RESIDUAL ALUMINIUM IONS IN WATER TREATED WITH 'ROCK ALUM' COAGULANTS

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***ABSTRACT*:** Safe and clean water is one of the essential natural resources necessary for human existence. Demand for clean and safe water has dramatically increased with the increase in human population and industrial developments. In most developing countries, small-scale water consumers use coagulants obtained from commercial shops to treat drinking water obtained from rivers, lakes and bore holes to remove turbidity and other suspended particles. These coagulants are preferred due to their convenience in use, low cost and is widely available in local market. Examples of these coagulants include 'Rock Alum'. Previous studies hypothesized that use of aluminium-based coagulants leaves residual aluminium in water making it unsafe for human consumption. Effects of aluminium exposure include neurological disorders such as Alzheimer's disease, reduced intellectual capability, renal failure, and bone defects. This study focused on analysis of treated de-ionized, river and borehole water samples treated with variable masses of the coagulant 'rock alum' ranging from a maximum of 0.125 g to a minimum of 0.0125 g in 500 cm^3 of water. The concentration of residual aluminium ions in treated water samples was determined using an Atomic Absorption Spectrometer. River water and borehole water samples were obtained from 3 different regions within central Kenya This study found that de-ionized water treated with 0.125 g and 0.0125g of coagulant in 500 cm^3 of water reported concentration of residual aluminium at an average of 7.41 and 1.9 ppm respectively. Analysis of river and borehole water samples treated with 0.125 g of coagulant in 500 cm^3 gave 4.23 and 3.47 ppm respectively. These concentrations are higher than 0.1 ppm which is the permissible limits the World Health Organisation (WHO) set for safe drinking water. These elevated levels are detrimental to consumers' health. These results provide important information to government bodies in charge of water provision and public health on the risks associated with water treated with commercial aluminium-based coagulants. This study recommends improvement in water treatment conditions to reduce residual aluminium ions.

***Index Terms*:** Residual aluminium ions, Neurological diseases, Rock alum and Water treatment.

1. INTRODUCTION

Safe and clean water is one of the essential natural resources necessary for human survival. Approximately 70% of the surface of the Earth's surface is covered by water. However, less than 4% of the water available is fit for human consumption [1]. The demand for clean and safe water has dramatically increased due to increase in human population and industrial developments [2]. In Africa, many residents rely on natural water sources such as rivers, streams, wells, and lakes for domestic uses [3]. Most of these sources provide untreated water which may contain physical debris, biological microorganisms and chemical contaminants. In addition, water may also contain contaminants related to agricultural activities such as farm chemicals and fertilizers [4], [5]. Water contamination can also result from landfills, illegal disposal and industrial processes, discharges of untreated industrial effluents to rivers and streams [6].

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Water treatment commonly involves use of coagulants such as aluminium sulphate, polyaluminium chloride, and iron salts [7], [8]. Upon addition of coagulants, suspended solids, organic matter, chemical contaminants, bacteria and coagulant residuals are removed from water in the form of flocs, which are macro-particles that can be removed through filtration and sedimentation [6], [9]. Most chemical coagulants used in water treatment removes over 90% of turbidity, heavy metals, and both organic and inorganic suspended particles. Other water treatment techniques include, ion-exchange, catalytic reduction-oxidation, membrane filtration, disinfection to mention among others [10]. These commercial water treatment methods are very efficient in water treatment and have low cost when applied in large scale. However, they are not convenient in domestic/ small scale use.

