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Hydrogeochemistry and human health risk assessment of heavy metal pollution of groundwater in Tarkwa, a mining community in Ghana

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ABSTRACT

The current study investigated the geochemical characterization and contamination of groundwater quality in Tarkwa, a mining community in Ghana. Total dissolved solids, electrical conductivity, calcium, magnesium, sodium, potassium, total dissolved solids, total hardness, heavy metals, and cyanide (CN) were all assessed in 74 groundwater samples. Other parameters assessed included pH, electrical conductivity, carbonate, bicarbonate, chloride, sulphate, nitrate, and pH. Using the method recommended by the USEPA, a health risk assessment was carried out. Among the parameters which exceeded their WHO recommended threshold were pH (4.9-7.7), Conductivity (150-1082), Turbidity (1.4-139.5), Magnesium (1.9 mg/L-395.5 mg/L), Nitrate (4.5 mg/L-760 mg/L), phosphate (2.3 mg/L-80 mg/L), Sulphate (10 mg/L-855.6 mg/L), CN (1.6mg/L-16 mg/L), As (0.001mg/L-0.015 mg/L), Hg (0.001 mg/L-0.005 mg/L), Cd (ND-0.009 mg/L), Mn (0.004 mg/L-1.38 mg/L), Ni (0.002 mg/L-0.118 mg/L). The study also showed that the groundwater in Tarkwa is naturally hard, making Ca and Mg ions the main ions determining the chemistry of the water. Similar to this, the most significant factors affecting water chemistry were rock dominance and precipitation dominance. Due to high levels of CN, As, and Cd, health risk evaluation revealed that prolonged intake of groundwater from Tarkwa is linked to both carcinogenic and noncancerinogenic health risk. Children were shown to be more vulnerable to both carcinogenic and noncancerinogenic health risks than adults.

Key words: Water Quality, Gibbs plot, hydrogeochemistry, Risk assessment, Groundwater.
1.0 INTRODUCTION
Currently, 2.4 billion people lack adequate sanitation, and 784 million people lack access to cleaner water sources (WHO, 2017). About 187 million people use surface water for drinking, with the majority of them (94%) living in remote areas in Sub-Saharan Africa (WHO/UNICEF, 2012). Groundwater that has been drilled as wells or boreholes primarily supplies rural and para-urban areas in Ghana, whereas treated surface water primarily supplies the country's urban centers. However, the majority of surface waters have become polluted as a result of industrial activities, and as a result, many people have turned to using groundwater. However, due to the high pollution levels, Ghana's groundwater resources have also become susceptible to contamination.

Groundwater is a less valuable resource than surface water because of its resistance to climate change, protection from common sources of pollution, and low cost of treatment before use (Carrad et al., 2019). However, according to Ntanganedzeni et al. (2018) and Tay et al. (2017), anthropogenic or natural contamination of groundwater can occur. The quality of groundwater can be affected by the weathering of rock and the related silicate, carbonate, and sulphate minerals, which is naturally facilitated by high temperatures and low pH levels (Chegheleh et al., 2020). As a result, groundwater's interactions during the hydrological cycle have an effect on its chemical composition. Due to this interaction, groundwater quality may be impacted by concentrations of undesired elements leaching into the ground (Ali, 2016). Similarly, commercial, residential, municipal, industrial, and agricultural activities could have an impact on groundwater composition (EPA, 1993). If not handled effectively, a number of practices, including the application of fertilizer and agrochemicals, leaks from landfills, and septic tanks, could contaminate groundwater (Amankona, 2010).

Illegal mining, or "galamsey," is currently Ghana's most concerning cause of groundwater and surface water pollution. These illegal miners engage in unethical practices, including the use of prohibited materials like cyanide and mercury (Hg). Similar to this, dredging soils and sediments in search of gold entails removing contaminants that have been buried in the sediment. As a result, water supply and quality have decreased in most mining settlements in Ghana (Donkor et al., 2006; Kuma, 2004). Due to the illegal mining activities and the depletion of water resources,
many communities in Ghana, mining locations run the risk of developing health issues brought on by considerably contaminated water bodies (Yeboah, 2013).

Groundwater hydrogeochemistry has been used to study the influence of human activity on groundwater quality, as well as the detection and interpretation of groundwater quality (Zakaria et al., 2021; Gibrilla et al., 2011; Kortatsi et al., 2008). Additionally, numerous hydrogeochemistry investigations conducted in Ghana were successful in identifying the causes of anthropogenic groundwater contamination as well as the dissolution of silicates. Anim-Gyampo et al., 2018, showed that weathering of silicates and cation exchange are the main factors that influence groundwater chemistry in the Atankwidi Basin. Similar findings were published by Egbi et al. (2018), who also named salinity, cation exchange, and silicate weathering as the primary determinants of groundwater chemistry. In 2019, Loh et al. investigated the hydrogeochemistry of the groundwater in Northern Ghana and identified anthropogenic sources of contamination. This stipulates the importance of hydrogeochemistry studies of water quality.

