ions on their surface. When Al₂(SO₄)₃ is used to coagulate and flocculate these particles, it can lead to the aggregation of particles into larger flocs. If these suspended particles contain conductive ions, their removal through coagulation and sedimentation can contribute to a reduction in water conductivity. However, the impact on conductivity will depend on the relative contribution of these particles to the total conductivity of the water.
- Organic Matter: Dissolved organic matter, containing various organic ions, plays a significant role in conductivity. The use of Al₂(SO₄)₃ for organic matter removal can result in a decrease in conductivity by eliminating specific dissolved organic ions. The extent of this removal is contingent upon the characteristics of the organic matter. Below are examples of organic ions:
 - Carboxylate ions: Derived from carboxylic acids featuring the -COOH functional group. Notable examples include the acetate ion (CH₃COO⁻) and formate ion (HCOO⁻).
 - Sulfonate ions: Originating from sulfonic acids, these ions exhibit the -SO₃H functional group. An illustrative example is the methylsulfonate ion (CH₃SO₃⁻).
 - Phenolate ions: Arising from phenols with an aromatic ring structure, a representative example is the phenolate ion $(C_6H_5O^-)$.
 - Amino acids: The fundamental building blocks of proteins, certain amino acids possess ionizable groups. Glycine, for instance, can exist as a zwitterion with both positive and negative charges.
 - Quaternary ammonium ions: Positively charged nitrogen atoms are commonly found in organic compounds like surfactants. The tetramethylammonium ion (N(CH₃)₄⁺) serves as an example.
 - Phosphonate ions: Derived from phosphonic acids containing phosphorus, the methylphosphonate ion (CH₃PO₃²⁻) is an illustrative instance.
 - Sulfate and Sulfonate Esters: Formed through the esterification of sulfuric or sulfonic acids, these ions include the ethyl sulfate ion (C₂H₅SO₄⁻) and methylsulfonate ester (CH₃SO₃⁻).

It is crucial to recognize that the specific organic ions present in water are contingent upon the source of organic matter and the composition of the organic compounds. When $Al_2(SO_4)_3$ is employed for organic matter removal, it engages with these organic ions, leading to the formation of insoluble aluminum complexes and subsequently contributing to a reduction in conductivity. The efficacy of this process is intricately linked to the characteristics of the organic matter present in the water.

- Phosphates: Dissolved PO₄³⁻ does not significantly contribute to conductivity since it has charged ions. Al₂(SO₄)₃ is often used to precipitate PO₄³⁻ by forming insoluble precipitates, which helps reduce the PO₄³⁻ concentration in water. While the reduction in PO₄³⁻ does not have a direct impact on conductivity, it can influence overall water quality.
- Heavy Metals: Dissolved heavy metals such as Fe³⁺ and Mn²⁺ can contribute to water conductivity as charged ions. Al₂(SO₄)₃ can promote the precipitation of these metals as insoluble precipitates, removing them from water. This can reduce the concentration of dissolved metal ions and potentially reduce conductivity.
- (3) Interactions between Water Quality Parameters (Suspended Particles, Turbidity, Organic Matter, PO₄^{3–}, Heavy Metals) and Ions Directly Impacting Conductivity (Na⁺, Ca²⁺, Mg²⁺, HCO₃⁻, Cl⁻): These interactions can be complex and depend on the chemical composition of the water as well as environmental conditions. Here is how these parameters can interact:
- Suspended Particles and Turbidity: Suspended particles often cause water turbidity. Ions such as Ca²⁺ and Mg²⁺ can bind to suspended particles, contributing to the formation of precipitates. Coagulation with coagulants like Al₂(SO₄)₃ can help group these particles into larger flocs, facilitating their removal and reducing turbidity.
- Organic Matter: Dissolved organic matter in water can complex with metal ions like Fe³⁺ and Mn²⁺, forming organometallic complexes. These complexes can contribute to water color and other quality issues. Al₂(SO₄)₃ can promote the precipitation of these complexes, aiding in the removal of organic matter.
- Phosphates: PO₄³⁻ can react with Ca²⁺ and Mg²⁺ ions to form insoluble precipitates such as Ca²⁺, Mg²⁺ and PO₄³⁻. However, under certain conditions, they can also react with heavy metals to form soluble complexes. Water treatment may aim to reduce PO₄³⁻ concentration by promoting their precipitation.
- Heavy Metals: Heavy metals like Fe³⁺ and Mn²⁺ can precipitate in the presence of Ca²⁺ and Mg²⁺ ions, forming insoluble precipitates. Al₂(SO₄)₃ is commonly used to promote this precipitation and remove these metals from water.
- Na⁺, HCO₃⁻, Cl⁻ Ions: In general, Na⁺, HCO₃⁻, and Cl⁻ ions do not directly react with the aforementioned water quality parameters. However, they can influence the overall chemical balance of water, pH, and other properties, which can, in turn, affect the solubility of compounds and chemical reactions in water.

3.3. Turbidity

Turbidity curves have been meticulously crafted for both the raw and treated water samples, offering a detailed and comprehensive representation in Figure 4. A thorough analysis of Figure 4 has revealed a notable and extensive variation in turbidity values within the raw water, ranging from 1.59 to 9.26 NTU. It is worth noting that these measurements have significantly exceeded the turbidity standards set by the WHO, which has established a maximum limit of 5 NTU. Conversely, when we shift our focus to the treated water samples, as illustrated in Figure 4, it becomes evident that the recorded turbidity values have consistently remained well below 2 NTU. These results have not only complied with but also significantly surpassed the defined tolerance threshold of <5 NTU, as documented in references [41,55]. The turbidity reduction rate has reached an impressive level, achieving a remarkable reduction of 98.05%. This accomplishment is a clear testament to the exceptional efficiency of the treatment process, demonstrating its ability to eliminate the vast majority of suspended particles responsible for the initial turbidity in raw water. This substantial reduction unequivocally reaffirms the effectiveness of employing Al₂(SO₄)₃ in the clarification process, underlining its pivotal role in enhancing water quality.



Figure 4. Variation in mean turbidity (NTU) over time (months) for raw and treated waters.

The process of reducing turbidity through $Al_2(SO_4)_3$ centers on promoting the formation of flocs, which act as agents for capturing and removing suspended particles. This mechanism involves two fundamental steps [56]: coagulation and flocculation, which work synergistically to purify water by aggregating impurities into larger, heavier structures, facilitating their removal.

Coagulation is the initial stage of this process, where $Al_2(SO_4)_3$ is introduced into the water. Upon contact with the water, $Al_2(SO_4)_3$ dissociates into Al^{3+} ions and SO_4^{2-} ions [57]. These Al^{3+} ions play a crucial role in destabilizing the suspended particles present in the water. They neutralize the electrostatic charges on the surface of these particles, which typically repel each other due to their like charges. By neutralizing these charges, the Al^{3+} ions allow for the suspended particles to come closer together, overcoming their natural electrostatic repulsion [57].

Flocculation follows coagulation and is the stage where the formation of flocs occurs. During this step, gentle mixing or stirring is applied to the water, which encourages the previously destabilized particles to collide and stick together [57]. This collision and adhesion process leads to the formation of larger, heavier aggregates, known as flocs. The flocs continue to grow as they capture more suspended particles and impurities in the water [57].

The key to the effectiveness of this process lies in the size and weight of the resulting flocs. Because they are larger and denser than individual particles, these flocs settle more rapidly in the water, aided by gravity. This settling process is known as sedimentation. As the flocs settle to the bottom of the water container or treatment tank, they entrap and encapsulate suspended particles and impurities within their structure [57].

Once the flocs have settled, they can be easily separated from the clear water through physical methods such as sedimentation basins or filtration systems [57]. The clear water is then collected for distribution, while the captured impurities remain trapped in the settled flocs [58].

It is essential to emphasize that turbidity, as observed, originates from suspended particles in water, including various components such as organic debris like clay and microscopic organisms [41,59]. Additionally, it is noteworthy that turbidity values often experience a significant increase during and after periods of precipitation. Furthermore, high turbidity can influence water temperature, leading to an elevation, while concurrently affecting the concentration of dissolved oxygen, which tends to decrease [41,56]. The significance of these parameters should not be underestimated, as their substantial influence

can have far-reaching implications, impacting not only overall water quality but also the equilibrium and stability of the surrounding ecosystem [41,42,60].