In Kenya and many other African countries, most people lack access to commercially treated, public tap water supplied by county governments. The need for clean water often compels local communities to carry out their own domestic water treatment from local streams, rivers, wells, or boreholes. Home water treatment typically involves the use of aluminium-based coagulants commonly known as "rock alum" purchased from local shops. These coagulants are preferred for domestic uses due to their low cost, ease of application and availability. However, water treated using these coagulants does not undergo quality checks by government agencies such as the Kenya Bureau of Standards (KEBS), unlike water from public treatment plants. As a result, harmful Water Treatment Residuals (WTR) such as residual aluminium ions, may remain in home-treated water, posing health risks to consumers. Water treatment using aluminium sulphate, significantly contributes to residual aluminium ions in the treated water intended for domestic use. Previous studies have reported that water treated using aluminium salts has contains higher concentrations of low molecular mass polyaluminium compounds. These molecules exhibit greater chemical reactivity and are readily absorbed into the human body. Consumption of water with elevated aluminium residues leads to poisoning [7], [11]. According to Krupińska (2020), aluminium sulphate and polyaluminium chlorides are commonly used in commercial drinking water treatment. They facilitate coagulation and remove particulate, colloidal, and other dissolved substances in water. However, residual aluminium ions may remain in water after treatment leading to human exposure upon consumption [9], [12].

Aluminium uptake through the gastrointestinal tract is proportional to between 0.1% and 0.3 %. Absorption occurs primarily in the upper intestines, where the elevated uptake is attributed to the lower pH [13], [14]. According to Alabi & Adeoluwa (2020), the kidneys help minimize the uptake of aluminium but cannot prevent the risk of its accumulation in other parts of the body. Mild exposure to aluminium results in general symptoms such as fatigue, confusion, dizziness and somnolence. Other symptoms include abdominal pains, nausea, vomiting, and sometimes diarrhea. When exposed to aluminium fumes, the victim may experience persistent coughing and difficulty breathing [16].

Acute exposure to aluminium in large quantities over a long period result in metal accumulation in bones, lungs, muscles, liver, and brain, affecting the victim's health. Exposure to aluminium levels exceeding the ability to excrete is directly related to the occurrences of Alzheimer's disease. In addition, other related neurological disorders such as multiple sclerosis and Parkinson's disease are attributed to aluminium exposure [17], [18].

2. MATERIALS AND METHODS

Four different "rock Alum" coagulants were obtained from different manufacturers. The coagulants are readily available in local shops in Kenya. De-ionized water was obtained from Murang'a University of Technology Analytical Research laboratory, while natural water samples were collected from three rivers; Chania, Thika and Maragwa. Boreholes water samples were collected from Juja, Thika and Maragwa in central Kenya. The water samples were analyzed for residual aluminium ions using Atomic Absorption Spectrometer (AAS) Ice 3000 series manufactured by Thermo-fisher in German.

This study investigated the concentration levels of residual aluminium ions in the treated de-ionized water. Water samples were treated using the 'rock alum' coagulants from four different coagulants as shown in Table 1.

Table 1: Different brands of coagulants used based on manufacturer.

COAGULANTS	ACTIVE COMPOUND	MANUFACTURER	NO. OF SAMPLES
Coagulant-1	Aluminium sulphate ($Al_2[SO_4]_3$)	Jolichem Suppliers Ltd., Nairobi, Kenya	3
Coagulant-2	Aluminium sulphate ($Al_2[SO_4]_3$)	Supreme Farmcare Ltd., Nairobi, Kenya	3
Coagulant-3	Aluminium sulphate ($Al_2[SO_4]_3$)	Vetagro suppliers Ltd., Nairobi, Kenya	3
Coagulant-4	Aluminium sulphate ($Al_2[SO_4]_3$)	Avachem Nairobi Kenya	3

Samples of “Rock alum” coagulants used in this study are shown in the images in the Figure 1 below.

Coagulant-1. Coagulant-2

Coagulant-3 Coagulant-4

Figure 1: Different coagulants used in this study.

De-ionized treated water sample was prepared by accurately weighing 0.125 g of coagulant using an analytical balance and adding it to 500 cm³ of de-ionized water. The solution was stirred using a magnetic stirrer for 30 minutes. It was then allowed to stand for 24 hours to enable coagulation and settlement of the flocs. The water sample was filtered using Whatman filter paper (90mm) and labelled as sample A.