One of Ghana's largest mining communities, Tarkwa is renowned for having abundant water resources that are used for domestic and agricultural purposes. It is recognized as a center for small-scale and illegal mining, nevertheless, 90% of the 20,000 small-scale miners operating now are illegal (Asklund and Eldvall, 2005). Numerous problems concerning heavy metal pollution of water bodies suggest that the community's water may not be of the highest quality (Kortatsi, 2004; Kuma, 2004). Poor water quality has been documented in mining regions both in Ghana and around the world in a number of studies (Arah, 2015; Gyamfi et al., 2019; Asare-Donkor and Adimado, 2013). It is well known that mining operations speed up the weathering and leaching of rocks, which can change the chemistry of groundwater. The hydrogeochemical study is one of the most important techniques for identifying and controlling the ways that groundwater interacts with rock minerals (Jeevanandam et al., 2007). For instance, prolonged aquifer storage, mineral species dissolution, and soil rock water contacts during groundwater flow and recharge are some of the interaction activities that are responsible for groundwater chemistry.
Acid mine drainage (AMD) causes groundwater in Tarkwa to get contaminated with heavy metals as a result of the area's high level of mining activity. AMD has become a brand-new environmental concern in Ghana with the start of open pit mining in 1990 (Hilson, 2002). Waste rocks that were later discovered to produce acid were dumped around the resulting pit, without any protections (Asamoah et al., 2009). Several researchers have documented acid mine drainage in Tarkwa (Kuma and Asamoah, 2009; Asamoah et al., 2009; Ofori-Sarpong and Amankwa, 2019). There is a need to evaluate the groundwater hydrogeochemistry and contamination because high AMD causes the leaching, dissolving, precipitation, and ion exchange processes of aquifer minerals, which have a significant impact on groundwater contamination and groundwater chemistry (Kangjoo, 2003; Panneerselvam et al., 2023; Li et al., 2021). Additionally, the extensive agricultural operations in Tarkwa could result in the leaching of minerals into groundwater resources, including phosphates and nitrates (Economou-Eliopoulos and Megremi, 2021; Li et al., 2021). In a same vein, it's been asserted that small-scale miners use chemicals like CN and Hg, which may contaminate groundwater in Tarkwa. (Emmanuel et al., 2018; Hou et al., 2023).

Water bodies that have been contaminated can have a variety of negative effects, especially because they can serve as a conduit for toxins to enter the food chain. Aside from being used as sources of drinking water, contaminated water used for irrigation or for animal use will transfer its pollutants to humans when they consume such things (Osae et al., 2023). The chemistry and quality of groundwater will be evaluated in this study together with the hydrogeochemical mechanisms that affect them. It will also assess the potential health effects of exposure to cyanide, heavy metals, and other pollutants in the groundwater in the study area. As a result, the physicochemical properties, major ions, trace metal elements, and nutrient content of groundwater samples would be investigated. Additionally, the USEPA's methodologies, such as estimated daily intake (EDI), hazard quotient (HQ), hazard index (HI), and lifetime cancer risk (LCR), would be used to assess the health risks associated with consuming such water in the research area.
2.0 Materials and Methods

2.1 Study Area
The study was carried out in Tarkwa, which found in the Western Region of Ghana between Latitude 4°5’ and Longitude 5°5’ (Figure 1). AngloGold Ashanti, Goldfields Ghana Ltd, and Ghana Manganese Company are three of the most well-known large-scale mining companies in Tarkwa. Additionally, Tarkwa is home to a number of small-scale mining firms. Large portions of the population are employed by these businesses. Due to its location within Ghana's main gold belt, which stretches from Konongo in the northeast through Tarkwa to Axim in the southwest, mining is the area's main industrial activity (Armah, 2022). Gold and manganese are the two most important minerals mined in the study area.

Although cocoa and oil palm are also produced, the majority of the people is employed in subsistence farming. The Banket Series, which can be further divided into a footwall and hanging wall barren quartzite, dominates the local geology at Tarkwa and is separated by a series of mineralized conglomerates and pebbly quartzites (Figure 2). Auriferous reefs and barren juvenile quartzites are interbedded in the well-established stratigraphy of the different quartzite groups. The research area contains both the weathered zone aquifer and the fractured zone aquifer. The zone where fresh and weathered rocks converge is where the weathered aquifer is mostly located. Due to the presence of clay and silt, these aquifers have low permeability despite having high porosity and storage. The fractured zone is below the aquifer in the transition zone which is characterized by Low storage but great transmissivity (Kortatsi, 2004). The region's main source of water supply is groundwater. The bulk of the larger municipalities in the area only have access to groundwater as a supply of water. To meet the need for drinkable water, the number of hand-dug wells and boreholes keeps increasing quickly (Kortatsi 2004).
Figure 1: Map of Tarkwa Nsuaem municipality
2.2 Sample Collection

500 mL polypropylene bottles were used to randomly collect 74 groundwater samples (wells and boreholes) in Tarkwa between January and March 2020 for the dry season and May to July 2020 for the wet season. The boreholes and wells ranged in depth from 21m to 85m. Many of the wells were manually dug, open, and some were unusually close to highly polluted streams. This is
owing to the fact that the majority of the region's surface water has been contaminated by illegal mining. Samples were collected in triplicate at each sampling site for physiochemical, heavy metals, and cyanide analysis. Samples were transferred to the lab for examination in coolers with ice packs after being acidified for heavy metal analyses to pH 2 by adding 2ml of trace metal grade HNO₃.