3.4. Pollution Parameters

3.4.1. Organic Matter

The graphical representations depicting organic matter concentrations in both untreated and treated water samples have been meticulously crafted, unveiling a wealth of intricate details within Figure 5. Expanding upon the insights gleaned from the analysis conducted in Figure 5, it becomes increasingly apparent that organic matter values in the untreated water samples exhibit significantly elevated levels, with a striking range spanning from 20.5 to 29.2 mg/L. This range of measurements naturally beckons further investigation into whether these values align with global regulatory standards. Nevertheless, the most pivotal observation emerges from the treated water samples (Figure 5), where the rigorous implementation of the $Al_2(SO_4)_3$ coagulation–flocculation process [41,61] culminates in a remarkable reduction in organic matter levels. The results unequivocally demonstrate that the coagulation–flocculation process with $Al_2(SO_4)_3$ has achieved an impressive reduction in organic matter, reaching an outstanding rate of 99.62%. This figure undeniably confirms the unquestionable effectiveness of this method in diminishing organic matter levels in treated water.



Figure 5. Variation in monthly mean organic matter (mg/L) over time (months) for raw and treated waters.

The specific types and concentrations of dissolved organic carbon (DOC) in river or stream water can vary widely depending on the characteristics of the water source, surrounding land use, geological features, and human activities in the area. Dissolved organic carbon refers to the fraction of organic carbon in water that is in a dissolved form. It includes a variety of organic compounds that have leached into the water from the surrounding environment.

The types of dissolved organic carbon in river or stream water can include:

- Humic Substances: Humic substances are complex organic compounds formed during the decay of plant and animal matter. They include fulvic acids, humic acids, and hymatomelanic acids. These substances contribute to the brown color often seen in water bodies.
- Amino Acids and Proteins: Organic nitrogen compounds, including amino acids and proteins, can be present in dissolved form in water. These compounds may come from decaying organic matter, animal waste, or anthropogenic sources.

- Carbohydrates: Sugars and other carbohydrates can be dissolved in water, contributing to the overall dissolved organic carbon content. These compounds may come from plant material or runoff from agricultural areas.
- Organic Acids: Various organic acids, such as fulvic acid and acetic acid, can be found in water. These acids contribute to the acidity of the water and can play a role in complexation reactions with metals.
- Dissolved Organic Nitrogen (DON): In addition to dissolved organic carbon, nitrogencontaining organic compounds may also be present. These include amines, amides, and other nitrogen-containing organic molecules.
- Microbial Metabolites: The activities of microorganisms in the water can lead to the production of organic compounds as metabolic byproducts. This can include substances like exopolysaccharides and extracellular enzymes.

The specific composition and concentrations of these dissolved organic carbon components will vary based on the specific conditions of the river or stream, including factors such as land use, vegetation, climate, and anthropogenic activities in the watershed.

 $Al_2(SO_4)_3$ plays a pivotal role in water treatment by establishing a foundation for effectively reducing the presence of organic matter. When introduced into water, $Al_2(SO_4)_3$ undergoes a chemical reaction, resulting in the formation of $Al(OH)_3$ precipitates, often referred to as "flocs". These flocs carry a positive charge, and their presence in the water augments the overall capacitance and enhances electrostatic interactions within the aquatic environment [15,47,62]. The adsorption process involving these positively charged flocs plays a crucial role in significantly reducing the concentration of dissolved organic matter in the water. In fact, this reduction can be quite remarkable, with reduction rates reaching as high as 99.62%. This substantial decrease in dissolved organic matter is of paramount importance in enhancing water quality. The removal of unwanted organics from the water has far-reaching implications. First and foremost, it contributes to the safety of the water supply. High levels of dissolved organic matter can introduce contaminants and impurities, potentially jeopardizing the health of those who consume or use the water. Additionally, it aids in ensuring that the water complies with stringent water quality standards, which are set to safeguard public health and the environment. By employing $Al_2(SO_4)_3$ to generate flocs that efficiently adsorb and eliminate dissolved organic matter, water treatment processes achieve a notable improvement in water quality. This, in turn, fosters a safer and more reliable water supply, meeting the expectations and requirements of both regulatory bodies and the communities they serve. The reduction in dissolved organic matter through the action of $Al_2(SO_4)_3$ thus underscores its vital role in the broader context of water treatment and the provision of clean, safe, and compliant drinking water.

In concordance with the discoveries made by Harrat and Achour, it is pertinent to underscore the potential formation of chlorination byproducts, including chloroform, resulting from the interaction between organic matter in surface waters and chlorine. These byproducts carry potential long-term toxicity risks, necessitating careful consideration.

This cautionary note underscores the paramount importance of meticulous deliberation in the decision-making processes related to water management [41,60,63]. It serves as a poignant reminder of the pivotal role played by $Al_2(SO_4)_3$ in the reduction in organic matter, ultimately contributing to the improvement of water quality and adherence to international regulatory standards.

3.4.2. Ammonium

The curves depicting the concentrations of NH_4^+ in both the raw water and treated water samples have been meticulously constructed, showcasing an exceptional level of precision, as exemplified in Figure 6. An analysis of these graphs reveals a striking conclusion: the levels of NH_4^+ in raw water exceed established drinking water standards (greater than 0.5 mg/L) (Figure 6), while treated water samples convincingly meet potability criteria (Figure 6), demonstrating a reduction rate of 100% (especially as the initial values were

VH4+ (mg/L)



very low). This marked disparity underscores the undeniable efficiency of the treatment process using $Al_2(SO_4)_3$.

Figure 6. Variation in monthly averages of ammonium (mg/L) over time (months) for raw and treated waters.

WHO standards

Treated water

Raw water

Indeed, the reduction in NH_4^+ in water can be indirectly influenced by factors such as conductivity, organic matter, turbidity, and pH. Understanding these interactions is crucial for optimizing water treatment processes and ensuring water quality in various contexts, from drinking water supply to the preservation of aquatic ecosystems. Here is how these parameters can interact [34,47,64,65]:

- Conductivity: Water conductivity is essentially a measure of its ability to conduct electrical current and is primarily influenced by the presence of dissolved ions. NH₄⁺ is one of these ions, and its concentration in water contributes to the total conductivity. Thus, water containing a significant amount of NH₄⁺ typically has higher conductivity. By reducing water conductivity (as found), you can potentially reduce the concentration of NH₄⁺ (especially when the initial values are very low), although this also depends on the contribution of other ions to conductivity.
- Organic Matter: Dissolved organic matter in water can be a key factor in reducing NH4⁺. Some organic compounds can complex with NH4⁺, forming complexes that may be less soluble. This can potentially promote the precipitation or adsorption of NH4⁺, contributing to its reduction.
- Turbidity: While turbidity itself is not directly related to NH₄⁺, it can have an indirect effect. Suspended particles in water can adsorb or trap NH₄⁺, altering its availability in the water. Reducing turbidity can, therefore, help maintain a more stable NH₄⁺ concentration in the water.
- pH: The pH of water is a crucial parameter for understanding the chemistry of NH₄⁺. At higher pH values (such as those mentioned between 7.65 and 8.19), a greater proportion of NH₄⁺ can convert to ammonia (NH₃), which is a gaseous and less ionic form. This conversion can be desirable in some situations to reduce the toxicity of NH₄⁺.

The presence of NH_3 nitrogen in surface waters can be attributed to a complex array of sources. These sources encompass the existence of plant materials along watercourses, the decomposition of organic matter of animal or human origin (humans excrete between 15 and 30 g of urea daily), industrial discharges, and fertilizer inputs, among others [41,47]. According to the comprehensive findings of Rodier et al., the application of chlorinebased oxidation treatment proves effective in removing dissolved NH_3 nitrogen from water. In this regard, it should be noted that biological oxidation of NH_3 could lead to the formation of anaerobic zones within certain parts of distribution networks, thereby generating unpleasant tastes and potentially contributing to pipe corrosion [47]. These findings emphasize the critical importance of maintaining adequate levels of NH_4^+ in water resources intended for human consumption, not only to ensure public health but also to preserve the integrity and long-term sustainability of the hydraulic infrastructure.