Solution B was prepared similarly by dissolving 0.025 g of coagulant in 500 cm³ of de-ionized water and procedure above repeated. De-ionized water sample C was prepared similarly but 0.0125 g of coagulant was used to treat 500 cm^3.Sample D was the control sample where 0.0 g (Zero) coagulant was added to 500 cm^3 of water solution D, as shown in Table 2 [19], [20], [21].

Table 2: Different masses of coagulants used to treat De-ionized water samples

Treated Water Samples	Mass of Coagulant Used (g)	Vol. of De-ionized Water Used (cm^3)
A	0.1250	500
B	0.0250	500
C	0.0125	500
D	0.0000	500

2.1 Treatment of natural water samples

A composite coagulant was prepared by mixing equal masses of all the four different coagulants. This was done by accurately weighing 4 g of each coagulant, mixing them and grinding in a mortar using pestle to form composite coagulant used in treatment of natural water samples. Water samples were collected from 3 different rivers labelled River-1(Chania), River-2(Thika) and River-3(Maragwa). Additionally, water samples were collected from 3 different boreholes, labelled Borehole-1(Juja), Borehole-2(Thika), and Borehole-3(Maragwa). The natural water samples were collected from rivers and boreholes in central Kenya as shown in Figure 2.

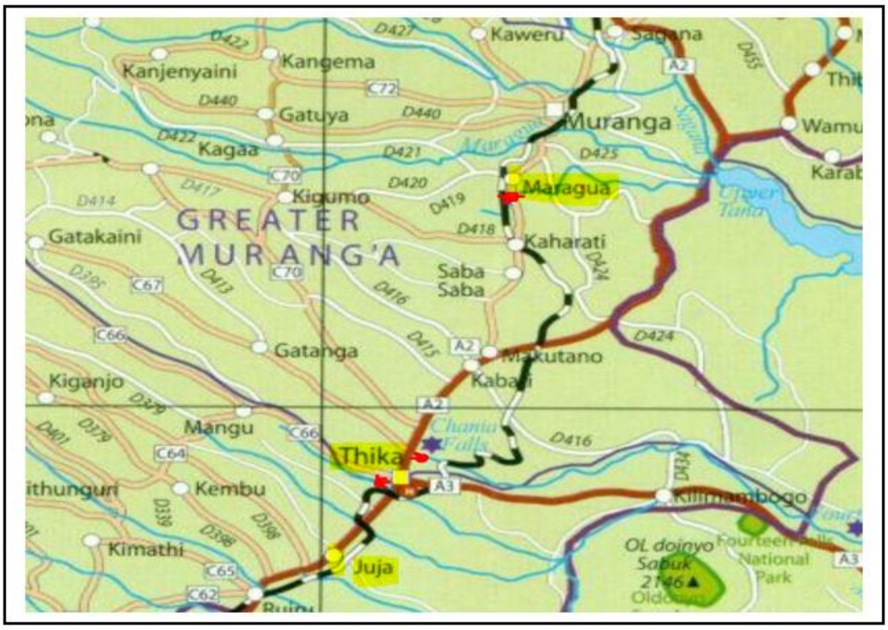

Figure 2: Water sampling sites. Red dots shows river water sampling site (Chania, Thika and Maragwa) rivers from bottom). The yellow dots shows borehole water sampling sites (Juja, Thika and Maragwa).

The natural water samples were treated with the composite coagulant prepared as described above. The mass of the composite coagulant used in water treatment varied from 0.1250, 0.0250 and 0.0125 g each in 500 cm^3 of natural water sample[20], [21]. The treated water samples were filtered using Whatman filter paper no. 41 and placed in labelled 25 ml volumetric flasks using the codes in the Table 2. The samples were analyzed using AAS in triplicate to determine the concentration of residual aluminium (Munyao, 2018; Abdulhussein *et al*., 2024).