2.3 Chemical analysis

2.3.1 pH and Conductivity

Approximately 30 mL of the sample was placed into a bottle and a combined pH and conductivity electrode TPS Smatchem multimeter calibrated with pH buffer 4 and 7 was used to measure the pH and conductivity of the samples.

2.3.2 Alkalinity

Utilizing a Metrohm Auto-titrator and TPS Multiparameter instrument, alkalinity was assessed titrimetrically. 50 mL of the sample was titrated with 0.02 M of HCl to pH 4.5 using methyl red as indicator to detect a color changed from yellow to orange. Equation 1 was used for calculating alkalinity.

\[
\text{Alkalinity as } \text{HCO}_3^- \text{ (mg/L)} = \frac{A \times N \times 50,000}{V} \quad (1)
\]

Where:

\[
A = \text{Volume of HCl used to titrate to pH 4.5 (mL)}; \quad N = \text{Normality of HCl};
\]

\[
V = \text{Volume of sample}.
\]
2.3.3 Turbidity

Turbidity of the samples was determined using a HACH Turbidimeter model 2100N. Before usage, the meter was calibrated using standard solutions of 2, 10, 50, and 200 NTU.

2.3.4 TDS

Gravimetric analysis was used to calculate the total dissolved solids. Filtrate from a 100 mL well-mixed sample was added to a beaker that had already been pre-weighed. After drying in an oven set to 100 °C, the samples were cooled in a desiccator, and the final weight was calculated. Equation 2 was used to calculate the TDS.

\[
\text{TDS (mg/L)} = \frac{(A-B) \times 1,000,000}{C}
\]

Where: \(A = \) weight of dried residue + beaker (g); \(B = \) weight of beaker (g); \(C = \) sample volume (mL)

2.3.5 Determination of Anions (Chloride, Nitrate, Phosphate and Sulphate)

The anions were determined using an Aquakem Discrete Analyzer Konelab 200. The technique employs colorimetric and photometric principles to measure the concentrations of anions in the samples. After calibrating the equipment with the necessary standards, the ions were measured. Certified reference materials, duplicates, and reagent blanks were used for quality control; those with recovery percentages greater than 90% were acceptable.

2.3.6 Total Recoverable Metals

According to the USEPA 3050B procedure (U.S. EPA. 1996), 50mL of water samples were digested with 2.5 mL of 1:1 HNO3 and 2.5 mL of 1:1 HCL. The concentration of Ca, Mg, Na, and K was determined directly from an aliquot of the digest using a Perkin Elmer Avio 500 ICPOES. The heavy metals (Cu, Cd, Cr, Pb, Hg, Ni, Zn, and Fe) were examined with Nexion 2000 ICPMS, which is outfitted with a quartz micro mist nebulizer and glass cyclonic spray chamber. Aliquots of the digests were also diluted with a diluent (2% v/v HNO3). The heavy metal analysis employed dwell periods of 60, 25, 100, 25, 50, 25 milliseconds for As, Cd Hg,
Pb, U, Zn, and Cr, respectively. All duplicate samples had a relative percentage difference less than 20%. Certified reference materials, duplicates, and reagent blanks were used for quality control; those with recovery percentages greater than 90% were acceptable.

2.3.7 Cyanide

The SKALAR Cyanide Analyzer was used to determine the concentration of cyanide in samples. In a reactor structure, this method combines UV digestion and distillation. Irradiation in a quartz coil breaks down strongly bound complex cyanides into free cyanide, which is then extracted from the sample matrix by distillation. Quartz is transparent to UV wavelengths from 200 to 400 nm; therefore, these wavelengths can pass through it. This makes it possible to degrade the strong metal cyanide complexes and thiocyanate. The instrument was calibrated with calibration standards followed by the analysis of samples. Certified reference materials, duplicates, and reagent blanks were used for quality control; those with recovery percentages greater than 90% were acceptable.

2.4 Health risk assessment

Human health risk has to do with estimating the possibility of negative health effects in humans exposed to toxicants in polluted settings. It entails assessing exposure, as well as noncarcinogenic and carcinogenic hazards. Owning to the behavioral and physiological variations between children and adults, the health risk assessment was done separately.