3.4.3. Phosphate

The curves representing the concentrations of PO_4^{3-} in both the raw water and treated water samples are displayed in Figure 7. A closer examination of Figure 7 reveals that the recorded levels exceed the limit of 0.5 mg/L in raw water sources. This observation emphasizes the natural presence of phosphorus in water sources, a characteristic influenced by the properties of the terrain and the decomposition of organic matter. It is worth noting that concentrations above 0.5 mg/L can be interpreted as potential indicators of pollution [41,47]. However, a striking contrast is observed in treated water samples, where complete removal of PO_4^{3-} is evident (Figure 7), demonstrating a reduction rate of 100% (Especially the initial values were very low). This finding unequivocally highlights the efficiency of the treatment process using $Al_2(SO_4)_3$. The residual presence of PO_4^{3-} in treated water is practically nonexistent, showcasing the exceptional performance of this chemical compound in contaminant removal.



Figure 7. Variation in monthly averages of phosphate (mg/L) over time (months) for raw and treated waters.

Indeed, the chemical reaction between $Al_2(SO_4)_3$ and PO_4^{3-} ions plays a pivotal role in water management and purification. When these two compounds come into contact, they react to form insoluble aluminum phosphate precipitates, creating a solid complex [66,67]. This precipitation process is of paramount importance as it effectively removes unwanted PO_4^{3-} ions from water. Once formed, these precipitates can be removed through separation methods such as sedimentation or filtration, significantly contributing to the reduction in PO_4^{3-} concentrations in water [68]. This chemical reaction thus plays an essential role in preserving water quality and protecting the environment by limiting algal proliferation and preventing eutrophication of aquatic ecosystems [69].

These remarkable results convincingly confirm the significant role of $Al_2(SO_4)_3$, which has proven to be a crucial factor in achieving a substantial reduction in PO_4^{3-} levels in treated

water. This conclusion underscores the positive impact of $Al_2(SO_4)_3$ usage on water quality enhancement and the preservation and balance of the surrounding aquatic ecosystem.

3.4.4. Nitrate

Research conducted by Rodier et al. demonstrates that NO_3^- results from natural decomposition processes, primarily orchestrated by nitrogenous microorganisms present in plant and animal proteins and originating from excrement [45]. It is also noteworthy that precipitation can introduce NO_3^- from nitrogen oxides and NH₃ present in the atmosphere. Furthermore, bacterial oxidation of NH₃ can play a significant role in elevating NO_3^- levels [41,70].

It is essential to consider the health implications of high NO_3^- concentrations in water. These adverse effects, such as mucous membrane irritation, nausea, vomiting, bloody stools, and even hypertension risks [41,71], underscore the crucial importance of monitoring and reducing NO_3^- levels in water resources intended for human consumption. This approach aims to ensure the health and well-being of communities dependent on these water sources.

Carefully plotted curves that visualize NO_3^- concentrations within the raw water and treated water samples are detailed in Figure 8. A thorough analysis of NO_3^- concentration variations reveals significant differences, with values ranging from 96.977 mg/L to 55 mg/L in raw water (Figure 8). These measurements manifestly exceed the current regulatory standards for waters, thereby highlighting a contamination issue to be considered. In contrast, the recorded values for treated water demonstrate a notable reduction and fall below those of raw water (Figure 8).



Figure 8. Variation in monthly averages of nitrate (mg/L) over time (months) for raw and treated waters.

It is imperative to emphasize that the WHO recommends a limit of 50 mg/L (NO₃⁻) for waters intended for human consumption [47]. After introducing Al₂(SO₄)₃, a reduction rate of 100% was recorded. This finding indisputably highlights the intrinsic efficiency of the treatment process using Al₂(SO₄)₃ in the drastic reduction in NO₃⁻ levels, thus contributing to compliance with water quality standards.

The chemical reaction between $Al_2(SO_4)_3$ and NO_3^- ions does not naturally lead to the formation of insoluble precipitates [72]. However, it is conceivable that this reduction involves the use of specific pH conditions, particular reagent concentrations, or other reaction parameters that promote NO_3^- reduction or precipitation [72]. These processes can be tailored to specific water treatment or effluent treatment needs. In our study, the significant reduction in NO_3^- within a pH range of 7.65 to 8.19 suggests that these pH conditions were favorable for NO_3^- reduction when using $Al_2(SO_4)_3$ as a treatment agent. Several mechanisms may explain this phenomenon [47,73–76]:

- 1. Precipitate Formation: In this pH range, $Al_2(SO_4)_3$ can react with NO_3^- to form aluminum nitrate precipitates or other insoluble reaction products. These precipitates can then be removed through sedimentation or filtration, resulting in NO_3^- reduction in the water.
- 2. Adsorption: Al₂(SO₄)₃ can also adsorb NO⁻₃ ions onto the surface of its particles or formed precipitates, thereby removing them from the aqueous solution.
- 3. Direct Chemical Reaction: Under specific pH conditions, it is possible that Al₂(SO₄)₃ directly reacts with NO⁻₃ to convert it into a non-reactive form or precipitate it:
- Effects on Physicochemical Parameters: Furthermore, the efficient removal of physicochemical parameters using Al₂(SO₄)₃ can have a positive impact on NO₂⁻ and NO₃⁻ concentrations in the water. Here are some possible reasons.
- Reduction in Organic Load: The removal of organic matter by Al₂(SO₄)₃ can reduce the availability of organic compounds that promote the growth of nitrifying bacteria, contributing to the reduction in NO₂⁻ and NO₃⁻.
- Reduction in Turbidity and Suspended Matter: The elimination of turbidity and suspended matter can contribute to water clarification, thereby improving the efficiency of biological processes responsible for converting NO₂⁻ into NO₃⁻ and subsequently reducing them.
- Impact on pH and Reaction Conditions: The use of Al₂(SO₄)₃ can influence the pH of the water, creating specific reaction conditions that favor the reduction in NO⁻₂ and NO⁻₃.
- 4. Removal of other Ions:
- The precipitates formed by the reaction between Al₂(SO₄)₃ and certain ions, such as PO₄³⁻, can trap other ions in solution. This precipitation can potentially sequester NO₂⁻ and NO₃⁻, removing them from the aqueous phase and contributing to their reduction.
- Reduction in Chemical Oxygen Demand (COD): The removal of organic matter and other impurities by Al₂(SO₄)₃ can reduce the chemical oxygen demand (COD) of the water. This can improve water quality in terms of chemical reactivity and promote conditions that reduce the presence of NO₂⁻ and NO₃⁻.
- 5. Not to forget, there chemical reactions involving NO₂⁻ and NO₃⁻ ions in relation to other physicochemical parameters such as pH, electrical conductivity, turbidity, organic matter, and metal ions.
- Reactions between NO₃⁻ and NO₂⁻: NO₃⁻ and NO₂⁻ ions are forms of oxidized nitrogen in water. They can be involved in conversion reactions, especially in natural environments and water treatment systems. Under acidic conditions (low pH) and in the presence of reducing microorganisms, NO₃⁻ can be reduced to NO₂⁻ in a process called denitrification. This reaction involves electron transfer:

$$NO_3^- + 2e^- + 2H^+ \to NO_2^- + H_2O$$
 (1)

Conversely, under more alkaline conditions (high pH), NO_2^- can be oxidized to NO_3^- . This reaction can be catalyzed by nitrifying bacteria:

$$2NO_2^- + O_2 \rightarrow 2NO_3^- \tag{2}$$

These reactions are essential for the nitrogen cycle in aquatic ecosystems.

 Reactions of NO₂⁻ Decomposition: NO₂⁻ ions can undergo decomposition under acidic conditions:

$$2NO_2^- + 8H^+ + 6e^- \to N_2 + 4H_2O$$
(3)

This reaction produces nitrogen gas and water.

- Oxidation and Reduction Reactions with Other Ions: Oxidation and reduction reactions involving NO₃⁻ and NO₂⁻ ions can be influenced by the presence of other metal ions. For example, Fe³⁺ can act as an oxidant for NO₂⁻ ions, forming ferrous Fe³⁺. This can be important in denitrification processes.
- Influence of pH and HCO₃⁻ Ions: The pH of the solution is a crucial factor in reactions involving NO₃⁻ and NO₂⁻ ions. Reduction reactions are generally favored in acidic conditions, while oxidation reactions are favored in alkaline conditions. The presence of HCO₃⁻ ions can also influence these reactions as they can act as buffers and modify the solution's pH.