3. RESULTS AND DISCUSSION

3.1 Residual aluminium ions in de-ionized water treated with "rock alum"

De-ionized water samples were treated with variable masses of the four coagulants from different manufacturers. The treated water samples were analyzed using atomic absorption spectrometer (AAS). The analysis of residual aluminium ions in the treated water revealed high residual aluminium concentrations. Coagulant-3 gave the highest concentration of residual ions, reaching a maximum of 15.01 ppm when 0.125 g of the coagulant was added to 500 cm^3 of de-ionized water. In contrast, coagulant-1 resulted in the lowest concentration of residual ions (7.41 ppm) for the same under the same conditions. It was observed that when no coagulant was used, the water samples exhibited undetectable levels of residual aluminium ions. This observation indicate that the residual ions originated from the use of coagulants during water treatment. Furthermore, the results showed a direct a direct relationship between coagulant mass and residual aluminium concentration: The greater the mass of coagulants used, the higher the aluminium concentration, as presented in Table 3.

Table 3: Concentrations of residual Al^{+3} ions in treated De-ionized water

Sample	Mass of Coagulant (g)	**Residual Al^{3+} concentrations (ppm)**				
		Coagulant-1	**Coagulant-2**	**Coagulant-3**	**Coagulant-4**	**Average**
A	0.1250	7.41±0.15^{a}	11.39±0.23^{b}	15.01±0.31^{c}	8.99±0.18^{a}	10.7
B	0.0250	2.36±0.05^{a}	4.06±0.08^{b}	4.83±0.10^{c}	3.47±0.07^{b}	3.68
C	0.0125	1.90±0.04^{a}	2.14±0.04^{a}	3.65±0.07^{b}	2.95±0.06^{b}	2.66
D	0.0000	0.00±0.00^{a}	0.00±0.00^{a}	0.00±0.00^{a}	0.00±0.00^{a}	0.00
p-value	**<0.001**	**<0.001**	**<0.001**	**<0.001**	**<0.001**	**<0.001**

Values are means±SE; values with different superscript letters in a row are significantly different at p-value < 0.001.

On average, Table 3 shows that the concentrations of residual ions were higher than the tolerable recommended levels of 0.1mg/L (0.1 ppm) by the World Health Organization (WHO, 2011). Coagulant-1, which had the lowest residual ions, yielded a value of 1.90 ppm when 0.0125 g of coagulant was added to 500 cm^3 of de-ionized water. These elevated levels of aluminium pose a significant health risk to consumers. This highlights the need to reconsider the use of "rock alum" in water treatment since it contributes to human exposure to residual aluminium ions, potentially leading to aluminium poisoning.

3.2 Residual aluminium ions in treated natural water

Unlike in de-ionized water, a composite coagulant prepared by mixing all the four coagulants tested was used to treat natural water samples. These samples were collected from various rivers and boreholes within central Kenya. On analysing the water using AAS, the river water samples, on average, gave lower residual aluminium ions concentration compared to treated de-ionized water. Specifically, when 0.125 g of the composite coagulant was used to treat 500 cm^3 of river and borehole water, average residual aluminium ions concentration were 4.23 ppm and 3.47 ppm respectively. These values were lower than average of 10.7 ppm observed in de-ionized water treated with the same mass of coagulant. The difference was attributed to the absence of suspended particles in de-ionized water which caused negligible turbidity. Consequently, fewer particles were available to form the floc leading to higher concentrations of residual metal ions. As shown in Figure 3, river water samples Therefore, negligible quantities were used to form the flocs, resulting in a high concentration of residual metal ions. In the Figure 3, river water samples formed flocs when treated with the coagulant. These flocs appeared as dirty white residues in the beaker on the right hand side (Figure 3).

Figure 3: Flocs in the treated river water samples

The study observed that flocs are formed from the settlement of suspended particles in the natural water, whereas de-ionized water contained none. Consequently, when equal volumes of natural water and de-ionized water samples were treated with the same mass of coagulant, the natural water samples exhibited lower residual aluminium concentrations than the de-ionized water. This is demonstrated in Tables 4 and 5 which presents the residual aluminium levels in river water and borehole water.

