



Assessment of Kaolinite as a Natural Remediation Agent for Lead (Pb) Contamination in Groundwater in Mirerani, Tanzania

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Abstract

This study explores the efficacy of kaolinite as a natural remediation for lowering lead (Pb) concentrations in groundwater resources in the Mirerani Manyara Region, Tanzania. The mining site exhibited a notably high mean Pb concentration of 0.025 mg/L, which exceeds the World Health Organization's guideline of 0.015 mg/L. In comparison, other regions, including agricultural (0.016 mg/L), residential (0.014 mg/L), and commercial (0.016 mg/L) areas, demonstrated lower Pb levels, yet remained concerning due to their proximity to mining operations. Adsorption of lead ions by kaolinite is a key natural mechanism for reducing Pb concentrations in water. The lead (Pb) levels initially ranged from 0.001 to 0.411 before the application of kaolinite, with a mean value of 0.019 and a standard deviation of 0.032. After the application of kaolinite, lead levels decreased with significant p-value of 0.0001, readings ranging from 0.000 to 0.026. The mean difference between initial and final Pb levels was 0.019 mg/L, with the removal efficiency (85%) /adsorption capacity of 0.385 mg/L at 95% confidence level with pH ranging from 4.4 to 8.9, with a stable decrease pH to 7. -and 80-minutes contact time. These findings indicate that kaolinite is a natural promising, sustainable, and cost-effective approach for treating Pb-contaminated groundwater. It offers insights into the application of natural remediation methods for Lead heavy metal removal.

Keywords · Keywords: Heavy Metal Lead (Pb) Adsorption, Groundwater pollution, Kaolin clay, natural remediation method.

1. Introduction

Lead (Pb) contamination in water is a pressing global concern that affects numerous regions worldwide. Investigations of this issue have highlighted the severity of contamination and explored various remediation methods. In the United States, for instance, the presence of lead in Groundwater sources has emerged as a significant challenge, predominantly attributed to ageing lead pipes in older infrastructure. A notable example is the Flint water crisis, where lead concentrations in drinking water exceeded 100 times the Environmental Protection Agency's (EPA) action level of 0.015 mg/L

due to inadequate corrosion control measures (Sobus, 2018).

Globally, approximately 2.2 billion individuals lack access to safe drinking water services, particularly in low-income and rural areas where the infrastructure for water purification and distribution is often insufficient. These populations frequently rely on natural water sources tainted by pathogens, heavy metals like Lead poisoning, and other harmful contaminants (Shabani, *et al.*, 2020). However, issues such as aging infrastructure, pollution, and sporadic regulatory lapses continue to pose risks, as evidenced by the Flint crisis (Ruckart *et al.*, 2019).

In developing countries, over 40% of the population lacks access to improved drinking water sources (UNESCO/WHO, 2019). Urbanization in many African cities has exacerbated lead exposure in drinking water due to factors such as uncontrolled traffic, rapid road expansion, industrial activities, and inadequate urban planning (Marandu, 2024). Consequently, lead concentrations in drinking water often surpass the World Health Organization's (WHO, 1989) guideline of 0.01 mg/L in several African nations (Obeng-Gyasi, 2022). Alarming, studies indicate that lead exposure accounted for approximately 54.3% of deaths among all age groups in Sub-Saharan Africa in 2017, with significant mortality linked to lead contamination in several countries, including South Africa (Bede *et al.*, 2018).

In Tanzania, lead exposure remains a significant environmental and public health concern, impacting groundwater sources and biodiversity. Research has identified lead concentrations in Tanzanian groundwater that exceed the WHO's recommended threshold of 0.01 mg/L (WHO, 2003). Access to clean drinking water is particularly challenging in rural areas, which rely heavily on groundwater for both domestic and agricultural needs (Fang *et al.*, 2019). Lead contamination levels above 0.01 mg/L pose considerable risks to human health and the environment, especially in regions with a history of mining, farming, or industrial activities as explained

by EWURA, in the year 2020. In response to these challenges, Tanzania has implemented measures to combat lead contamination, with an acceptable limit set at 0.01 mg/L under Tanzania Bureau of Standards (TBS, 2016).