2.4.1 Exposure assessment

Estimated daily intake (EDI) was employed to calculate the exposure of humans to heavy metals and cyanide from direct ingesting using equation 3 adopted from US EPA methods (1992). Estimates were established for two distinct groups: children (as a sensitive group) and adults (as the general population).

\[
EDI = \frac{C \times IR \times EF \times ED}{BW \times AT}
\]  

Where EDI (mg/kg/day) is the estimated daily dosage consumption by ingestion, C is the concentration of metal (mg/kg) in the food, IR is the ingestion rate (kg/day), EF is the Exposure frequency, ED is the exposure duration, BW (Kg) is the Standard body weight and AT is the
time duration of human exposure. Table 1 shows the parameter used to calculate the expected daily intake.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>IR</td>
<td>0.2g/day for children and 0.1g/day for adults</td>
<td>USEPA, 2012</td>
</tr>
<tr>
<td>EF</td>
<td>180 days/year</td>
<td>USEPA, 2012</td>
</tr>
<tr>
<td>ED</td>
<td>6 years for children and 24 years for adults</td>
<td>USEPA, 2012</td>
</tr>
<tr>
<td>BW</td>
<td>70kg for adults and 15kg for children</td>
<td>USEPA, 2012</td>
</tr>
<tr>
<td>AT</td>
<td>365 *ED</td>
<td></td>
</tr>
</tbody>
</table>

2.5.2 Non carcinogenic risk

The Hazard quotient and Hazard index were used to assess noncarcinogenic health risk.

2.5.2.1 Hazard quotient

Equation 4 was used to determine the Hazard Quotient (HQ). “A hazard quotient is the ratio of the potential exposure to a substance and the level at which no adverse effects are expected” (USEPA, 1989). Table 2 shows the specific reference doses (RD) for each heavy metal and is adopted from (USEPA, 2012).

<table>
<thead>
<tr>
<th>Heavy Metal</th>
<th>Ref Dose</th>
<th>CSF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cd</td>
<td>0.001</td>
<td>0.0061</td>
</tr>
<tr>
<td>Cr</td>
<td>1.5</td>
<td>0.041</td>
</tr>
<tr>
<td>Pb</td>
<td>0.04</td>
<td>0.0085</td>
</tr>
<tr>
<td>Fe</td>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td>Zn</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Element</td>
<td>Concentration</td>
<td>Reference Value</td>
</tr>
<tr>
<td>---------</td>
<td>---------------</td>
<td>-----------------</td>
</tr>
<tr>
<td>Cu</td>
<td>0.04</td>
<td>0.00084</td>
</tr>
<tr>
<td>Ni</td>
<td>0.02</td>
<td>0.0003</td>
</tr>
<tr>
<td>As</td>
<td>0.0003</td>
<td>1.5</td>
</tr>
<tr>
<td>Hg</td>
<td>0.0001</td>
<td></td>
</tr>
</tbody>
</table>

\[ HQ = \frac{EDI}{RD} \]  \hspace{1cm} (4)

### 2.5.2.2 Hazard Index

A Hazard Index (HI) method was used to assess the total potential for non-carcinogenic consequences posed by more than one chemical (USEPA, 1989). To be able to estimate the HI for a mixture of chemicals, the hazard index of that mixture is calculated using equation 5 (USEPA, 1989)

\[ HI = \sum HQ \]  \hspace{1cm} (5)

“If the HI value is less than one, the exposed population is unlikely to experience obvious adverse health effects. If the HI value exceeds one, then adverse health effects may occur” (USEPA, 1989).

### 2.5.3 Carcinogenic risk assessment

Carcinogenic risks are calculated by assessing the likelihood of a person acquiring cancer as a result of exposure to a carcinogen throughout a lifetime. As illustrated in equation 6, the carcinogenic health risk is determined using a cancer slope factor. (USEPA, 1989):

\[ LCR = EDI \times SF \]  \hspace{1cm} (6)

Where LCR is the lifetime cancer risk and SF is the slope factor (mg/kg/day).

LCR above $1 \times 10^{-4}$ is considered unacceptable, risks below $1 \times 10^{-6}$ are known to have significant health effects, and risk lying between $1 \times 10^{-4}$ and $1 \times 10^{-6}$ is said to be in an acceptable range (USEPA, 1989).
2.6 Ground water chemistry

2.6.1 Gibbs Plot

The Gibbs plot was used to determine the link between water composition and the lithological characteristics. The Gibbs plot was obtained by plotting TDS against Cl−/(Cl− + HCO₃⁻) for anions and TDS against Na/Na+Ca for cations (Gibbs 1970).

2.6.2 Ionic dominance

The correlations between the major ions: (Ca²⁺+Mg²⁺); (Ca²⁺+Mg²⁺) vs (HCO₃⁻ + SO₄²⁻); (Ca²⁺ + Mg²⁺) vs (Cl⁻ vs SO₄²⁻) were used to investigate the mechanisms influencing groundwater chemistry and the sources of ions in groundwater in the study area.

2.7 Statistical Analysis

The mean and standard deviations were calculated using exploratory data analysis and the significant differences. Prior to statistical analysis, the Shapiro-Wilks test was employed to assess the normality of the data. The data were found to have a normal distribution. All plots and statistical analysis were done using SPSS 24.

3.0 RESULTS AND DISCUSSIONS

3.1 Physicochemical parameters of groundwater in Tarkwa

The nature, quality and type of water is mostly determined by the physicochemical properties of the water. The physicochemical parameters of 74 groundwater samples from Tarkwa were analyzed statistically and the descriptive statistics are presented in Table 3. There was a statistically significant difference between the physicochemical parameters (P=0.0001; P<0.05) indicating a great variability in data.

