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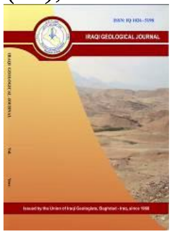

# Geochemical Characteristics and Evaluation of the Groundwater and Surface Water in Limestone Mining Area around Gunungkidul Regency, Indonesia

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## Abstract

Ten groundwater and six surface water samples were collected near the limestone mining region in Semin, Gunungkidul Regency, Daerah Istimewa Yogyakarta Province, Indonesia. This study is to investigate water characteristics using the geochemical method and to evaluate the groundwater and surface water for drinking and irrigation purposes in the study area. The samples were collected in 1000 ml polyethylene bottles. Prior to filling with sampling water, these bottles were rinsed to limit the chance of contamination. The hydrochemical characteristics of groundwater and surface water were determined using a Piper diagram. Groundwater samples were evaluated based on the Indonesia Minister of Health Regulation No: 492/ Menkes/Per/IV/2010 about the Requirements of Drinking Water Quality, while for irrigation water standards, surface water samples were evaluated based on the Government Regulation of the Republik Indonesia Number 82-year 2001 about Water Quality Management and Water Quality Control. All water samples are classified as $CaHCO_3$ hydrochemical facies, except SMN-07 as CaCl hydrochemical facies. The element Ca is assumed to arise from the limestone dissolution of the Wonosari-Punung Formation in the $CaHCO_3$ hydrochemical facies, whereas the $HCO_3$ element is thought to derive from the dissolution of calcite or aragonite, which is usually abundant in limestone. Meanwhile, the element Cl in one CaCl hydrochemical facies sample could come from a variety of sources, including weathering of soil and rocks, industrial and home waste discharge, seawater intrusion, wastewater discharge, rainfall, and irrigation return flow. The surface water quality in the study area is suitable for irrigation. The quality of groundwater in the study area, although most of them are acceptable drinking water quality standards, there are several locations that do not accept the standards for drinking water quality, especially pH and Total Dissolved Solids.

**Keywords:** Hydrochemical facies; Piper Diagram; Water drinking quality; Irrigation water quality; Water contamination

## 1. Introduction

In recent years, assessing water quality in order to manage any region has become a necessary study. It is also useful for determining the chemical composition of water, preventing further

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deterioration of its quality, and avoiding future pollution for irrigation and drinking water (Hamzaoui-Azaza et al., 2020). Chemical and physical criteria are used to assess water quality, which is influenced by hazardous materials, dissolved inorganic contaminants, natural organic compounds, sediments, and bacteria that have been collected over time (Aravinthasamy et al., 2020). Both groundwater and surface water play critical roles in determining people's quality of life and society's long-term viability. Due to global population growth, groundwater exploitation for daily needs, industrial, and irrigation purposes is quickly increasing. It has resulted in the depletion of groundwater sources, difficulties with water quality, and increased withdrawal costs. Natural and anthropogenic activities have an impact on groundwater quality, and recovering the original quality of previously contaminated groundwater is difficult (Karunanidhi et al., 2020). Different anthropogenic influences have severely polluted groundwater, and continuous consumption has greatly exacerbated the health concerns. Vomiting, diarrhea, stomachache, skin rashes, and other symptoms of polluted drinking water can occur quickly. Long-term consequences could include malignancy, such as leukemia (Marghade, 2020). In an unprecedented climate change scenario, natural processes involved in water dynamics and quality include natural subsidence, surface topography, marine storms, saltwater intrusion, and water-rock (sediment) interaction processes (Greggio et al., 2020). Septic tanks, organic and inorganic waste disposal, on the other hand, are the most common anthropogenic sources of groundwater pollution (Naik et al., 2021). It is critical to examine the water quality because natural and anthropogenic influences can pollute the aquatic environment (Asih et al., 2022). Gunungkidul is a regency of Indonesia's Daerah Istimewa Yogyakarta (DIY) Province. Gunungsewu Geopark, one of the oldest tropical karst landscapes lasting for around 120 kilometers, is located in the eastern section of the province and contains a variety of geographical landscapes ranging from coastal regions, hills, and hilly locations. Massive limestone mining was explicitly prohibited throughout the regency in 2011 under the Gunungkidul Regency's legislation concerning “Spatial Planning and Territory Gunungkidul District year 2010 to 2030” due to indicators of environmental degradation, notably in the karst area (Rahayu, 2018). Mining has an impact on groundwater discharge and results in changes in groundwater quality. Quarrying is the most common method of mineral extraction, and this practice has resulted in a large number of pits that have been abandoned without adequate closure and or rehabilitation. Because they are filled with rainwater during the rainy season, these pits pose significant environmental dangers and it flows around the residence wells, where it then becomes a supply of water for the surrounding community for whatever uses, especially during the dry season (Eyankware et al., 2018). Groundwater around the mining areas is the principal source of domestic water. It is vital to identify the water quality condition and characteristics of groundwater chemical evolution as a result of mining (Wang et al., 2019). Groundwater quality is critical to the security and preservation of groundwater. As a result, assessing the quality of groundwater is critical not only for current consumption but also for future consumption (Madhav et al., 2018). The main reasons for continuing to undertake studies all over the world are the exploitation and deterioration of groundwater (Beg et al., 2021). The groundwater geochemical facies are crucial in determining the primary factors and dominating sources of many metrics, water types, and processes that influence water chemical properties. Surface water percolates into aquifers and is hence referred to as groundwater, and its geochemical facies aids in determining how water quality changes over time (Kumar et al., 2020). As a result, the study of groundwater geochemical properties, geochemical processes involved, and their evolution in natural water circulation processes is beneficial not only to the optimal use and protection of this resource but also to the planning of groundwater environment improvements (Zamroni et al., 2021). Therefore, the objective of this study is to investigate water characteristics using the geochemical method and to evaluate the groundwater and surface water for drinking and irrigation purposes in Gunungkidul Regency, Indonesia.