Table 4: Concentrations of residual Al^{+3} ions in treated river water

Sample	**Statistics**	**Mass of Coagulant (g)**	**Residual Al^{3+} concentrations (ppm)**			
			River 1	**River 2**	**River 3**	**Average**
A	N=3	0.1250	4.68 ± 0.09^a	5.73 ± 0.11^b	2.28 ± 0.05^c	4.23±0.89
B	N=3	0.0250	1.04 ± 0.02^a	1.69 ± 0.03^b	1.43 ± 0.03^b	1.39±0.17
C	N=3	0.0125	0.56 ± 0.01^a	0.97 ± 0.02^b	1.51 ± 0.03^c	1.01±0.24
D (Control)	N=3	0.0000	0.98 ± 0.02^a	0.05 ± 0.001^b	0.60 ± 0.01^c	0.54±0.24
p-value	**<0.001**	**<0.001**	**<0.001**	**<0.001**	**<0.001**	**<0.001**

Values are means±SE; values with different superscript letters in a row are significantly different at p-value < 0.001

Table 5: Concentrations of residual Al^{+3} ions in treated borehole water

Sample	**Statistics**	**Mass of Coagulant (g)**	**Residual Al^{3+} concentrations (ppm)**			
			Borehole 1	**Borehole 2**	**Borehole 3**	**Average**
A	N=3	0.1250	4.06 ± 0.08^a	3.33 ± 0.07^b	3.02 ± 0.06^b	3.47±0.28
B	N=3	0.0250	2.03 ± 0.04^a	1.06 ± 0.02^b	0.98 ± 0.02^b	1.36±0.31
C	N=3	0.0125	0.25 ± 0.005^a	0.84 ± 0.02^b	1.01 ± 0.02^c	0.70±0.21
D (Control)	N=3	0.0000	0.20 ± 0.004^a	0.01 ± 0.0002^b	0.00 ± 0.00^b	0.07±0.06
p-value	**<0.001**	**<0.001**	**<0.001**	**<0.001**	**<0.001**	**<0.001**

Values are means±SE; values with different superscript letters in a row are significantly different at p-value < 0.001.

On average river water contained higher concentrations of residual aluminium ions than borehole water. When 0.125 g of composite coagulant was used in 500 cm^3 of water samples, river water samples gave an average of 4.23 ppm while borehole gave 3.47 ppm of residual aluminium ions. At a lower dosage of 0.0125 g of composite coagulant, river and borehole water samples yielded averages of 1.01 and 0.070 ppm respectively.

This observation was attributed to presence of higher concentrations of aluminium ions in untreated water samples in river water samples compared to borehole water. Analysis of the control samples indicated aluminium concentration of 0.54 ppm in river water and 0.07 ppm in borehole untreated water, as shown in Table 6. These elevated levels in river water may be linked to human activities such as the discharge of agricultural and industrial wastes into natural water sources, which contribute to increased residual ions in untreated water samples.

Table 6: Overall comparison of residual Al^{3+} across the three water types

Water Type	**Statistics**	**Mean residual Al^{3+} ±SE (ppm) at different coagulant doses**		
		0.1250g/500mL	**0.0250g/500mL**	**0.0125g/500mL**
De-ionized water	N=12	10.70 ± 1.52^{a}	3.68 ± 0.50^{b}	2.66 ± 0.36^{c}
River water	N=9	4.23 ± 0.89^{a}	1.39 ± 0.17^{b}	1.01 ± 0.24^{b}
Borehole water	N=9	3.47 ± 0.28^{a}	1.36 ± 0.31^{b}	0.70 ± 0.21^{c}
p-value	**<0.001**	**<0.001**	**<0.001**	**<0.001**

These results are consistent with a study conducted by Angelova *et al* (2020) on the determination of aluminium ions in Iskar rivers in Bulgaria, which reported concentrations ranging between 0.148 and 0.199 ppm. This concentration was higher than the recommended maximum limit of aluminium in drinking water set by the environmental quality standards of Bulgaria, which allows up to 0.3 ppm. The high aluminium content in the river water was attributed to the mineral and chemical composition of the local soils [23].