3.4.5. Nitrite

Meticulously plotted curves representing NO₂⁻ concentrations within the raw water and treated water samples are detailed in Figure 9. A comprehensive examination of the data reveals a reduced presence of NO₂⁻, as clearly highlighted in Figure 9. Consistent with Kurnaz's observations (2004), NO₂⁻ is typically found at modest levels in natural waters. However, it is noteworthy that even at these levels, the measured concentrations exceed the WHO regulatory standards (maximum value of 0.1 mg/L). Nevertheless, a striking contrast emerges for treated water samples, with a significant observation: NO₂⁻ values have been reduced to zero (100% reduction rate, especially as the initial values were very low) (Figure 9), thus underscoring the complete elimination of this harmful substance. This undeniable discovery highlights the intrinsic efficiency of the treatment process using Al₂(SO₄)₃, ensuring a drastic reduction in NO₂⁻ levels and thereby ensuring compliance with rigorous water quality standards.



Figure 9. Variation in monthly averages of nitrite (mg/L) over time (months) for raw and treated waters.

The origins of NO_2^- in raw waters are diverse and can be attributed to several factors, including [41]:

- Partial oxidation of NH₃ nitrogen, leading to NO₂⁻ formation under the influence of nitrifying bacteria [41].
- Reduction of NO₃⁻, primarily of agricultural origin, into NO₂⁻ through the action of denitrifying bacteria [77].

Research by Rodier et al. (2009) also mentions the possibility of NO_2^- formation as a byproduct during the water chlorination process. This precaution is crucial to maintaining adequate chlorination while ensuring an optimal level of safety for consumers [47].

Regarding water intended for human consumption, it is important to note that the WHO recommends a provisional guideline value of $3 \text{ mg/L} (\text{NO}_2^-)$ [47], thus highlighting the fundamental importance of monitoring and maintaining NO₂⁻ levels in accordance with health guidelines to ensure the purity and safety of drinking water for the health and well-being of the population.

Our study has illuminated several relevant aspects related to the removal of NO_2^- from water using $Al_2(SO_4)_3$ as a treatment agent. The results reveal a complex yet effective mechanism that relies on several key elements: [15,47,74,78].

- Decomposition of NO₂⁻: NO₂⁻ present in water can undergo decomposition under acidic conditions, typically at a low pH. This chemical reaction leads to the production of gaseous nitrogen (N₂) and water (H₂O). This NO₂⁻ decomposition is an essential element of the elimination process and significantly contributes to its reduction in water.
- Oxidation and reduction reactions with other ions: Oxidation and reduction reactions involving NO₃⁻ and NO₂⁻ ions can be modulated by the presence of other metal ions, notably Fe³⁺. These reactions can play a crucial role in water treatment systems, promoting the conversion of NO₂⁻ into NO₃⁻ or their complete elimination.
- Influence of pH and HCO₃⁻ ions: The pH of the solution plays a crucial role in reactions between NO₂⁻ and other ions. Acidic conditions generally favor reduction reactions, while alkaline conditions favor oxidation reactions. Additionally, the presence of HCO₃⁻ ions can act as a buffer, altering the solution's pH and, consequently, influencing the progression of reactions.

3.5. Overall Mineral

3.5.1. Calcium, Magnesium, and Total Hardness

The meticulously crafted concentration curves depicting the levels of Ca^{2+} , Mg^{2+} , and TH in both untreated and treated water samples are intricately detailed in Figures 10–12. A comprehensive examination of Figure 10 reveals a notable variation in Ca^{2+} levels in untreated water, spanning from 240.3 to 276.35 mg/L. Similarly, a meticulous analysis of monthly average Mg^{2+} concentrations, as presented in Figure 11, unveils a range of fluctuation from 168.04 to 202.1 mg/L in untreated water. It is of utmost importance to underscore that both of these parameters surpass the prescribed WHO standards for water quality.

In consonance with insights offered by Rodier et al., the productivity of freshwater ecosystems has been intricately correlated with the presence of Ca^{2+} and Mg^{2+} salts, reaching its zenith with a total Ca^{2+} alkalinity of 25 mg/L [47]. This association underscores the pivotal role of these elements in the chemical composition and inherent quality of natural water bodies.

Water hardness, arising from the collective concentrations of Ca^{2+} and Mg^{2+} , assumes a central position in the assessment of water potability. The remarkably elevated levels documented for these two elements, as depicted in Figure 12, unequivocally classify the water as "very hard".

It is important to emphasize that water hardness not only affects human health but also significantly influences pipe corrosion. In line with Rodier's research, hard water, characterized by high concentrations of Ca^{2+} and Mg^{2+} , generally exhibits lower corrosivity compared to soft water. This discrepancy can be attributed to the formation of a protective layer of carbonates that precipitate onto the inner surfaces of pipes, effectively mitigating the risk of corrosion [41,79,80]. These intricate interactions between water parameters and their ramifications on the quality and stability of water distribution infrastructure accentuate the critical necessity for judicious and balanced management of these essential elements to ensure continuous access to high-quality potable water.



Figure 10. Variation in monthly averages of calcium (mg/L) over time (months) for raw and treated waters.



Figure 11. Variation in monthly averages of magnesium (mg/L) over time (months) for raw and treated waters.

Following the introduction of $Al_2(SO_4)_3$ into the contaminated water, concentrations of Ca^{2+} and Mg^{2+} in treated water samples underwent a reduction attributable to $Al_2(SO_4)_3$ treatment, albeit remaining slightly elevated (Figures 10 and 11). Nonetheless, they remain within the limits recommended by the WHO. Our findings have unequivocally demonstrated that the prudent application of $Al_2(SO_4)_3$ resulted in substantial reductions, ranging from 55.35% to 76.66% for Ca^{2+} ions and from 76.58% to 91.37% for Mg^{2+} ions. The reduction in total water hardness ranged from 67.40% to 78.69%. These results underscore the effectiveness of $Al_2(SO_4)_3$ as a coagulant for mitigating water hardness under specific conditions, thereby facilitating the precipitation of Ca^{2+} and Mg^{2+} ions as insoluble salts.

As indicated in the conductivity reduction mechanism, Ca^{2+} and Mg^{2+} ions are among the key parameters that influence conductivity. Therefore, by reducing conductivity (as found), Ca^{2+} and Mg^{2+} ions will be automatically reduced by one of the mechanisms mentioned in the conductivity reduction process, notably pH [81].



Figure 12. Variation in monthly averages of total hardness (mg/L) over time (months) for raw and treated waters.

Indeed, when $Al_2(SO_4)_3$ is introduced into water, it reacts with the Ca^{2+} and Mg^{2+} ions present in the solution. The mechanism of this reaction is based on the formation of insoluble precipitates, primarily calcium hydroxide (Ca(OH)₂) and magnesium hydroxide (Mg(OH)₂). This process is influenced by the pH of the water, with the pH range of 7.65 to 8.19 being particularly favorable. In this pH range, OH⁻ are relatively abundant, thereby facilitating the formation of precipitates. The chemical equation summarizing this mechanism is as follows:

Formation of hydroxide precipitates:

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+$$

$$\tag{4}$$

Precipitation reactions for Ca²⁺ ions and Mg²⁺ ions, respectively:

$$Ca^{2+} + 2OH^{-} \rightarrow Ca(OH)_2 \text{ (precipitate)}$$
 (5)

$$Mg^{2+} + 2OH^- \rightarrow Mg(OH)_2$$
 (precipitate) (6)

Subsequently, these insoluble precipitates are removed from the system through sedimentation or filtration processes, resulting in a substantial reduction in the concentrations of Ca^{2+} and Mg^{2+} ions in the water and, consequently, a reduction in water hardness.

The initial pH of the water between 7.65 and 8.19 promotes the formation of these precipitates, thereby optimizing the effectiveness of $Al_2(SO_4)_3$ as a water hardness reduction agent.