In Mirerani, various government agencies, including the Ministry of Water and the National Environment Management Council (NEMC), conduct regular monitoring of water quality to facilitate prompt responses to contamination and Lead removal (Tarimo, 2007; and Ahmad *et al.*, 2022). There are natural remediation methods, such as utilizing natural materials like clay, sand, peat moss, and goethite, which can act as adsorbents. Banana peels and orange peels can also be used to remove lead through bio-adsorption as well as reverse osmosis (Rodriguez, 2022).

This study selected kaolinite clay, which is cost-effective solution for reducing lead concentrations, in rural areas lacking sophisticated water treatment infrastructure. Despite ongoing efforts, lead concentrations in Groundwater of Mirerani, where levels range from 0.015 to 0.91 mg/L, exceed both the WHO, 2020 and Tanzania's limits of 0.015 and 0.01 mg/L, respectively. Lead affects multiple body systems, neurological, haematological, gastrointestinal, cardiovascular and renal systems, causing cancer (WHO, 2019). The Mirerani community faces severe health implications due to

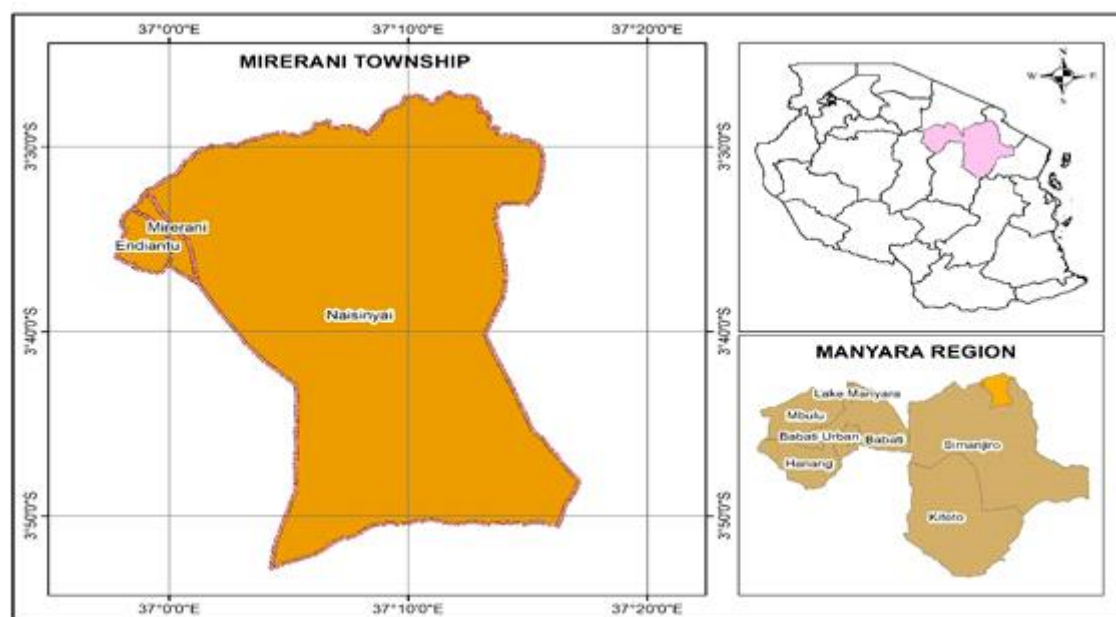


Figure 1: Location of Mirerani Area (*Mirerani Township Authority (2024)*).

elevated lead levels in shallow wells. This study aims to assess lead (Pb) concentration in Groundwater sources in the Mirerani area and assess the efficacy of kaolinite as a natural remediation method in the Manyara Region of northern Tanzania.