**2. Geology and Hydrogeology of the Study Area**

The study area is located in the Surakarta-Giritontro Quadrangle of Java's regional geological map (Fig.1.). Mandalika Formation, Semilir Formation, Wonosari-Punung Formation, and Alluvium Formation are the exposed formations in the research area, from oldest to youngest (Surono et al., 1992). Magmatic activity lasting from the late Oligocene to the early Miocene resulted in the Mandalika Formation, which is made up of andesite, andesite breccias, and basalt, tuff. Breccia and lava were also deposited, with a large area of coverage (Idrus et al., 2021; Susilo et al., 2021). The Semilir Formation was developed in a marine environment that gradually transformed into land during the Early-Middle Miocene. It consists of tuff, tuffaceous sandstone, dacitic pumice breccia, and shale. It was formed by an old volcano in the DIY Province's southern region. It is made of relatively soft rock that has experienced a lot of weathering and erosion (Surono et al., 1992; Saputra et al., 2016; Pambudi et al., 2018). The Wonosari-Punung Formation was developed in a shallow marine environment during the Middle Miocene-Pliocene period. Limestone, conglomeratic limestone, siltstone, tuffaceous sandstone, and marly-tuffaceous limestone make up this formation (Surono et al., 1992; Atmoko et al., 2018; Pambudi et al. 2018). Along the river, alluvium is made up of clay, silt, sand, and gravel, which is created by denudation processes on steep and very steep terrain (Saputra et al., 2016).

Gunungkidul Regency is divided into three zones based on topographic, morphological, and hydrological characteristics, namely the northern zone (Batur Agung Zone), the middle zone (Ledok Wonosari Zone), and the southern zone (Gunungsewu Zone). The Batur Agung Zone is the first zone. On the north, it has mountainous terrain, while on the west, it has steep slopes. Because of the abundant water supply in the Batur Agung zone, various types of perennial and food crops can thrive. This zone includes the districts of Nglipar, Patuk, Gedangsari, Semin, Ngawen, and the northern part of Ponjong. Ledok Wonosari Zone is the second zone. It is located in Gunungkidul Regency's central region, with a flat to slightly undulating topography and a comparatively deep layer of soil with higher fertility than other zones. This zone includes the districts of Wonosari, Playen, Karangmojo, central Ponjong, and northern Semanu. Gunung Sewu is the third zone. Gunungsewu zone is located in Gunungkidul Regency's southern section, with a mountainous topography that leaves no landscape with a relatively narrow area. Dry ground moors predominate, with thin soil solum and nutrient-poor soil. Furthermore, there are very few water sources in this area. Drought calamities strike this region on a regular basis (Srihartanto and Widodo, 2020). The karst area of Gunungkidul Regency contains a fertile aquifer system, as evidenced by groundwater drilled wells utilized for irrigation or domestic reasons. The quantity of groundwater in Gunungkidul Regency varies by location, ranging from a medium potential for domestic use and a bad potential for irrigation to an exceptional potential for both irrigation and purposes (Purwanto et al., 2020).