3.5.2. Chloride

The meticulously plotted curves representing Cl^- concentrations within the raw water and treated water samples are detailed in Figure 13. Careful observation of these curves reveals a significant variation in Cl^- levels in raw water, ranging between 510 mg/L and 562.52 mg/L (Figure 13). This concentration range notably exceeds the standards established by the WHO water quality guidelines. However, the treatment of water samples using the $Al_2(SO_4)_3$ -based process has yielded remarkable results by reducing $Cl^$ concentrations to considerably lower levels, with most falling below 90 mg/L (Figure 13). It is worth noting that a slight exception was observed in November, when a value of 124 mg/L was recorded. Nevertheless, even this value, remaining below the limits set by



the rigorous Official Journal of the Algerian Republic standards (2011) [41,55], demonstrates the unequivocal effectiveness of the treatment process.

Figure 13. Variation in monthly averages of chloride (mg/L) over time (months) for raw and treated waters.

In addition to its impact on water quality compliance, Cl⁻ has broader implications. Its effect on the taste of water is an important aspect to consider, especially when concentrations reach or exceed 250 mg/L, which can alter the perceived quality of drinking water. Interestingly, the presence of Ca²⁺ and Mg²⁺ can mitigate this unpleasant taste. However, beyond the taste aspect, Cl⁻ can also play a role in the corrosion of water distribution infrastructure, particularly in the case of stainless steel materials. The concentration of 50 mg/L marks a critical threshold beyond which the risk of corrosion increases significantly [47]. These factors underscore the need for constant monitoring of Cl⁻ levels to ensure high-quality drinking water and the long-term sustainability of distribution networks. In conclusion, a comprehensive analysis of Cl⁻ concentrations in the raw water and treated water samples, as detailed in Figure 13, highlights the exceptional efficiency of the $Al_2(SO_4)_3$ -based treatment in reducing Cl^- levels. This approach ensures compliance with rigorous water quality standards while preserving public health and the integrity of water distribution infrastructure. The water treatment process involving the use of $Al_2(SO_4)_3$ in coagulation-flocculation provides an effective method for reducing the concentration of Cl⁻ ions in water. This treatment relies on complex chemical reactions that occur in a specifically controlled environment. Al₂(SO₄)₃ is introduced into the water to be treated, generating Al^{3+} ions and SO_4^{2-} ions. Maintaining the pH of the water within an optimal range, typically between 7.65 and 8.19 in this case, is crucial to promote the coagulation process. Subsequently, the formation of Al(OH)₃ occurs under the influence of the appropriate pH, where Al³⁺ ions react with water molecules to create Al(OH)₃ according to the equation [15]:

$$2Al^{3+} + 6H_2O \rightarrow 2Al(OH)_3 + 6H^+$$
 (7)

Following this, Cl^- ions present in the water interact with Al(OH)₃, leading to the formation of insoluble alumina (Al₂O₃). The reaction is described as follows:

$$2Al(OH)_3 + 6Cl^- + 3/2O_2 \rightarrow Al_2O_3 + 6OH^- + 3Cl_2$$
(8)

In this reaction, chlorine undergoes oxidation by losing electrons, and it is oxidized from a lower oxidation state (in chloride ions, Cl⁻) to a higher oxidation state in chlorine

gas (Cl₂). The oxidation of chlorine is accompanied by the reduction of aluminum ions to form insoluble alumina (Al₂O₃) and hydroxide ions (OH⁻) as byproducts.

This reaction results in the formation of insoluble alumina, chlorine gas (Cl_2), and OH^- as byproducts.

The formed alumina appears in the form of solid, insoluble flocs. These flocs have the ability to capture various impurities present in the water, such as suspended particles, organic matter, and excess ions. Due to their greater weight compared to water, the precipitated alumina flocs settle at the bottom of the treatment tank. The precipitated alumina flocs are then removed from the treatment tank, resulting in a substantial reduction in the concentration of Cl^- ions, as well as other impurities, in the treated water [47].

This mechanism explains how $Al_2(SO_4)_3$, under appropriate pH conditions and with an adequate dosage, can effectively reduce the concentration of Cl⁻ ions in water by promoting the formation of insoluble alumina. It is worth noting that the efficiency of this process depends on various factors and must be tailored to the specific water treatment conditions [82]. The reduction rate of 95.13% found in this study demonstrates the success of this process in this particular context.

3.5.3. Bicarbonate

The meticulously plotted curves representing HCO_3^- concentrations within the raw water and treated water samples are detailed in Figure 14. Upon careful scrutiny of Figure 14, it is evident that HCO_3^- levels in raw water exceed the defined limits for potable water. In contrast, treated water samples adhere to stringent potability standards, unmistakably illustrating the efficiency of the treatment process using $Al_2(SO_4)_3$ (Figure 14).



Figure 14. Variation in monthly averages of bicarbonate (mg/L) over time (months) for raw and treated waters.

Rodier et al. astutely highlighted that the HCO_3^- concentration in water is closely correlated with pH and temperature, two key parameters that exert significant influence on the chemical composition of water [47]. Natural levels of total alkalinity, which can reach up to 400 mg/L of CaCO₃, although considered relatively moderate, should not be overlooked due to their potential to induce corrosion issues in plumbing systems, underscoring the necessity for cautious and balanced management of these concentrations [41,83].

However, the relevance of maintaining adequate HCO_3^- levels extends beyond mere normative compliance. In reality, these concentrations play a crucial role in preserving water quality as well as the long-term integrity of water distribution infrastructure [41]. The innovative treatment process using $Al_2(SO_4)_3$ that is examined in this study not only ensures the sanitary safety of water but also helps mitigate potential corrosion-related risks, thus preserving the durability and integrity of distribution networks. This holistic approach underscores the importance of comprehensive chemical parameter management to ensure a supply of high-quality water while safeguarding public health and the sustainability of water infrastructure [84].

 $\rm HCO_3^-$ ions are present in water and contribute to its alkalinity. To reduce the $\rm HCO_3^-$ concentration in water, it is generally necessary to use specific treatment techniques, such as the addition of acids (such as sulfuric acid) to neutralize the $\rm HCO_3^-$ and reduce alkalinity [85]. These acids react with $\rm HCO_3^-$ to produce water, carbon dioxide (CO₂), and the corresponding salts:

$$HCO_{3}^{-} + H^{+} \rightarrow H_{2}O + CO_{2} + \text{salts}$$

Example: $2HCO_{3}^{-} + H_{2}SO_{4} \rightarrow 2H_{2}O + 2CO_{2} + SO_{4}^{2-}$ (9)

This reaction decreases the HCO₃⁻ concentration and lowers the alkalinity of water. As previously observed for the pH parameter, a decrease in the water's pH was caused by the introduction of Al₂(SO₄)₃ into the water, which can explain the reduction in HCO₃⁻ in our case [85]. Al₂(SO₄)₃ can chemically react with the HCO₃⁻ in the water, forming precipitates that are removed from the system during the coordilation–flocculation process

precipitates that are removed from the system during the coagulation–flocculation process. The reduction in the pH from 8.19 to 4.5 is likely due to this chemical reaction between $Al_2(SO_4)_3$ and HCO_3^- , which can result in the precipitation of HCO_3^- as solid. This process can be effective in removing some of the HCO_3^- from the water.

Furthermore, a significant decrease in pH from 8.19 to 4.5 indicates acidification of the water, which can effectively lead to a decrease in the HCO_3^- concentration in the water [82]. When the pH is significantly lowered, HCO_3^- can transform into gaseous carbon dioxide (CO₂) and water (H₂O). This can also explain the reduction in HCO_3^- concentration that was observed.

All of these explanations demonstrate a reduction rate of 60%, which was achieved and highlights the effectiveness of $Al_2(SO_4)_3$ in this process.

3.5.4. Sulfate

Meticulously plotted curves representing SO_4^{2-} concentrations within the raw water and treated water samples were meticulously crafted and are presented in detail in Figure 15. A thorough analysis of this figure brings to light a significant characteristic: SO_4^{2-} levels in raw water markedly exceed established thresholds, reaching values even beyond 540 mg/L (Figure 15). This observation clearly underscores that SO_4^{2-} concentrations in raw water do not adhere to the standards prescribed by the WHO water quality norms (Figure 15). However, the efficacy of the treatment process, employing $Al_2(SO_4)_3$, has succeeded in reducing these SO_4^{2-} concentrations in treated water, though a few values remain slightly above the limit. Importantly, these values still fall within the strict limits recommended by the Official Journal of the Algerian Republic (2011) (<400 mg/L), irrefutably affirming the effectiveness of the treatment process.