With a mean pH of 6.8, the pH was in the range of 4.9 to 7.7. The humic soil and the local acidic latrite are responsible for the acidic pH of the study area (Boateng et al., 2016). The mean pH was appropriate for use as drinking water because it was within the WHO permitted range of 6.5-8.5. However, waters with an acidic pH are not recommended for drinking because they may cause consumers to experience health issues including acidosis. In a similar vein, an acidic pH encourages the release of chemical pollutants like heavy metals. According to Bradl, 2004, at
lower pH there is low adsorption of heavy metals to organic constituents in aquatic systems resulting in the release on heavy metals bound to organic particles. This is a result of competition between H+ and heavy metals at the binding sites of organic materials. Similar to this, the pH adsorption edge is seen near neutral pH, which favors the release of heavy metals to some extent in comparison to higher pH (Bradl 2004). Thus, the pH of Tarkwa water encourages the release of heavy metals bound to water particle particles. The pH found in our study is consistent with research done by Boateng et al, 2016 in the Ejisu Juaben municipality and that of Duncan et al., 2016 in the cape coast municipality.

According to the electrical conductivity (EC), which ranged from 150 to 1080, some of the sampling spots exceeded the WHO threshold of 500 S/Cm. According to Radojevic and Bashkin (2006), the EC measurement measures the capacity of water samples to carry electric currents, which is related to the amount of ionized chemicals present in the water. According to the WHO (2011) and Radojevic and Bashkin (2006), greater EC values at some sampling locations signify the presence of inorganic salts and organic materials in high concentrations as dissolved ions. Since acidic pH facilitates the release of organic bound ions, the presence of significant dissolved salts supports the results of acidic pH obtained at some sample points. The mean EC recorded was 398.5 which was however below the WHO criteria of 500 µS/Cm.

Total dissolved solids (TDS) are composed of calcium, chlorides, and magnesium (Zhang et al., 2017). The TDS ranged from 21 to 873 mg/L. This suggests that certain sampling stations may have exceeded the WHO-recommended limit of 500 mg/L. However, the mean TDS value of 271 mg/L was below the WHO standards. TDS levels in bodies of water may be increased by human activities such agriculture, water use, industrial processes, and mining (2013) Caedo-Argüelles et al. The overall taste of water can be changed by a high TDS. Similar to this, TDS readings exceeding 500 ppm necessitate additional testing for harmful particles like heavy metals that can harm both people and aquatic organisms.

Groundwater from Tarkwa is perceived to be hard water as the mean total hardness was above 180mg/L (Diggs and Parker, 2009). The range of total hardness (TH), with a mean value of 323.2 mg/L, was 70.5-3358.8 mg/L. The measured concentration of divalent metal cations is
frequently used to characterize TH. The only two divalent cations that are found in considerable concentration in the majority of waters are calcium and magnesium (Diggs and Parker, 2009).

Natural waters frequently contain calcium and magnesium ions, both of which have high concentrations. Magnesium and calcium levels in Tarkwa water were observed to be between 1.9 and 395 mg/L and 3.8 to 693 mg/L, respectively. Calcium and magnesium are mostly derived from carbonate minerals such as calcite and dolomite (Deelman, 2021; Stanienda, 2016). These ions are released into the water as a result of ion exchange between the minerals, soil, and water. This is made easier by the medium's pH since at lower pHs, more ions are liberated than at higher pHs. However, the average Ca and Mg ion concentration fell within the WHO guidelines.

Sulfates and chlorides may originate from both natural and anthropogenic sources. Sulphates may originate from minerals like gypsum (CaSO2·2H2O) and barite (BaSO4) (Jeevanandam et al. 2006; Kumar et al. 2009). Results ranged from 10.5 to 95 mg/L for chlorides and from 10 to 855 mg/L for sulfates, respectively. Cl concentrations were below the WHO recommended level at all sample sites. Chloride in excess is frequently employed as a tracer for groundwater contamination and as a pollution indicator (Loizidou and Kapetanios 1993). According to Walker et al., 1991, anthropogenic sources and the solubility of Cl-bearing evaporation deposits are two common causes of high Cl ion concentration in groundwater. Water with a high chloride concentration has a salty taste and can lead to kidney stones, asthma, hypertension, and osteoporosis (McCarthy, 2004). Some sampling stations had sulphate concentrations that were higher than the WHO criteria. Higher sulphate concentrations may arise from sewage and industrial effluents (Zak et al., 2021). In the context of high Ca and Mg levels, excess sulphate might result in health issues such gastrointestinal discomfort (Suthar et al. 2009).