Panggang, Bribin-Baron, Ponjong, Donoharjo-Pringkuku, Pracimantoro-Wuryantoro, and Sadeng Subsystems are the six primary hydrogeological subsystems in Gunungkidul Regency. The Panggang Subsystem is one of the hydrogeology blocks with major water resource issues, specifically a scarcity of clean water. Aside from the lack of a significant subsurface system, the paucity of water resources is due to the hydrogeological conditions of the area, which are characterized by small-discharge springs that are sparsely distributed along the limestone formation's outer edge. As a result, the central Panggang Subsystem has limited water resources. This hydrogeological subsystem features a fairly productive aquifer with fissure, fracture zone, and solution channel groundwater flow (Riyanto et al., 2019). The Gunungkidul Regency Regional Government, through its local water company (PDAM) Tirta Handayani, meets the local demand for clean water by pumping raw water from four underground rivers, namely the Seropan, Bribin, Baron, and Ngobaran (Bribin-Baron system), and constructing distribution networks. The Bribin-Baron subterranean system, which has a considerable flow discharge throughout the year, is the principal river system in the Gunungsewu Karst Area. PDAM has the ability to deliver 2,945 liters per second of raw water, with 1,000 liters per second from Baron, 950 liters per second from Seropan, 875 liters per second from Bribin, and 120 liters per second from Ngobaran. However, these values do not account for the maximum amount of extractable capacity. PDAM Tirta Handayani currently serves 50.32 percent of Gunungkidul's total population and 69.51 percent of the entire population in the service area. From 34,890 in 2013 to 47,325 in 2018, the number of homes connected to the pipeline has increased year after year (Widyastuti et al., 2020).