2. Materials and Methods

2.1 Description of the Study Area

This study was carried out in the Mirerani area (Naisinyai, Endiamtu, and Mirerani Wards), which lies between latitude 03° 34' 59" South and longitude 37° 0' 29" East. The administrative unit is in the Simanjiro district of the Manyara region, northern Tanzania. Lead (Pb) contamination in Groundwater poses a serious environmental threat (WHO, 2017). Kaolinite, a natural adsorbent, has shown potential in mitigating Pb levels in water (Makris *et al.*, 2007 and Kumar *et al.*, 2018). Prior research confirms its efficacy in removing heavy metals from water (Fang *et al.*, 2019; Zhang *et al.*, 2020).

This study focuses on the application of kaolinite in the Mirerani area of Tanzania, where significant Pb contamination has been documented (Tarimo, 2007 & Mkongo *et al.*, 2019). Lead exposure, particularly through contaminated water, poses grave health risks, notably for children and pregnant women, potentially leading to lead poisoning and resulting health issues such as developmental delays, cognitive impairments, and increased risks of cardiovascular diseases in adults. Various techniques have been historically employed to remediate lead in water, with kaolinite's natural method being the focus of this analysis, utilizing kaolin clay to effectively adsorb lead ions. By passing water through kaolinite clay particles, lead ions are captured, allowing for their removal from drinking water. This method is favored due to kaolinite's accessibility, cost-effectiveness, and environmental sustainability. The study manipulates concentrations in water samples to assess the efficacy of the kaolinite method and optimize its operational parameters.

2.2 Data Collection

This study employed a quantitative research method whereby 223 Groundwater samples were collected. The Groundwater samples were collected from various sources, including wells and boreholes in the Mirerani Area. The Taro Yamane Formula (Yamane, 1970) was used to calculate the study sample size, $n = N/(1+Ne^2)$, where n = sample size, N = population under study, where in this study included 502 Groundwater sources allocated in Mirerani, Naisinyai and Endiamtu wards, and e = margin of error, which in our case is 5%. This formula is used because the population is finite, hence: $n = 502/(1+502*(0.05)^2)$; hence $n = 223$.

The study used a stratified sampling technique to select 223 Groundwater sources. In this technique, the Groundwater sources were divided into three wards: Mirerani ward, which had 207 Groundwater sources; Naisinyai ward, which had 163 Groundwater sources; and Endiamtu ward, which had 132 Groundwater sources. To determine the proportion of sample sizes for each ward based on their respective populations, the study calculated the following: For Mirerani ward, the proportion of Groundwater sources was 207/502. When multiplied by the total number of water sources in the study (223), this resulted in a sample size of 91 water sources. For Naisinyai ward, the proportion of Groundwater sources was 163/502. When multiplied by the total number of water sources in the study (223), this resulted in a sample size of 73 water sources. For Endiamtu ward, the proportion of Groundwater sources was 132/502. When multiplied by the total number of water sources in the study (223), this resulted in a sample size of 59 water sources.

After determining the proportional sample sizes for each ward, the study randomly selected the required number of water sources from each ward. This included 91 Groundwater sources from Mirerani ward, 73 Groundwater sources from Naisinyai ward, and 59 Groundwater sources from Endiamtu ward, making a total of 223 samples. Samples were collected by sterile plastic 1L bottles and were

labelled, kept in a cool and dark environment to minimize degradation, kept at 4°C stored in HNO₃ acid-washed bottles, EPA (301) _AF. R3 protocol standards. About 1.599 g of lead nitrate/1000 mg/L water was used-contact time 30 minutes and cools 30 minutes. Control samples were from other areas away from mining areas, but the characterization of Kaolin material was not done in this investigation.

2.3 Sample Preparation, Dosage, and pH range

Subsequently, collected samples underwent filtration through a 0.45 µm membrane filter. This process aimed to remove any suspended particles and bacteria present in the samples. The Kaolinite materials were prepared for water treatment in the laboratory by initially subjecting them to the grinding

accounted for increased adsorptive sites and surface area to increase adsorption efficiency at pH range of 5 to 6 which reached maximum at pH 7 due to neutralization. Analytical Grade (A.G.) Nitric acid (HNO₃) with a specific gravity of 1.4 (65%) and hydrochloric acid (HCl) with a specific gravity of 1.18 were utilized. Kaolin _Al₂Si₂O₅(OH)₄-activated by acid (H₂ SO₄) treatment to increase porosity, & surface area for Pb adsorption.