Similarly, a study carried out in Turkey on aluminium content in natural waters and soils, reported that most aluminium in natural water originates from interaction of water with rocks. In the Canakkale-Kirazli region in Turkey, rocks were found to contain approximately 7.8 - 8.2 % aluminium. This contributed to some natural waters containing more than 100 mg/L aluminium, particularly, in acidic conditions. However, aluminium concentration reduced with increase in pH due to low solubility especially with in pH range of 6-8 [24].

A study by [25] analyzed residual aluminium ions in treated water samples from different areas in Malaysia. Water samples were collected from 8 sampling stations from other water treatment plants. The study reported high concentrations of residual aluminium ions. It was observed that the concentration increased with an increase in water acidity, while near-neutral water samples exhibited the lowest concentration. The aluminium concentrations ranged from 0.001 to 8.00 ppm, exceeding the permissible Malaysian legal limits of 0.2 ppm. The residual aluminium ions were attributed to the use of aluminium-based water coagulants in water treatments [25], [26].

Previous studies have shown that natural raw water generally contain lower aluminium levels than treated water. For example [9] investigated the effects of aluminium-based coagulants on residual aluminium ions concentrations in treated and found that untreated water had concentrations ranging between 0.0001 and 1.0 ppm. However, aluminium content increased significantly at lower pH values, particularly below pH of 5. This was attributed to the low solubility of aluminium in water at a pH of 6-8. Therefore, aluminium content is generally lower in natural surface water where the pH is about 7. The aluminium present in natural water results from water interacting with naturally occurring aluminium salts in the soil. The study also reported that tap water contained elevated aluminium ions, attributed to using aluminium salts as coagulants. Aluminium sulphate, when used in water treatment, increases the dissolved aluminium in water due to its lower molecular weight than poly aluminium coagulants [9].

High aluminium content in tap water makes it unsafe for human consumption due to various health challenges related to aluminium poisoning [27]. Consumption of contaminated water results in compromised central nervous system, Alzheimer's disease, Renal failure, bone defects, among others [17]. The total exposure to aluminium via consumption depends on the method of water treatment and the specific coagulant used. The situation is aggravated in areas where treated water already has high natural aluminium content from physicochemical and mineralogical sources [28].

Previous studies have reported that the amount of residual aluminium ions in treated water depends on physical factors such as the amount of coagulant used, coagulation temperature, pH, and turbidity of the treated water [29]. Most developed countries, such as France, Poland, Sweden, Japan, and the United States of America, have set maximum limits for the aluminium content in drinking water to be less than 0.1 ppm. These limits are set to ensure the safety of the consumers [9].

4. CONCLUSIONS

This study concludes that water treated using coagulants contains a high concentration of residual aluminium ions. These concentrations were beyond the tolerable limits of aluminium ions in water meant for human consumption as set by the World Health Organisation (WHO). The study recommends that elevated aluminium content can be reduced or eliminated all together by avoiding the use of excessive aluminium-based coagulants per given volume of water. However, the concentration of residual aluminium ions reduced with an increase in the quantity of Total Suspended Solids (TSS) in water. Therefore, the dosage of coagulant should match the extent of the suspended particles. Also, ensuring adequate mixing by agitation at the dosing stage may reduce residual metal ions as well as providing sufficient settlement time for maximum flocculation and efficient floc filtration.

The study recommends application of water remediation techniques to remove residual metal ions from treated water before human consumption. These methods include adsorption, membrane filtration, ion exchange, electrochemical degradation, photo-Fenton oxidation, and reverse osmosis.

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