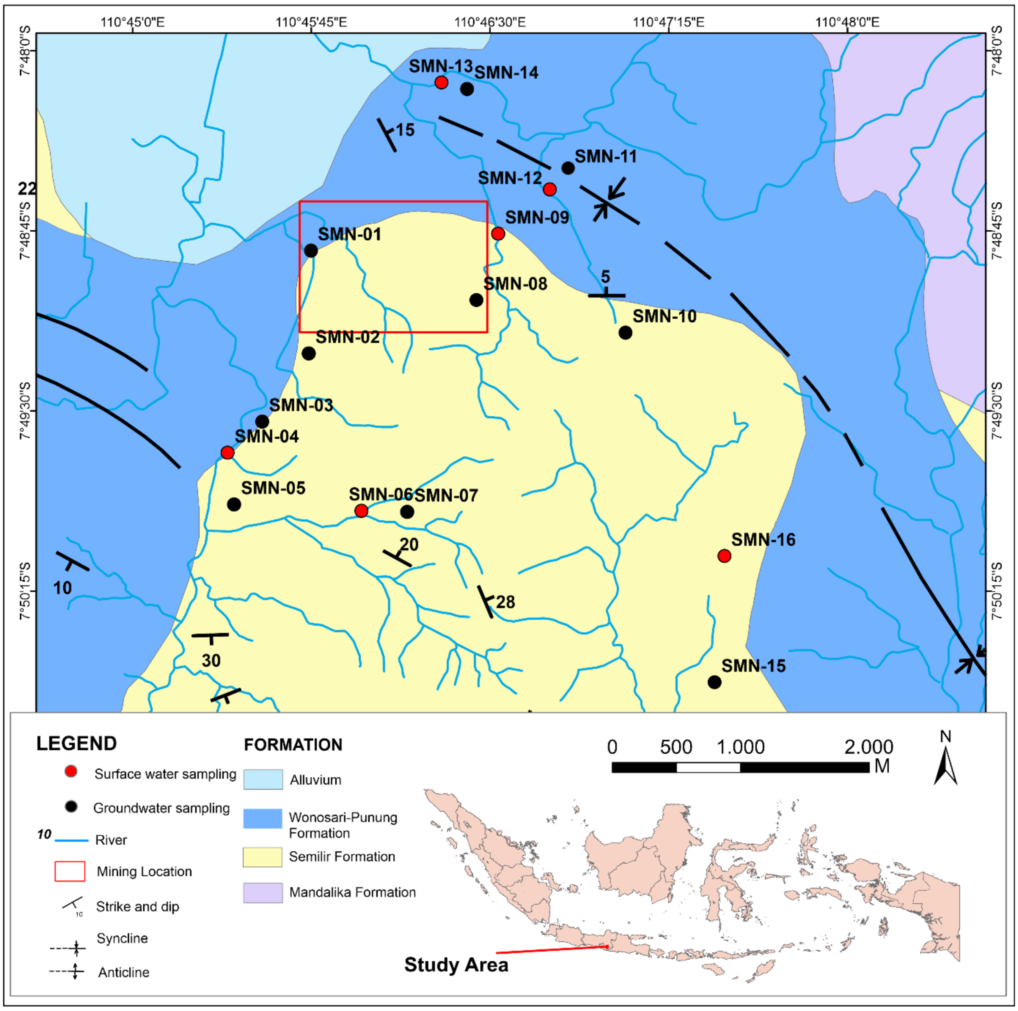


**Fig.1.** Geological map of the study area (Modified from Surono et al., 1992)

## 3. Materials and Methods

On June 20, 2021 (drought season), ten groundwater and six surface water samples were collected near the limestone mining region in Semin District, Gunungkidul Regency, Daerah Istimewa Yogyakarta Province, Indonesia (Fig. 2.). The study area is a limestone mining area where local communities use rudimentary mining tools to extract limestone. Mining operations have been ongoing since the 1990s. The limestone mining area is around 1.5 square kilometers in size. Surface water samples were taken from rivers near the mining region, while groundwater samples were acquired from household wells. The samples were collected in 1000 ml polyethylene bottles. Prior to filling with sampling water, these bottles were rinsed to limit the chance of contamination.