2.4 Data Analysis

Atomic Absorption Spectrometry (AAS) is the most common technique for determining biological and environmental samples. The Arusha Urban Water Supply and Sanitation Authority (AUWSA) laboratory analyzed the water samples to test for lead (Pb) concentrations. The analytical method also

Table 1: Levels of Lead (Pb) Concentration in Groundwater Sources in the Mirerani Area (Source: Field Data, 2024 and WHO/FAO: Rajat Nag and Enda Commins, 2022)

AREA	Minimum	Maximum	Mean	Std. Dev	WHO (2011)
Agriculture area	.00300	.05700	.0158935	.01084764	0.1 mg/kg
Residential area H ₂ O	.00300	.11000	.0144247	.01437486	10 µg/l
Commercial area H ₂ O	.00300	.05200	.0162222	.01176462	0.001 m/L
Mining area H ₂ O	.00100	.41100	.0255301	.04967586	0.005 mg/L
Mirerani Overall	0.0010	0.41100	0.019000	0.0325646	(5 µg/L)

and sieving process to achieve a specific particle size. The Kaolinite was then washed and dried to remove impurities before being activated through acid (HCl) treatment and dried thoroughly to increase its porosity and surface area and create more adsorption properties for lead (Pb) reduction in Groundwater. The filtered water samples were then treated with kaolinite natural clay. The kaolinite was added to the samples at a high dosage range starting with 5 g/L to 8.5 g/L, and the mixture was stirred for 30 minutes at room temperature. The treated samples were then allowed to settle for 30 minutes before being filtered through a 0.45 µm membrane filter. The study selected dosage range (5–8.5 g/L) high which

included statistical data analysis using the Statistical Package for Social Science (SPSS, Version 26) programme (Davis, 2021). The package conducts multivariate statistical analyses, including correlations, factors, and clusters of statistical data. Microsoft Excel and the GraphPad instant program were used for paired sample t-test correlations and the Chi square (χ^2) test on level of significance.

The filtered samples were then analyzed for lead (Pb) concentrations using an AAS instrument. The AAS instrument was calibrated using a series of standards with known lead (Pb) concentrations. The instrument was also equilibrated with a blank sample to remove any background signal. The AAS data was

Table 2: Measurement of Lead (Pb) and pH levels (Source: Field Data, 2024)

	N	Minimum	Maximum	Mean	Std. Dev
Initial Pb	223	0.001	0.411	0.019	0.032
Final Pb	223	0.000	0.026	0.004	0.004
Initial pH	223	4.370	8.910	7.523	0.435

analyzed using a statistical software package. The Pb concentration was Pb 1.599 g of lead nitrate/1000 mg/L calculated based on the peak area and the calibration curve. All measured lead concentrations in both the control and experimental groups were recorded and compared against the World Health Organization's (WHO) guideline for lead in drinking water, which is 0.015 mg/L. This comparison helped assess the water samples' compliance with the acceptable lead concentration levels. The lead concentrations in the water samples were compared against the WHO guideline for lead in drinking water, which is 0.015 mg/L, to assess compliance with acceptable levels.

3. Results and Discussion

3.1 The Efficacy of Kaolinite as a Natural Remediation for Lowering Lead (Pb) Concentrations in Groundwater Sources in Mirerani

The study found significant variations in lead concentrations across the agricultural, residential, and commercial sectors. In the agricultural sector, lead levels ranged from 0.003 mg/L to 0.057 mg/L, averaging 0.016 mg/L, suggesting potential contamination from lead-based pesticides or fertilizers. This is concerning as it exceeds the WHO guideline of 0.01 mg/L for lead in drinking water, indicating the need for monitoring and mitigation measures. In the residential sector, lead concentrations ranged from 0.003 mg/L to 0.051 mg/L, with an average of 0.014 mg/L, likely due to lead-based paints or pipes in older buildings, also exceeding WHO (2011) guidelines. The commercial