Research by Rodier et al. has already highlighted that the use of $Al_2(SO_4)_3$ in water coagulation processes can lead to an increase in SO_4^{2-} levels [47]. However, the addition of $Al_2(SO_4)_3$ to water can sometimes lead to a reduction in the SO_4^{2-} concentration in the solution. This observation may seem counterintuitive. This reduction can be explained by various mechanisms, including reactions with other ions present in the water, pH changes induced by $Al_2(SO_4)_3$, or specific reactions related to the particular application of water treatment. These findings emphasize the importance of understanding, in detail, water chemistry and the reactions that occur when chemicals are added for water treatment.

It is also important to note that excessive SO_4^{2-} concentrations can alter the taste of water, imparting an undesirable bitterness and potentially causing laxative effects in individuals unaccustomed to such concentrations [47]. This observation underscores the vital imperative of maintaining appropriate SO_4^{2-} levels in drinking water to ensure both consumer health safety and satisfaction.



Figure 15. Variation in monthly averages of sulfate (mg/L) over time (months) for raw and treated waters.

The rigorous examination of SO_4^{2-} concentrations, coupled with the evident success of $Al_2(SO_4)_3$ treatment, eloquently illustrates the critical necessity to diligently monitor and manage chemical water parameters at all stages of purification [41,86,87]. This comprehensive approach not only ensures compliance with quality standards but also helps preserve taste and consumer safety, thereby reinforcing confidence in the consistent provision of reliable and wholesome drinking water.

3.6. Undesirable Parameters

3.6.1. Iron

Carefully plotted curves representing Fe^{3+} concentrations within the raw water and treated water samples reflect the result of meticulous investigation, and are detailed in Figure 16. A thorough analysis of these curves reveals an intriguing feature: concentrations of Fe^{3+} in both the raw and treated water samples remain remarkably low, maintaining an almost negligible presence, below 0.3 mg/L [41]. Notably, it is remarkable to observe that Fe^{3+} concentration values for raw water samples have been brought down to undetectable levels following treatment (total reduction, especially the initial values were very low), unequivocally demonstrating the undisputed effectiveness of the treatment process in eliminating Fe^{3+} presence from the water (Figure 16). Indeed, $Al_2(SO_4)_3$ reacts with Fe^{3+} ions in the water, resulting in the formation of precipitates and contributing to the removal of Fe^{3+} . The chemical reactions can be articulated as follows.

The detailed mechanistic depiction of the reaction between aluminum sulfate and iron(III) ions, forming iron(III) hydroxide, includes the following simplified steps:

Dissociation of aluminum sulfate:

$$Al_2(SO_4)_3 \to 2Al^{3+} + 3SO_4^{2-}$$
 (10)

Reaction with hydroxide ions:

$$2\mathrm{Al}^{3+} + 6\mathrm{OH}^{-} \to 2\mathrm{Al}(\mathrm{OH})_{3} \tag{11}$$

• Formation of the aluminum–iron(III) complex:

$$Fe^{3+} + Al(OH)_3 \rightarrow Fe(OH)_3 + Al^{3+}$$
(12)

$$Fe(OH)_3 \rightarrow Fe(OH)_3 \downarrow$$
 (13)

In this mechanistic sequence, Al^{3+} ions resulting from the dissociation of aluminum sulfate react with OH^- ions to produce $Al(OH)_3$. Subsequently, Fe^{3+} irons interact with aluminum hydroxide, forming a complex that precipitates as iron(III) hydroxide (Fe(OH)_3).



Figure 16. Variation in monthly averages of iron (mg/L) over time (months) for raw and treated waters.

The precipitate formed is $Fe(OH)_3$, which is insoluble in water and precipitates as a solid. Fe^{3+} is thus removed from the solution in the form of a precipitate. This method is commonly used in water treatment to eliminate Fe^{3+} , which is crucial for improving the quality of drinking water or meeting environmental requirements for wastewater discharge. Once the precipitate is formed, it can be separated from the water using various water treatment methods.

On the other hand, in-depth research by Rodier et al. provides a captivating perspective by highlighting that even at concentrations as minimal as 0.05 mg/L, Fe³⁺ can impart an unpleasant taste to water, potentially affecting consumer perception [47]. This observation underscores the crucial importance of maintaining adequate Fe³⁺ levels to ensure a pleasant and discomfort-free consumption experience.

Furthermore, extensive studies by Friedman et al. have established a substantial link between pipeline corrosion and the release of contaminants like Fe^{3+} into the water distribution network. This interconnection also raises questions regarding the potential impact of cast Fe^{3+} pipelines, which could contribute to Fe^{3+} introduction in the system [41]. Additionally, the accumulation of Fe^{3+} within this system can result from intricate oxidation and settling processes, emphasizing the vital importance of implementing preventive measures to mitigate these undesirable phenomena.

The vigilance given to monitoring and careful management of Fe³⁺ concentrations, combined with tangible proof of treatment efficacy, signifies a rigorous approach to ensuring the quality and safety of drinking water [41]. This proactive approach goes beyond mere compliance with stringent quality standards; it aims to guarantee ongoing consumer satisfaction and reinforce confidence in the reliability of their drinking water supply.

3.6.2. Manganese

Meticulously traced curves representing Mn²⁺ concentrations within raw water and treated water samples are the result of a meticulous and detailed scientific approach, and are revealed in depth in Figure 17. Scrutinizing Figure 17 closely reveals a captivating

revelation: Mn^{2+} concentrations in both the raw and treated water samples remain minute, maintaining an impressively low trace level, below 0.05 mg/L (Figure 17). This remarkable finding unequivocally highlights the unwavering effectiveness of the treatment process deployed in eliminating Mn^{2+} presence from the water. Equally impressive is the fact that Mn^{2+} concentrations in raw water samples have been reduced to undetectable levels following treatment (total reduction, especially when the initial values were very low), offering tangible proof of the undeniable efficacy of this treatment process.



Figure 17. Variation in monthly averages of manganese (mg/L) over time (months) for raw and treated waters.

Indeed, the introduction of $Al_2(SO_4)_3$ into water containing Mn^{2+} ions initiates a reaction leading to the formation of insoluble precipitates—specifically manganese(II) hydroxide (Mn(OH)₂) [88]. The overarching chemical reaction governing the precipitation of Mn^{2+} in the presence of $Al_2(SO_4)_3$ is delineated as follows:

Dissociation of aluminum sulfate:

$$Al_2(SO_4)_3 \to 2Al^{3+} + 3SO_4^{2-}$$
 (14)

Reaction with hydroxide ions:

$$2\mathrm{Al}^{3+} + 6\mathrm{OH}^{-} \to 2\mathrm{Al}(\mathrm{OH})_{3} \tag{15}$$

• Formation of the aluminum-manganese(III) complex:

$$Mn^{2+} + Al(OH)_3 \rightarrow Mn(OH)_2 + Al^{3+} + OH^-$$
(16)

• Precipitation of the complex:

$$Mn(OH)_2 \rightarrow Mn(OH)_2 \downarrow$$
 (17)

In this reaction, Mn^{2+} ions react with hydroxide ions, OH^- , derived from the decomposition of $Al_2(SO_4)_3$, thus forming insoluble $Mn(OH)_2$ precipitate in the water. This precipitate can then be removed by settling or filtration, thereby contributing to water purification by eliminating undesirable Mn^{2+} ions [89].

It is crucial to emphasize that the primary source of Mn²⁺ in surface waters largely stems from industrial activities and fertilizer plants, as confirmed by extensive research by Rodier et al. [47]. In these contexts, Mn²⁺ tends to appear in an oxidized and precipitated form, rendering it particularly amenable to removal through traditional clarification processes [41].

Beyond its impact on water taste quality, it is imperative to note that Mn^{2+} can impart an unpleasant taste to water, even at low concentrations (0.02 mg/L). Moreover, it can lead to the formation of a black layer inside pipelines, which may detach and alter the visual appearance of water, rendering it uninviting for consumption [47]. This finding underscores the crucial importance of carefully monitoring Mn^{2+} levels, even at minimal rates, to ensure a pleasant consumption experience free from taste or visual disturbances. In summary, the rigorous approach adopted to manage Mn^{2+} concentration significantly contributes to preserving the quality and acceptability of drinking water, thus enhancing consumer confidence and satisfaction with their water supply [41].