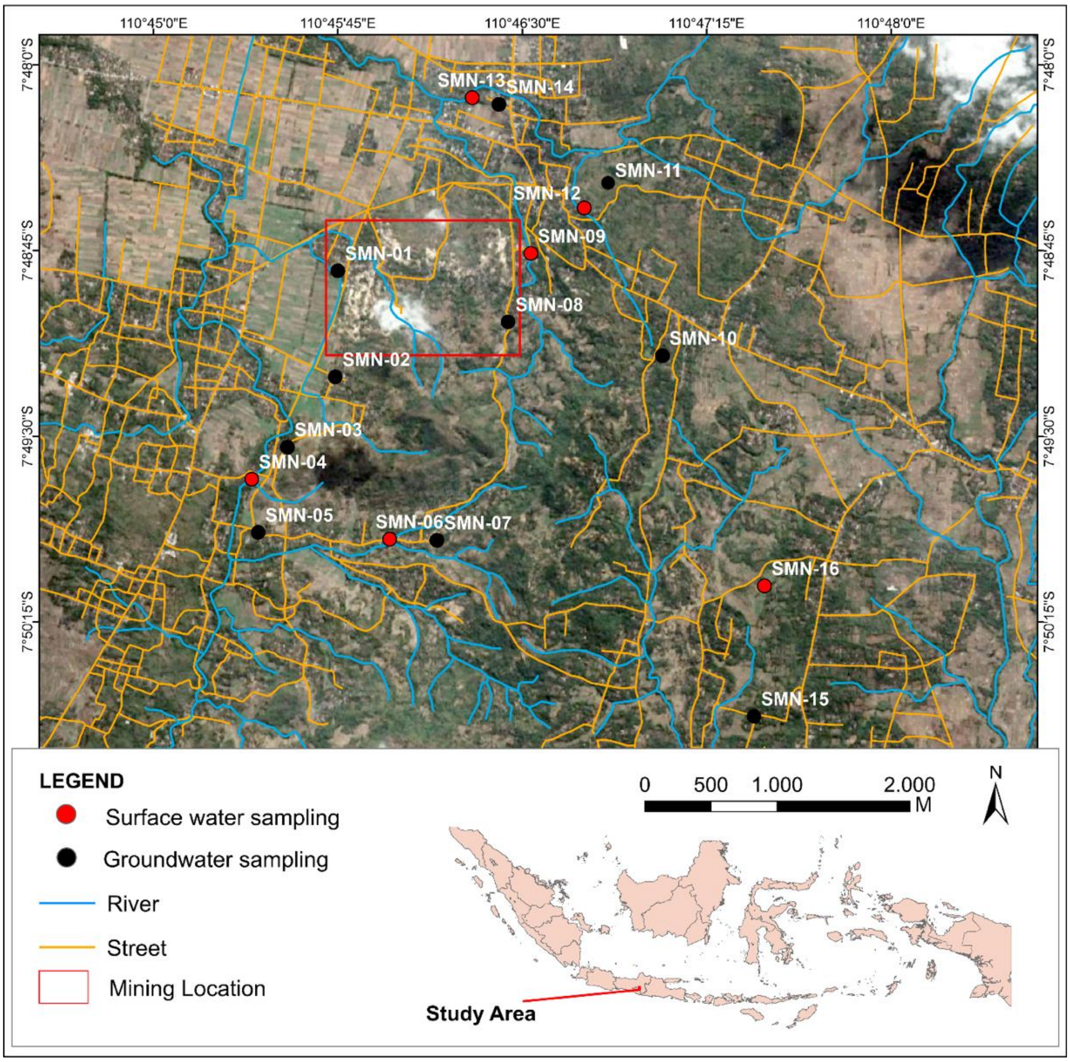


**Fig.2.** The satellite image shows the location of the study area and the groundwater and surface water sampling sites

The main ions: $Ca^{2+}$, $Na^{+}$, $Mg^{2+}$, $K^{+}$, $SO_4^{2-}$, $Cl^{-}$, $HCO_3^{-}$, and $NO_3^{-}$ were analysed, Total Dissolved Solids (TDS) meter and Electrical Conductivity (EC) were measured in situ. Sodium ($Na^{+}$), magnesium ($Mg^{2+}$), potassium ($K^{+}$), calcium ($Ca^{2+}$), bicarbonate ($HCO_3^{-}$), and chloride ($Cl^{-}$) ions were measured using the titrimetric method. The ions sulfate ($SO_4^{2-}$) and nitrate ($NO_3^{-}$) were detected using a spectrophotometer. A pH meter was used to determine the pH. TDS was measured with a TDS meter. EC was determined using a conductivity meter. The hydrochemical characteristics of groundwater and surface water were determined using a Piper diagram (Soleimani et al., 2018). Groundwater samples were evaluated based on the Indonesia Minister of Health Regulation No: 492/ Menkes/Per/IV/2010 about the Requirements of Drinking Water Quality (Indonesian Ministry of Health, 2010), while for irrigation water standards, surface water samples were evaluated based on the Government Regulation of the Republik Indonesia Number 82 year 2001 about Water Quality Management and Water Quality Control (State Secretary of the Indonesian Republic, 2001).

## 4. Results and Discussion

The findings of the geochemical investigation of groundwater and surface water in the research region are shown in Table 1. The findings of the chemical analysis were checked for ion-balancing faults

(≤20%). Piper's trilinear diagram (Piper, 1944) was created to assess geochemical sources and the connection between the research area's underlying lithology and water chemistry. According to Fig.3., in the study area, the interpretation of Piper's trilinear diagram for groundwater and surface water revealed that Ca, no dominant type, $HCO_3$, and $SO_4$ are the predominant cations and anions, therefore, all water samples are classified as CaHCO3 hydrochemical facies, except SMN-07 as CaCl hydrochemical facies. The concentration of bicarbonate in the studied area is higher than that of other main anions. Three theories have been offered to explain the high bicarbonate content. First, high-bicarbonate water is produced by the dissolving of calcite and dolomite. Second, the high bicarbonate level is due to hydrogeological influences in the area. Finally, the high partial pressure of $CO_2$ indicates that the carbonate system is open (Liu et al., 2019). The study area of the hydrogeological condition is spring with a small discharge but ample water availability and is sparsely distributed along the limestone formation's outer edge (Riyanto et al., 2019; Srihartanto and Widodo, 2020). $CaHCO_3$ and carbonate behave differently depending on the amount of free $CO_2$ in the water. The amount of $CO_2$ that should be present in water to keep the bicarbonate species in solution is referred to as $CO_2$ in equilibrium. When $CO_2$ levels are high, the concentration of bicarbonate rises to maintain equilibrium (Alvarez-Bastida et al., 208). Furthermore, recent geological processes such as rock weathering and water coming into contact with rocks might have an impact on the hydrochemical facies in the research area (Zamroni et al., 2020). The element Ca is assumed to arise from the limestone dissolution of the Wonosari-Punung Formation in the $CaHCO_3$ hydrochemical facies, whereas the $HCO_3$ element is thought to derive from the dissolution of calcite or aragonite, which is usually abundant in limestone. The Wonosari-Punung Formation, among numerous formations in the study area, is the only one that contains limestone. Meanwhile, the element Cl in one CaCl hydrochemical facies sample could come from a variety of sources, including weathering of soil and rocks, industrial and home waste discharge, seawater intrusion, wastewater discharge, rainfall, and irrigation return flow (Hamzaoui-Azaza et al., 2020).