sector showed lead levels from 0.003 mg/L to 0.052 mg/L, with an average of 0.016 mg/L, posing a risk of lead contamination in drinking water. The mining sector had the highest mean lead concentration at 0.025 mg/L. Excessive lead concentrations in these sectors can lead to various health problems. In children, lead exposure can cause cognitive and behavioral issues, such as reduced IQ and developmental delays, while adults may face higher risks of high blood pressure, heart disease, stroke, and kidney damage. Lead can also affect reproductive health, causing fertility issues and pregnancy complications. These findings align with WHO (2019)'a study which revealed that, lead exposure impacts cognitive performance, growth, and hearing in children, and causes cardiovascular, central nervous system, kidney, and fertility problems in adults.

The study found that using the kaolinite natural method effectively reduces lead concentration in Groundwater by 85% while maintaining water quality. The pH levels remained relatively stable, ranging from 4.370 to 8.910, with a slight decrease in mean pH to mean 7.5 with Sdv 0.4. This dual benefit highlights kaolinite as a cost-effective, environmentally friendly solution for lead contamination. These findings align with Karakaya *et al.* (2021), who noted kaolinite's ability to stabilize water, and Yang *et al.* (2020), who emphasized its low cost and eco-friendly.

Table 3: One-Sample Test

	Test Value = 0			Mean D	95% Confidence Interval of the Diff	
	t	df	Sig. (2-tailed)		Lower	Upper
Initial Pb	8.815	222	.0001	.019052	.01479	.02331
Final Pb	14.711	222	.0001	.003668	.00317	.00415

3.2 The Effectiveness of Kaolinite Natural Method in Reducing Lead (Pb) Concentrations in Groundwater Sources

The study employed t- test and linear regression to determine the relationship between kaolinite natural methods and reducing lead (Pb) concentrations in Groundwater sources. The findings in Table 3 revealed that the application of kaolinite effectively reduced lead (Pb) concentrations in Groundwater sources in the Mirerani Area. Initially, lead levels ranged from 0.001 to 0.411 mg/L. After the application of kaolinite, these levels decreased significantly by 85% overall in the three areas investigated, with readings ranging from 0.000 to 0.026 mg/L. The findings of this study are in the same line with those of Mwanga, (2020); Abdul-Razzaq *et al.* (2020) and Al-Saad *et al.*, (2021) study which

Regression model summary revealed a R square value of 0.471, which signifies that approximately 47.1% of the variation in lead concentrations could be explained by the presence of kaolinite in the water. The Adjusted R square value was 0.469, which is slightly lower than the Initial Pb R square 0.079 value due to the removal of multicollinearity between predictors in water. The standardized coefficient, Beta was 0.687 approximately 68.7%, suggesting increase in the final Pb concentration emphasize the importance of addressing high initial lead concentrations before applying kaolinite remediation methods to ensure optimal results.

The study found that the application of kaolinite effectively reduced lead (Pb) concentrations in Groundwater sources in the Mirerani Area. The findings of this study align with Sakar and Makris, 2020; Hussain and Khaleefa (2021)'s studies which

Table 4: Coefficients Results (Dependent Variable: Final Pb)

Model	Unstandardized Coefficients		Standardized Coefficients	t	Sig.
	B	Std. Error	Beta		
1	(Constant)	.002		10.226	.000
	Initial Pb	.079	.687	14.041	.000

found that kaolinite natural method and bentonite clay had a high capacity for adsorbing heavy metals, particularly lead, from contaminated water sources in Tanzania.

The findings in Table 4 also revealed that was significant relationship between application of Kaolinite Natural Method and reduction of Lead (Pb) concentrations in Groundwater Sources, since the p-value was 0.00 which is less than 0.05.

found that the kaolinite is a natural method that is both readily accessible and cost-effective. Kaolinite's affordability makes it an attractive option for large-scale applications, while its environmental friendliness is emphasised by the fact that it produces no toxic by-products or adverse environmental impacts. Also, kaolinite has dual benefits of economic viability and environmental safety, reinforcing its suitability for widespread use in environmental

remediation efforts. Thus, kaolinite is effective in maintaining water quality and stability while reducing lead levels, thereby emphasizing its role as a key component in sustainable environmental management practices.