Nitrates and NO2-levels were within the permitted ranges of 4.5–760 mg/L and 1.2–30 mg/L, respectively. This suggests that several sampling locations had nitrate and nitrite concentrations that were higher than the WHO's standards of 3 mg/L and 45 mg/L, respectively. Nitrates are typically absent from deeper wells due to the reducing conditions (Hojberg et al., 2017). Therefore, nitrates can be used as an indicator of the interaction between groundwater and land use (Lee et al., 2020). High nitrate levels in groundwaters in Tarkwa are a sign of shallow
aquifers and anthropogenic intrusion. This can be attributed to the use of fertilizers and sewage pollution (Bijay-Singh and Craswell, 2021). High nitrate intake is linked to a number of health impacts, including gastric cancer and other effects on children and pregnant women (Rao, 2006). Nitrites are created when ammonium converts into nitrate during the nitrification process (Eletta et al. 2010). Nitrites can have a health impact that is orders of magnitude worse than nitrates. This is due to the fact that nitrites have the ability to oxidize hemoglobin to methemoglobin, which impairs the flow of oxygen throughout the body (Alsabahi et al. 2009; Chapman 1992).

Phosphate in water systems may originate from raw sewage, pipes rich in detergent or from agricultural fields (Akoto et al. 2010, Akpabli and Drah 2001; Sinha et al. 2000). Results for phosphate were within the range of 2.3 to 80 mg/L, and the mean value of 13.3 mg/L was greater than the WHO recommendation of 2.5 mg/L. Given Tarkwa's extensive agricultural lands, this is hardly unexpected. The high phosphate can thus be attributed to run off from agricultural fields as a result of the use of phosphate fertilizers.
Table 3: Physicochemical parameters of groundwater from Tarkwa

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Mean</th>
<th>Standard Deviation</th>
<th>Minimum</th>
<th>Maximum</th>
<th>WHO</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>6.8</td>
<td>0.7</td>
<td>4.9</td>
<td>7.7</td>
<td>6.5-8.5</td>
</tr>
<tr>
<td>Conductivity</td>
<td>398.5</td>
<td>291.6</td>
<td>150</td>
<td>1082.0</td>
<td>500.0</td>
</tr>
<tr>
<td>HCO₃⁻</td>
<td>109.5</td>
<td>92.4</td>
<td>0.1</td>
<td>353.7</td>
<td>500.0</td>
</tr>
<tr>
<td>Turbidity</td>
<td>24.4</td>
<td>28.5</td>
<td>1.4</td>
<td>139.5</td>
<td>&lt;5</td>
</tr>
<tr>
<td>TDS</td>
<td>271.7</td>
<td>208.2</td>
<td>21.0</td>
<td>873.0</td>
<td>500.0</td>
</tr>
<tr>
<td>TH</td>
<td>323.1</td>
<td>380.6</td>
<td>70.5</td>
<td>3358.8</td>
<td></td>
</tr>
<tr>
<td>Cl⁻</td>
<td>39.1</td>
<td>23.6</td>
<td>10.5</td>
<td>95.0</td>
<td>250.0</td>
</tr>
<tr>
<td>Ca²⁺</td>
<td>71.8</td>
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3.2 Hydrogeochemical process

Anionic dominance was in the order NO₃->SO₄²->HCO₃->Cl->PO₄->NO₂- and cationic dominance was in the order Ca²⁺ > Mg²⁺ > Na⁺. The interaction between the various primary ion concentrations was used to study the evolution of groundwater in the Tarkwa municipality. Studying the weathering processes in the basin is made possible by the chemistry of important ions in groundwater (Brijraj and Kaur 2007). Ca and Mg were the most dominant cations and bicarbonate, nitrates, and sulfates were the most dominant anions in Tarkwa's groundwater. Calcium and magnesium ratio below Two (2) indicate dissolution of dolomite and calcite and above 2 reflects the effect of silicate on groundwater chemistry (Figure 3a) (Boateng et al, 2006; Rajib et al., 2019). The majority of the calcium to magnesium ratio distribution is above the 2-threshold value, as shown by Figure 3a. This shows that silicate dissolution was responsible for the increase in Ca and Mg concentration in the water. Significant amounts were discovered below 2, though, which suggests that some calcite and dolomite weathering may also contribute to the ions in solution.

Also, the majority of the samples are also above the equiline in the scatter map of Ca+Mg vs HCO₃+SO₄ (Figure 3b), indicating that Ca and Mg ions predominate in Tarkwa's groundwater. If more points are below the median line, the dissolution of ions can be attributed to ion exchange (Nadeem et al., 2016; Cerling et al., 1989; Fisher and Mulican 1997). The fact that the majority of points emerged above the median line suggests that the ion release was not caused by simple ion exchange mechanism. However, a sizable amount below the equiline suggests that groundwater also contains a sizable number of ions due to ion exchange. In natural water, alkalis have primary bonding affinity with SO₄ and Cl (Nazzal et al., 2014). However, a poor correlation was found between Ca+Mg VS SO₄+Cl suggesting that not all alkalis are consumed by SO₄ and Cl (Figure 3c). This corroborating the fact that Ca and Mg ions are the dominating ions. Additionally, it suggests that groundwater in Tarkwa is contaminated by anthropogenic activities. It is not strange that calcite, dolomite, silicate dissolution and even anthropogenic sources favor the release of ions into groundwater in Tarkwa. This is due to the large number of illegal miners in Tarkwa who employ various chemicals to aid in the breakdown of minerals from diverse sources. The unfavorable outcome of these processes is the release of
heavy metals from minerals, which contaminates groundwater and renders it unfit for irrigation and drinking.
The relationship between water composition and lithological characteristics has frequently been examined using the Gibbs plot (Kumar et al., 2015; Madhav et al., 2018). TDS against Cl/(Cl− + HSO4−) is used to determine the Gibbs ratio for anions (Figure 4a), and TDS against Na/(Na+Ca) is used to calculate the Gibbs ratio for cations (Figure 4b). The Gibbs plot demonstrates that the primary processes controlling water chemistry are rock dominance and, to a lesser extent, precipitation dominance. The samples with a rock dominance are on the left side of the Gibbs diagram, have moderate TDS values, but a lower Na/(Na+Ca) and Cl/(Cl+HCO3) ratio (Qiying et al., 2020). The primary mechanism regulating the chemistry of groundwater is rock water interphase. The samples in the precipitation dominance fall on the lower right side of the Gibbs diagram; the TDS value is lower, while the values of Na/(Na+Ca) and Cl/(Cl+HCO3) are higher,
indicating that atmospheric precipitation is a factor influencing the chemical composition of groundwater (Qiying et al., 2020).