3.6.3. Aluminum

The carefully traced curves representing Al³⁺ concentrations within raw water and treated water samples reflect the outcome of a meticulous and rigorous scientific investigation, detailed comprehensively in Figure 18.



Figure 18. Variation in monthly averages of aluminum over time (months) for raw and treated waters.

A thorough analysis of data pertaining to raw water reveals both an intriguing and significant observation: Al^{3+} concentrations are virtually imperceptible. However, following the application of treatment involving the use of $Al_2(SO_4)_3$, a marginal presence of Al^{3+} emerges, characterized by negligible levels that align perfectly with standards governing treated waters.

It is essential to highlight the pivotal role played by $Al_2(SO_4)_3$ as a coagulant in water treatment processes. This substance, widely employed to facilitate coagulation and clarification, introduces Al^{3+} into the water as an inevitable byproduct [41,90]. However, cautious vigilance is warranted due to the observation that the use of $Al_2(SO_4)_3$ may lead to a 50% likelihood of increasing Al^{3+} concentrations in treated water compared to raw water [41,91].

This discovery underscores the need to closely monitor Al³⁺ levels in treated water, thereby reinforcing commitment to safety and quality standards. Furthermore, it raises pertinent concerns about potential effects on human health, prompting a judicious and prudent management approach toward the use of Al³⁺-based coagulants in treatment processes. As a result, a proactive and balanced approach is imperative to maintaining drinking water quality while minimizing potential risks associated with Al³⁺ presence [41]. This health-focused approach enhances consumer confidence in the ongoing reliability and safety of their drinking water supply.

3.7. Alkaline Parameters

3.7.1. Potassium

The meticulously traced curves representing K^+ concentrations within raw water and treated water samples are the result of a rigorous scientific methodology, detailed extensively in Figure 19. A thorough analysis of Figure 19 reveals a striking characteristic: K^+ concentrations in raw water do not meet potability standards. In contrast, treated waters adhere perfectly to these stringent norms (Figure 19). It is widely known that Al₂(SO₄)₃ itself generally does not have a significant effect on the concentration of K^+ in water. The chemical reactions that occur when Al₂(SO₄)₃ is added to water are primarily related to the coagulation of suspended particles and the formation of Al(OH)₃ precipitates. K^+ is a distinct chemical element and does not directly react with Al₂(SO₄)₃ within this process [19]. However, it can indirectly react with Al₂(SO₄)₃ in certain situations, depending on the chemical composition of the water and specific conditions. For instance [34,76,81,92]:

- pH of the water: Al₂(SO₄)₃ is often used in water treatment to adjust pH. When added to water, it can react with OH⁻ present in the water to form Al(OH)₃ precipitates. This reaction can influence the water's pH by increasing it, which, in turn, can affect the solubility of K⁺ salts in the water.
- Complexation: In some waters, K⁺ ions can form complexes with OH⁻ ions or with Al(OH)₃ precipitates, depending on the pH and concentration. These complexes can impact the availability of K⁺ in the water. Potassium exhibits versatile complexation behaviors across different chemical contexts. Here are illustrative instances of potassium complexes:
 - Potassium Hydroxide Complex (KOH): In alkaline solutions, potassium ions can form complexes with hydroxide ions, resulting in potassium hydroxide (KOH). The equilibrium reaction is:

$$K^+ + OH^- \rightleftharpoons KOH$$
 (18)

• Aluminum Potassium Complex: In waters containing aluminum ions, potassium ions can form complexes with aluminum hydroxide precipitates. The complexation reaction may involve the formation of species like Al(OH)₄⁻ and can be represented as:

$$K^{+} + (Al(OH)_{4})^{-} \to K(Al(OH)_{4})$$
(19)

• Potassium Cyanide Complex (KCN): Potassium can form complexes with cyanide ions, resulting in potassium cyanide (KCN). The equilibrium can be expressed as:

$$K^{+} + CN^{-} \rightleftharpoons KCN \tag{20}$$

• Potassium Tetrafluoroborate Complex (KBF₄): Potassium can also form complexes with fluoroborate ions, as seen in the compound potassium tetrafluoroborate (KBF₄):

$$K^{+} + BF_{4}^{-} \rightleftharpoons KBF_{4} \tag{21}$$

 Potassium Chloride Complex (KCl): While not a traditional complex, in certain contexts, the dissolution of potassium chloride in water can be considered as a form of complexation:

$$\mathrm{KCl} \rightleftharpoons \mathrm{K}^{+} + \mathrm{Cl}^{-} \tag{22}$$

These examples illustrate different scenarios where potassium ions interact with other ions to form complexes, and each complex has its unique properties and significance in various chemical systems.

As seen in our study, a significant reduction in the concentration of K^+ in the polluted water, approximately a 97.42% reduction, occurred after the addition of $Al_2(SO_4)_3$ in the treatment process. This notable reduction can be attributed to several interrelated factors. Firstly, the initial pH of the polluted water, ranging between 7.65 and 8.19, played a crucial role. At this slightly alkaline pH, $Al_2(SO_4)_3$ reacted with OH⁻ ions in the water

to form Al(OH)₃ precipitates (Al(OH)₃). These precipitates acted as coagulation agents, facilitating the agglomeration of suspended particles. Additionally, the high pH influenced the solubility of K⁺ salts, reducing their presence in the treated water. This reduction in K⁺ could also be due to complex interactions between Al₂(SO₄)₃ and K⁺ ions, forming complexes that decreased the concentration of soluble K⁺.



Figure 19. Variation in monthly averages of potassium (mg/L) over time (months) for raw and treated waters.

This proactive approach to treatment using $Al_2(SO_4)_3$ and monitoring of K⁺ concentrations, coupled with the concrete demonstration of its effectiveness, testifies to responsible and thoughtful management of the drinking water supply. It enhances consumer confidence in the reliability and continuous quality of the water they consume daily, while highlighting sustained efforts to meet the most demanding standards of water safety and quality [41].

3.7.2. Sodium

The meticulously elaborated curves representing Na⁺ concentrations within raw water and treated water samples reflect a rigorous scientific approach, the results of which are detailed in Figure 20. A thorough analysis of Figure 20 highlights a remarkable observation: Na⁺ concentrations in raw water exceed the defined limits for drinking water, while treated waters maintain perfect compliance with these stringent standards (Figure 20). This undeniable finding attests convincingly to the exceptional effectiveness of the Al₂(SO₄)₃based treatment in reducing Na^+ levels. As previously mentioned, $Al_2(SO_4)_3$ is commonly used to coagulate suspended particles in water, allowing them to clump together into larger flocs that can be easily removed through filtration. However, it does not have a direct effect on the Na⁺ concentration. Nevertheless, it can have an indirect effect on the Na⁺ concentration in water, although this effect is generally limited and depends on specific water treatment conditions. When $Al_2(SO_4)_3$ is used as a coagulant in water treatment, it promotes the formation of flocs by aggregating suspended particles. These flocs primarily contain solid impurities present in the water, such as organic matter, colloids, suspended particles, etc. In the coagulation-flocculation process, some soluble impurities, including Na^+ compounds, may also be trapped or adsorbed by the flocs formed by $Al_2(SO_4)_3$. This can result in a reduction in the concentration of Na⁺ in the treated water. This demonstrates the 92.64% reduction that has been recorded.



Figure 20. Variation in monthly averages of sodium (mg/L) over time (months) for raw and treated waters.

It is relevant to note that this achievement extends beyond merely satisfying regulatory requirements; it also holds deeper significance. By ensuring adequate Na⁺ levels in drinking water, this treatment process actively contributes to preserving consumer health and wellbeing. Treated waters become a trusted source for the population, delivering quality water that can be consumed with confidence [41].

The positive implications also extend to the realms of infrastructure safety and environmental sustainability. By maintaining Na⁺ concentrations at acceptable levels, potential risks of pipeline and equipment corrosion are reduced, which in turn can extend their useful life and minimize maintenance costs.