4. Discussion

4.1 The Efficacy of Kaolinite as a Natural Remediation Strategy for Lowering Lead (Pb) Concentrations in Groundwater Sources in Mirerani

The study found significant variations in lead concentrations across the agricultural, residential, and commercial sectors. In Table 1, the agricultural sector, lead levels ranged from 0.003 mg/L to 0.057 mg/L, averaging 0.016 mg/L, suggesting potential contamination from lead-based pesticides or fertilizers. This is concerning as it exceeds the WHO guideline of 0.015 mg/L for lead in drinking water, indicating the need for monitoring and mitigation measures. In the residential sector, lead concentrations ranged from 0.003 mg/L to 0.051 mg/L, with an average of 0.014 mg/L, likely due to lead-based paints or pipes in older buildings, also exceeding WHO, 2020 guidelines. The commercial sector showed lead levels from 0.003 mg/L to 0.052 mg/L, with an average of 0.016 mg/L, posing a risk of

lead contamination in drinking water.

The mining sector had the highest mean lead concentration at 0.025 mg/L as presented in Figure 2. The excessive lead concentrations in these sectors can lead to various health problems. In children, lead exposure can cause cognitive and behavioural issues, such as reduced IQ and developmental delays, while adults may face higher risks of high blood pressure, heart disease, stroke, and kidney damage. Lead can also affect reproductive health, causing fertility issues and pregnancy complications. These findings align with Santucci and Scully, (2020) & WHO (2020)' studies which revealed that, lead exposure impacts cognitive performance, growth, and hearing in children, and causes cardiovascular, central nervous system, kidney, and fertility problems in adults.

High lead (Pb) concentrations in the environment have significant environmental and socioeconomic implications. Environmentally, lead is a neurotoxin that can cause decreased growth and reproduction in plants and animals, and neurological effects in vertebrates. Socioeconomically, lead exposure can lead to reduced productivity and a drag on GDP, particularly in low and middle-income countries (World bank, 2023).

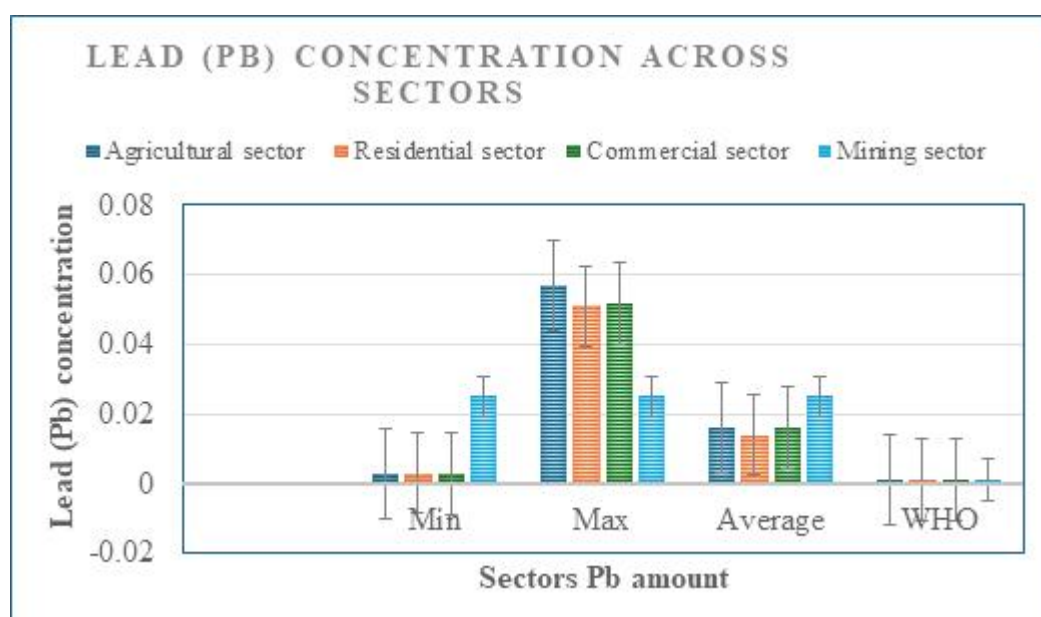


Figure 2: Lead (Pb) concentration across sectors in Mirerani areas (2024)