Figure 4: Gibbs plot for anions (a) and Cations (b)
3.4 Chemical parameters of groundwater in Tarkwa

The descriptive statistics of heavy metals and CN are presented in Table 4. Results were in the range of 1.6-16, 0.0001-0.015, 0.0001-0.005, 0.009, 0.001-0.009, 0.001-0.008, 0.0001-0.01, 0.004-1.375, 0.002-0.118, 0.001-0.006, 0.001-0.145 for CN, As, Hg, Cd, Cu, Cr, Pb, Mn, Ni, Zn and Co, respectively. Some Hg, As, Cd, Mn, and CN sample locations exceeded their corresponding WHO standards. This is consistent with studies by Li et al. (2018), Hadzi et al. (2018), and Rama et al. (2021), which found high amounts of heavy metals in mining sites. Environmental pollution has resulted from illegal mining activities in Tarkwa, including the use of CN and Hg. Numerous studies have revealed that consuming water that contains a lot of heavy metals is harmful to one's health. High-density lipoprotein (HDL) levels rising and immune system function deteriorating have both been linked to zinc (Chasapis et al., 2012). At extremely high concentration, Ni could cause abdominal pain, an increase in red blood cells, and a decline in lung function (Zambelli et al., 2016). High Pb concentrations are linked to health issues such behavioral issues and elevated arterial pressure (Esmaeilzadeh et al., 2019; Vasseghian et al., 2020). Extreme Cd exposure can also result in bone problems and other health problems. Cd exposure is extremely harmful and can cause cancer, according to numerous research (Vasseghian et al., 2020; Jarup et al., 2000).

Table 4: Concentration of heavy metals and CN in groundwater from Tarkwa

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<tr>
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<th>Mean (mg/L)</th>
<th>Standard Error</th>
<th>Minimum (mg/L)</th>
<th>Maximum (mg/L)</th>
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<td>0.015</td>
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<tr>
<td>Hg</td>
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<td>0.0001</td>
<td>0.0001</td>
<td>0.005</td>
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<tr>
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3.5 Correlation of parameters

A high correlation was observed between Ca, Mg and TDS and SO$_4$ and TDS which stipulates the dominance of these ions in groundwater in Tarkwa (Table 5). Similarly, a high correlation was observed between Ca and Mg which indicates that they originate from similar sources. These sources derive from the breakdown of silicates, calcite, and dolomite in the rock water interphase (Deelman, 2021; Stanienda, 2016). The presence of ions in solution may be altered by an ion exchange mechanism, as evidenced by the strong association found between SO$_4$ and Ca (Haldar, 2020). Furthermore, a strong association (0.564) between nitrate and sulphate was found. According to several studies, common sources of excess nitrates and phosphates in groundwater include septic systems, agricultural fertilizers, animal feed and manure, residential sewage, industrial waste waters, sanitary landfills, and garbage dumps (Zhang and Qiu, 2019; Sun et al., 2019; Wand et al., 2020; Lu et al., 2019). Nitrate and phosphate had moderate correlations with Cu and Cd, indicating that certain heavy metals originate from anthropogenic sources such as residues from fertilizer application (Zhang and Qiu; Wand et al., 2020).

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### Table 5: Correlation of parameters

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</table>
3.6 Non carcinogenic health risk: Estimated daily intake (EDI) and Hazard quotient (HQ) and Lifetime cancer risk (LCR)

Table 6 shows the estimated daily intake of CN and heavy metals in groundwater in Tarkwa. The estimated daily intake evaluates the amount of chemical pollutants that are consumed on a daily basis (Chamannejadian et al., 2013). The results for adults and children ranged from 0.34 to 2.78 and 0.00-0.003 for CN, 0.00-0.003 and 0.00-0.001 for As, 0.00-0.001 and 0.00-0.002 for Hg, 0.00-0.002 and 0.00-0.003 for Cd, and 0.00-0.002 and 0.00-0.003 for Pb respectively. The results for Cd and Pb were within their acceptable daily limit. Hg and As, however, exceeded the advised daily limit. Numerous publications have noted high arsenic concentrations in drinking water sources in mining sites (Wurl et al., 2018; Chandio et al., 2021). Additionally, it has been reported that illegal miners in Ghana and the Tarkwa municipality used mercury (Hg) (Saim, 2021). Due to the danger mercury poses to human health, numerous intervention strategies have been developed, including the Minamata Convention on Mercury, which many governments, including Ghana, have ratified. Long-term exposure to Hg and As is not recommended because they may have a negative impact on health.