The pH of the groundwater samples varies from 6.2 to 9.8. According to the Indonesian Minister of Health Regulation No: 492/ Menkes/Per/IV/2010, the permitted pH range is 6.5-8.5. Most places are within legal drinking water quality limits (SMN-01, SMN-02, SMN-03, SMN-05, SMN-06, SMN-08, SMN-10, SMN-12, and SMN-14), but a handful is not (SMN-07 and SMN-15). Surface water samples have pH levels that vary from 7.2 to 7.9. According to the Government Regulation of the Republik Indonesia Number 82 year 2001, the allowable pH level limit is 5-9. All of the surface water samples were found to be safe to use as irrigation water. The presence of carbonic acid, which is formed by $CO_2$ and $HCO_3$ in water and changes the pH level in the groundwater, indicates that the nature of groundwater and surface water is alkaline. The basicity is also attributable to the quarrying of limestone and limestone rock dissolution (Madasamy et al., 2021). It is critical to emphasize the mechanisms underlying the events going place in this domain. The resultant solution has a pH of 8.3, which is close to the pKa of the weak acid bicarbonate ion $HCO_3$ (pKa = 8.4) when water is in equilibrium with both atmospheric $CO_2$ and carbonate-containing rock. Any additional acid or base will not cause further pH shifts. This is a natural process for balancing significant lime dissolution from the underlying formation with lime precipitation from the water body (Eyankware et al., 2018). The high pH value in groundwater and surface water could be related to the location's high dissolving rate (Aziz et al., 2008). The EC values in the groundwater samples range from 108 to 1223 μS/cm, whereas TDS levels are between 54.5 and 602.7 mg/L. TDS has an allowed level of 500 mg/L, according to the Indonesian Minister of Health Regulation No: 492/ Menkes/Per/IV/2010. Most locations are within allowed limits for drinking water quality (SMN-01, SMN-03, SMN-07, SMN-08, SMN-10, SMN-12, SMN-14, and SMN-15), but a few are not (SMN-02 and SMN-05). Surface water samples include EC values ranging from 301 to 566 μS/cm and TDS values ranging from 148 to 281.5 mg/L. TDS has an acceptable level of 2000 mg/L, according to the Government Regulation of the Republik Indonesia Number 82 year 2001. All of the surface water samples were found to be safe to use as irrigation water. TDS levels are associated with EC levels. This means that places with high EC values are also areas with high TDS concentrations. The lower the EC value, the less water is available for plants to use (Eyankware et al., 2018). The substantially wider range of TDS values suggests that geohydrological factors had a stronger influence on mine water TDS (Shidong et al., 2021). TDS can also be found in runoff, fertilizers, and inorganic minerals containing calcium bicarbonate, nitrogen, iron phosphorus, sulfur, and other elements (Al-Dabbas et al. 2016).

The concentrations of $SO_4$ in groundwater samples range from 12 to 184 mg/L. According to the Indonesian Minister of Health Regulation No: 492/ Menkes/Per/IV/2010, the allowable $SO_4$ limit is 250 mg/L. The quality of the drinking water is acceptable at all places. Meanwhile, according to Government Regulation Number 82 of the Republik Indonesia for irrigation water, there is no $SO_4$ quality standard limit. $SO_4$ is mostly derived by the breakdown of evaporation rocks such as gypsum and anhydrite, or from the entry of runoff water enriched in soluble sulfate from the surface soil (Al-Dabbas et al., 2016). A high $SO_4$ content, particularly in wells near the end of the natural groundwater flow system, is influenced by natural hydrogeochemical evolution from $HCO_3$ in the recharge zone to $SO_4$ along a longer path, and finally to Cl groundwater (Hamzaoui-Azaza et al., 2020).The concentrations of $NO_3$ in groundwater samples range from 0.01 to 8.14 mg/L. According to Indonesian Minister of Health Regulation No: 492/ Menkes/Per/IV/2010, the allowable limit of $NO_3$ is 50 mg/L. The quality of the drinking water is acceptable at all places. Meanwhile, according to Government Regulation Number 82 of the Republik Indonesia for irrigation water, there is no quality standard limit for $NO_3$. $NO_3$ is a stable form of combined nitrogen and oxygenation system that is particularly water-soluble. As a result, it can easily pass through the soil and into the aquifer unit. However, there is an essential control in nature when groundwater goes below a confining unit, reducing availability to oxygen ions and lowering $NO_3$ concentrations (Hamzaoui-Azaza et al., 2020). Due to the usage of fertilizers by farmers, $NO_3$ is one of the most complex pollutants to both water and soil from a health standpoint (Al-Dabbas et al., 2016).