3.7.3. Total Alkalinity

The carefully traced curves representing TAC concentrations within raw water and treated water samples testify to a meticulous and systematic scientific approach, the results of which are minutely detailed in Figure 21. A meticulous analysis of Figure 21 reveals a striking characteristic: TAC values in raw water samples are remarkably high, far exceeding established standards for ensuring water potability. However, this situation takes a significant turn with treated water samples, where a substantial reduction in TAC levels is observed (Figure 21), bringing them into alignment with stringent potability criteria. This undeniable observation vividly highlights the exceptional effectiveness of the $Al_2(SO_4)_3$ -based treatment process in significantly reducing TAC levels, thereby contributing to ensuring the quality of distributed water.

Unlike acidity, which reflects the concentration of hydrogen ions in water, alkalinity represents the presence of bases and salts of weak acids. In the context of natural waters, alkalinity primarily originates from HCO_3^- , carbonates, and hydroxides present. However, it should be noted that other salts of weak acids can also be detected, potentially introducing interferences in the analysis process, such as humic acids, PO_4^{3-} , citrates, and tartrates. Additionally, the presence of ionic silica can impact measurements, especially when the water pH exceeds the threshold of 8.5. This awareness underscores the importance of considering these factors when assessing total alkalinity in water [41,47]. Indeed, $Al_2(SO_4)_3$ itself does not significantly add or remove bases or acids from water. However, it can influence water hardness, which is a measure of the concentration of dissolved Ca^{2+} and Mg^{2+} cations in water. When $Al_2(SO_4)_3$ reacts with these cations, it can form insoluble



precipitates, thus reducing water hardness [47]. This can sometimes lead to a decrease in the TAC of the water, although it depends on the composition of the original water.

Figure 21. Variation in monthly averages of total alkalinity (mg/L of CaCO₃) over time (months) for raw and treated waters.

In our study, a significant removal was recorded for parameters such as electrical conductivity, turbidity, organic matter, NH_4^+ , PO_4^{3-} , NO_3^- , NO_2^- , Ca^{2+} , Mg^{2+} , TH, Cl^- , HCO_3^- , SO_4^{2-} , Fe^{3+} , Mn^{2+} , Al^{3+} , K^+ , and Na^+ in our water treatment system using $Al_2(SO_4)_3$. It is indeed possible that this has an indirect impact on TAC. TAC is largely determined by the presence of HCO_3^- and carbonate (CO_3^{2-}) ions in the water, among other alkaline species [47]. By reducing the concentration of these other ions and forming precipitates with other compounds present in the water, $Al_2(SO_4)_3$ can contribute to a decrease in TAC by removing alkaline chemical species. However, it is important to note that the impact on TAC may vary depending on specific treatment conditions and the initial composition of the water. Therefore, our finding of a reduction in TAC after the addition of $Al_2(SO_4)_3$ seems consistent with the results obtained for other parameters.

The use of $Al_2(SO_4)_3$ in water treatment has demonstrated significant results in modifying the TAC of water. When applied under initial pH conditions ranging from 7.65 to 8.19, this coagulant has shown exceptional effectiveness in reducing water TAC. This reduction, measured at 96.13%, attests to ability of $Al_2(SO_4)_3$ to interact with alkaline compounds in water, leading to the formation of insoluble precipitates. This process not only facilitated water clarification but also resulted in a decrease in pH. These findings highlight the crucial importance of $Al_2(SO_4)_3$ as an effective treatment agent for reducing alkalinity under specific water quality conditions.

Furthermore, it demonstrates that $Al_2(SO_4)_3$ also has an indirect impact on TAC by removing alkaline compounds, in addition to its contribution to the removal of other water contaminants. This further enriches our understanding of the efficacy of this coagulant in water treatment.

3.8. Dry Residue

The carefully plotted curves depicting the concentrations of DR within raw water and treated water samples bear witness to the methodical and rigorous application of a scientific approach, the detailed results of which are thoroughly expounded in Figure 22.



Figure 22. Variation in the monthly averages of the dry residue (mg/L) as a function of time (months) of the raw and treated waters.

The determination of DR in water holds fundamental importance in evaluating its chemical composition. This measurement estimates the total amount of non-volatile and suspended solids present in the water after thorough evaporation. It serves as a key indicator for assessing the quality and composition of dissolved elements in the analyzed water [41,47,93].

Figure 22 offers a vivid visualization of the spatial distribution of DR levels in the raw and treated water samples. A comprehensive analysis reveals that the values of DR in raw water significantly exceed the limits defined for drinking water, with some values even surpassing 2000 mg/L (Figure 22). Conversely, treated water samples exhibit levels that scrupulously adhere to strict potability standards (Figure 22). This tangible observation undeniably highlights the exceptional efficiency of the Al₂(SO₄)₃-based treatment process in substantially reducing DR levels, thereby significantly contributing to ensuring the superior quality of water intended for distribution. Indeed, the use of $Al_2(SO_4)_3$ in water treatment has resulted in a substantial reduction in various water quality parameters, encompassing electrical conductivity, turbidity, organic matter, NH₄⁺, PO₄³⁻, NO₃⁻, NO₂⁻, Ca²⁺, Mg²⁺, TH, Cl⁻, HCO₃⁻, SO₄²⁻, Fe³⁺, Mn²⁺, Al³⁺, K⁺, and Na⁺, as well as the total alkalinity content (TAC). Since all of these parameters have been reduced, including those that directly influence DR, it is evident that DR would be significantly reduced, where DR represents the cumulative amount of dissolved solids in the water, encompassing salts, minerals, and various other compounds. This reduction in DR was indeed remarkable, with an impressive reduction rate of 88.14% observed in the recorded data.

It should be emphasized that the result obtained for DR can be influenced by various parameters, such as temperature and the duration of the evaporation process. At temperatures between 100 and 105 °C, a portion or all of the interstitial water as well as the water of crystallization of certain compounds can evaporate. When the temperature exceeds 105 °C, hydrogen carbonates can transform into carbonates, resulting in the release of carbon dioxide. Beyond these temperatures, complex chemical reactions can occur, involving the loss of NH₃, hydrogen sulfide, and organic matter. Different analysis temperatures, such as 180 °C and 525 °C, thus enable the dissection of the different components of DR and a better understanding of its multifaceted composition [41,47,93].

In summary, the rigorous scientific approach adopted to evaluate DR in water, coupled with tangible proof of treatment effectiveness, attests to a committed stance in preserving

the optimal quality of drinking water, considering the intricate aspects of its composition and characteristics.

4. Conclusions

This comprehensive study on the quality of raw and treated water across a series of key parameters has underscored the crucial importance of the $Al_2(SO_4)_3$ -based treatment process in ensuring high-quality drinking water. The obtained results have enabled an in-depth analysis of concentrations in various elements such as NO_2^- , Ca^{2+} , Mg^{2+} , Cl^- , SO_4^{2-} , Fe^{3+} , K^+ , Na^+ , DR, and many others. The remarkable effectiveness of the Al₂(SO₄)₃based treatment has been clearly demonstrated through a significant reduction in levels of various contaminants throughout the year, thereby ensuring compliance with national and international potability standards. Elevated concentrations of certain elements in raw water, such as Ca²⁺, Mg²⁺, and Cl⁻, have been substantially mitigated after treatment, while other elements like Fe³⁺ have been virtually eliminated, attesting to the undeniable efficiency of the process. These findings also highlight the complexity and interdependence of different parameters influencing water quality, such as alkalinity, hardness, and the presence of dissolved substances. They underscore the need for a comprehensive and balanced approach to maintain the quality of distributed water, while considering public health and infrastructure corrosion considerations. Ultimately, this study demonstrates that continuous monitoring, in-depth scientific research, and the application of effective treatment methods are essential to ensure a safe and high-quality supply of drinking water. The collected data and drawn conclusions contribute not only to the understanding of water's chemical composition but also to the implementation of proactive strategies aimed at upholding the health and safety of the population, while preserving precious water resources.

Supplementary Materials: The following supporting information can be downloaded at: https://www. mdpi.com/article/10.3390/w16030400/s1, Table S1. Water Quality Analysis Throughout the Year: A Comprehensive Overview of Physicochemical Parameters in Raw and Treated Water; Table S2. WHO standards of water quality parameters.

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