The hazard quotient as a result of exposure to heavy metals are presented in Figure 5-10 for CN, As, Hg Cd and Pb, respectively. The hazard quotient for CN for all sampling points for children, as well as for sampling point 26 for both adults, was above the recommended guideline of 1, indicating noncancerous health risk are associated with water consumption within the Tarkwa municipality. Cd for sampling point 44-72 for both adults and children were also above the recommended guideline of 1. According to studies by Kwaanssa-Ansah et al. (2017) and Hendry-Hofer et al. (2019), CN is one of the most dangerous compounds. Small doses of CN may result in headaches, weakness, nausea, and vomiting, while large doses may result in panting, irregular heartbeat, seizures, fainting, and even quick death (Arbabi et al., 2015). However, the hazard quotients for Pb and Hg were below the advised level. As a result of the high concentration of As noted, an LCR was additionally performed (Fig. 9). According to the LCR, drinking water for an extended period of time in Tarkwa posed a danger for cancer. Children were discovered to be at higher risk for both carcinogenic and non-carcinogenic health risks.
Table 6: Estimated daily intake (mg/kg/day)

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<td>74</td>
<td>0.0003</td>
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<td>Hg Adult</td>
<td>0.0001</td>
<td>0.0001</td>
<td>1.71E-05</td>
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<td>74</td>
<td>0.0001</td>
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<td>Hg Child</td>
<td>0.0002</td>
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<td>2.67E-05</td>
<td>0.002</td>
<td>74</td>
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<tr>
<td>Cd adult</td>
<td>0.0003</td>
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<td>4.29E-06</td>
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<td>74</td>
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<td>Cd child</td>
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<td>6.67E-06</td>
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<td>74</td>
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<tr>
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Figure 5: Hazard quotient for CN in groundwater from Tarkwa

Figure 6: Hazard quotient for Arsenic in groundwater from Tarkwa
Figure 7: Hazard quotient for mercury in groundwater from Tarkwa

Figure 8: Hazard quotient for Cadmium in groundwater from Tarkwa
Figure 9: Hazard quotient for Lead in groundwater from Tarkwa

Figure 10: Lifetime cancer risk for arsenic in for adults and children
CONCLUSION

In Tarkwa, a mining settlement in Ghana, the hydrogeochemical mechanisms that influence groundwater chemistry and chemical pollutants in groundwater were assessed. The findings indicated that the primary mechanisms regulating the chemistry of the water are rock dominance. As a result, interaction between the water and rock interphase, notably Silicates, ultimately responsible for the presence of ions in solution that alter water quality. The discharge of ions from the rock interphase is facilitated by unlawful mining practices that use chemicals like Hg and CN.

Anthropogenic causes of contamination were also found. The presence of contaminants in groundwater was indicated by the parameters pH, Conductivity, Turbidity, Magnesium, Nitrate, Phosphate, Sulfate, CN, As, Hg, Cd, Mn, and Ni, which were all above their WHO recommended thresholds. The amount of nitrates was higher than the WHO-recommended threshold, which is an indication of a shallow aquifer that receives runoff from agricultural fields. Elevated concentrations of CN, As, Cd, and Hg were discovered, and this was ascribed to the illegal mining activities within the municipality. Due to high levels of CN, As, and Cd, prolonged usage of water from the Tarkwa municipality may pose a both carcinogenic and noncarcinogenic health risk. Children were shown to be more vulnerable to both carcinogenic and noncarcinogenic health risk than adults.

Legalization of mining activities is recommended to regulate the activities of miners within the community. Enforcement of the Hg act will help reduce the use of Hg and minimize their impact on the environment. Also, education on the proper chemical handling and efficient mining methods will help reduce the presence of chemical such as Hg and CN in the environment.
Declarations

Ethics approval and consent to participate: Not applicable.

Availability of data and material: The authors confirm that the data supporting the findings of this study are available within the article.

Competing interests: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Authors' contributions: Richard Osae and Harry Amoono Gwira conceived of the presented idea. All authors discussed the analytical methods required to complete the research and were involved in the sampling process. The analytical test was performed by Christopher Abasiya, Michael Peasah and Seyram Loh. Map of the study area was developed by Seyram Loh. Statistical analysis was performed by Richard Osae, Felix Owusu and Harry Amoono Gwira and all authors discussed the results and contributed to the manuscript. The manuscript was written by Richard Osae, Felix Owusu and Harry Amoono and the final manuscript was discussed by all authors.
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