**Table 1.** The geochemical data of groundwater and surface water in the study area, concentrations in Mg/l

| Location | Sample Type | Depth (m) | pH | Ec | TDS (Mg/l) | Ca | K | Mg | Na | Cl | $SO_4$ | $HCO_3$ | $NO_3$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| SMN-01 | GW | 4.95 | 7.3 | 810 | 407 | 114.17 | 1 | 18.56 | 30 | 37.2 | 72 | 396 | 0.95 |
| SMN-02 | GW | 2.6 | 6.9 | 834 | 514 | 94.87 | <1 | 33.22 | 39 | 84.9 | 34 | 415.4 | 2.16 |
| SMN-03 | GW | 1.1 | 6.9 | 579 | 291 | 51.46 | 3 | 30.28 | 39 | 20.8 | 41 | 344.9 | 1.74 |
| SMN-04 | SW | 0 | 7.8 | 566 | 281.5 | 77.18 | 2 | 27.35 | 25 | 8.5 | 118 | 370.5 | 0.14 |
| SMN-05 | GW | 1 | 7.0 | 1223 | 602.7 | 86.03 | 2 | 6.35 | 72 | 86.5 | 89 | 402 | 0.52 |
| SMN-06 | SW | 0 | 7.9 | 402 | 199.5 | 40.2 | 1 | 21.98 | 30 | 10.5 | 52 | 281 | 0.54 |
| SMN-07 | GW | 0.65 | 6.2 | 502 | 260 | 44.22 | 1 | 27.84 | 28 | 25 | 184 | 83 | <0.01 |
| SMN-08 | GW | 2.55 | 6.7 | 533 | 259 | 55.07 | 2 | 28.33 | 28 | 18.5 | 21 | 198 | 8.14 |
| SMN-09 | SW | 0 | 7.6 | 530 | 261.5 | 66.73 | 2 | 15.63 | 25 | 10 | 63 | 293.8 | 0.07 |
| SMN-10 | GW | 1.3 | 7.1 | 900 | 452.5 | 140.7 | 2 | 13.19 | 30 | 30.5 | 31 | 463.1 | 0.16 |
| SMN-11 | SW | 0 | 7.5 | 446 | 218 | 61.1 | 1 | 10.25 | 14 | 9.5 | 18 | 217.2 | 0.44 |
| SMN-12 | GW | 3.6 | 7.0 | 469 | 233 | 84.42 | 1 | 10.26 | 14 | 9 | 18 | 249.1 | 0.67 |
| SMN-13 | SW | 0 | 7.2 | 492 | 248.5 | 75.58 | 3 | 15.14 | 22 | 9.5 | 18 | 293.8 | 0.13 |
| SMN-14 | GW | 3.4 | 7.0 | 600 | 300 | 96.48 | 1 | 17.59 | 19 | 13 | 25 | 287.4 | 1.33 |
| SMN-15 | GWr | 1.7 | 9.8 | 108 | 54.5 | 8.04 | <1 | 7.33 | 14 | 10 | 12 | 76.6 | 0.65 |
| SMN-16 | SW | 0 | 7.5 | 301 | 148 | 45.83 | 1 | 21 | 19 | 7 | 82 | 242.7 | <0.01 |