4.2 The Effectiveness of Kaolinite Natural Method in Reducing Lead (Pb) Concentrations in Groundwater Sources

The study in Table 2 found that, after applying the kaolinite natural method to reduce lead concentration in Groundwater, the water quality was maintained. Various factors contribute to the interaction between the Pb ions and the kaolinite surface. At lower pH values, the surface of the kaolinite material possesses active sites that become positively charged. The positive charge on the kaolinite surface is pH-dependent which increases with decreasing pH values. This leads to the presence of Al-O-H₂⁺ kaolinite species. As a result, there is a competition between H⁺ ions and Pb metal ions for available kaolin adsorption sites. However, as the pH increases, this competition diminishes due to increased negative charge of these active sites on the kaolinite surface. For instance, pH levels remained relatively stable, ranging from 4.4 to 8.9, with a slight decrease in mean pH to 7. Other factors are initial Pb concentration, dosage, contact time and other competing ions. Optimizing the contact time can maximize the removal of lead from contaminated water (Prozialeck *et al.*, 2022). By ensuring sufficient interaction time, it is possible to achieve higher levels of lead reduction, resulting in cleaner and safer water for consumption and use. The study findings imply that employing the kaolinite natural method results in a dual benefit: not only does it effectively lower lead concentrations, but it also helps maintain the stability and quality of the water samples.

This dual advantage highlights the significant potential of kaolinite as a cost-effective and environmentally friendly solution for addressing lead contamination in water sources. The findings of this study are on the same line with those conducted by Karakaya *et al.*, (2021) which found that, kaolinite improves water stability due to its unique properties. When added to water, kaolinite particles create a stable suspension by bonding with water molecules and forming a protective layer on the surface. This layer helps to prevent the coalescence of water droplets and the settling of suspended particles,

thereby increasing the overall stability of the water. Additionally, kaolinite has a high cation exchange capacity which allows it to adsorb and remove contaminants from the water, further enhancing its stability. Furthermore, the study findings implying that the application of kaolinite not only contributes to effective environmental management practices but also proves to be a cost-effective and eco-friendly approach. By utilizing kaolinite, authorities in Mirerani areas can enhance their sustainability efforts, reduce operational costs, and minimize their environmental footprint. These findings align with the study by Yang *et al.* (2020) which revealed that kaolinite is a natural, abundant, low-cost, and eco-friendly material. However, kaolinite's effectiveness compared to other methods like; Electrokinetic remediation (EKR), Biochar and phytoremediation can be limited by its lower surface area and cation exchange capacity compared to other clay minerals like montmorillonite, which may require acid activation modifications to enhance its performance.

5. Conclusions

This study revealed that the mining area of Mirerani exhibits the highest mean lead concentration at 0.025 mg/L, highlighting substantial environmental and health risks associated with mining operations. In contrast, lower mean lead concentrations were observed in agricultural (0.016 mg/L), residential (0.014 mg/L), and commercial (0.016 mg/L) areas. This treatment did not significantly impact other critical water quality parameters, such as pH, thereby ensuring the water's safety and suitability for consumption. Based on these findings, the study concludes that kaolinite natural method is an effective strategy for reducing lead (Pb) concentrations in Groundwater within Mirerani, Manyara Region, Tanzania. These provide useful information on the use of kaolin clay as non-hazardous and efficient adsorbent in removal of Lead (Pb) from contaminated groundwater.

It is recommended that the government maintain ongoing monitoring of Lead levels in Groundwater in the Mirerani areas to ensure compliance with safety

standards. Furthermore, the widespread adoption of the kaolinite natural method is encouraged in regions experiencing elevated Lead concentrations in their water sources for sustainable groundwater quality management.

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