*GW: Groundwater
SW: Surface water

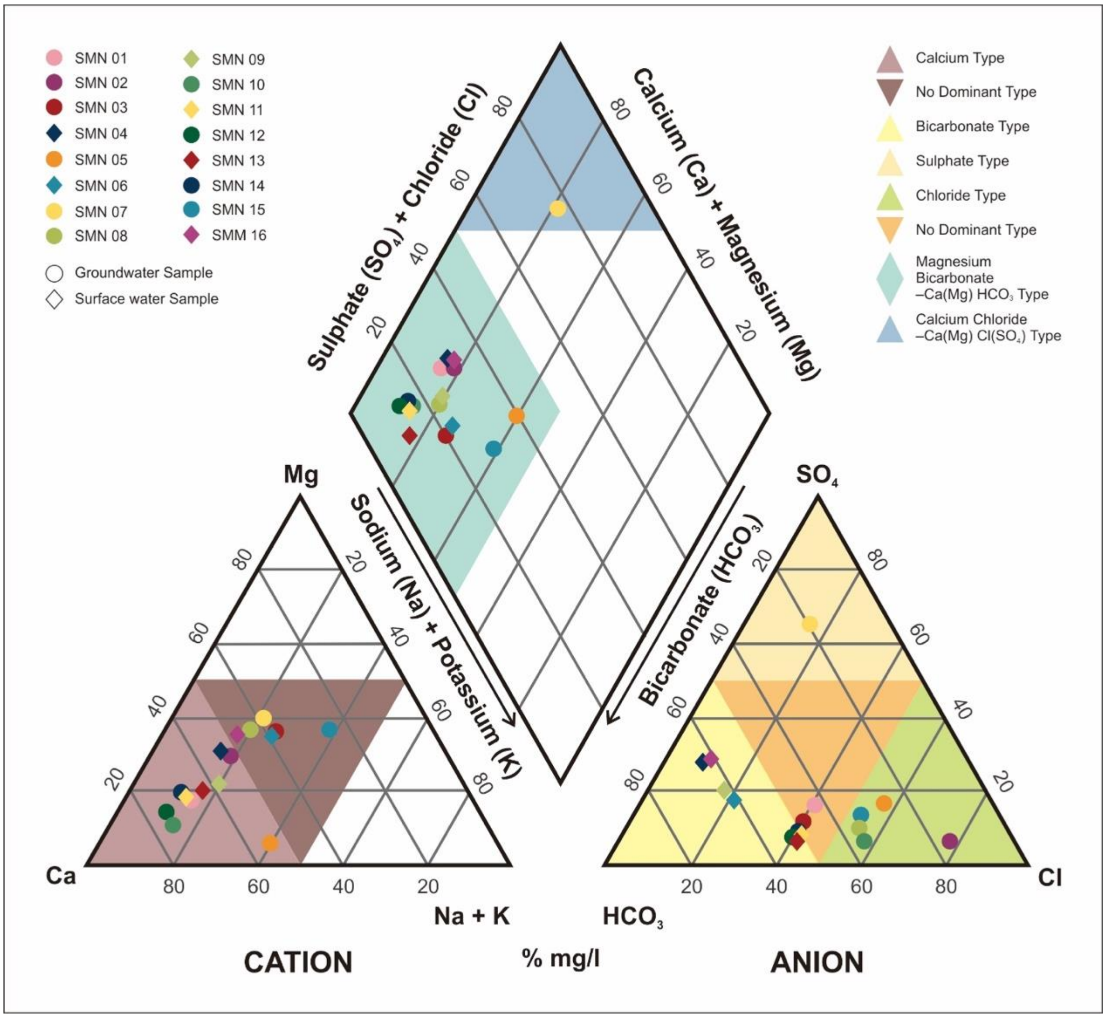


**Fig.3.** Piper's trilinear diagram for water samples in the study area

## 5. Conclusions

All water samples are classified as $CaHCO_3$ hydrochemical facies, except SMN-07 is classified as CaCl hydrochemical facies. The $Ca^{2+}$ ion is assumed to arise from the limestone dissolution of the Wonosari-Punung Formation in the $CaHCO_3$ hydrochemical facies, whereas the $HCO_3^-$ ion is thought to derive from the dissolution of calcite or aragonite, which is usually abundant in limestone. Meanwhile, the $Cl^-$ ion in one CaCl hydrochemical facies sample could come from a variety of sources, including weathering of soil and rocks, industrial and home waste discharge, seawater intrusion, wastewater discharge, rainfall, and irrigation return flow. The surface water quality in the study area is suitable for irrigation. However, the quality of groundwater in the study area mostly acceptable for drinking water, there are several locations that do not accept the standards for drinking water quality, especially pH and TDS. It is essential for the government to carry out treatment to improve drinking water quality and make policies related to water in the mining area and groundwater management so that mining activities do not pollute the water around the study area.

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