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Characterization of Water Pollution in Tropical Epikarst Spring in Gunungsewu Karst Area, Java Island, Indonesia

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Abstract

Groundwater vulnerability in karst areas is generally considered high due to the development of secondary porosity caused by the dissolution processes. However, several epikarst springs still exhibit a large influence of diffuse flow with underdeveloped conduit flow.. This study aims to characterize pollution in epikarst springs located in the tropical region. Water quality standard is used in this study to assess the water pollution in the Gedong epikarst spring. Analysis of dominant major ions, hydrogeochemical facies, dominant hydrogeochemical processes, as well as analysis of hydrogeochemical processes occurring in groundwater (including pollution) were analyzed by Schoeller diagrams, Piper diagrams, and scatter. The characterization results displays that the water quality of Gedong Spring has HCO_3^- and Ca^{2+} values that exceed WHO standards. Analysis of Schoeller diagrams and Piper diagrams on this spring shows that these two dominant major elements are produced from the enrichment process due to groundwater processes in the limestone aquifers of the Wonosari Formation. This is also supported by the results of Scatter plot which shows that the dominant ionic Gedong Spring comes from limestone dissolution. Further analysis showed an influence of agricultural activity is present in 1 of 25 samples taken from the epikarst. The analysis outcomes exhibit a unique characteristic of epikarst springs, namely the dominance of diffuse flow moving through the rock matrix which causes high HCO_3^- and Ca^{2+} content throughout most of the season. On the other hand, this type of flow makes it difficult for pollutants caused by anthropogenic activities in the form of agriculture and domestic use to pollute the water inside. This study presents the pollution characteristics of an epikarst spring in a tropical region with bi-seasonal characteristics, analyzed multitemporally throughout the year to highlight the contrasting effects of wet and dry seasons.

1. Introduction

Karst aquifers are aquifers that play a major role in providing clean water worldwide [1,2]. However, several previous studies have stated that many karst aquifers worldwide are experiencing pollution [35]. The most common pollution in karst aquifers comes from agricultural and domestic activities in settlements [6, 7]. This can be seen from the high nitrate, sulfate, phosphate, chloride, and fecal colli parameter, which the main product of pollution in many karst springs [8, 9].

Karst aquifers are highly vulnerable compared to other aquifers type (porous aquifer) due to several flow types, namely the conduit, diffuse, and fissure flow [10]. Conduit and fissure flow are the most vulnerable to pollution [11]. Conduit flow is a flow that enters from the earth surface directly into the karst aquifer through a tunnel or ponor with a minimum diameter of 0.3 m, so pollutants will enter the underground river system unfiltered and respond very quickly to rainfall, while the fissure flow that causes moderate pollution potential in small fractures with diameter < 0.3 m [12]. The diffuse flow has the lowest vulnerable since it flows through the voids between grains, resulting in a relatively slow flow with a deliberate process [13]. Diffuse systems are often found in areas that still have epikarst zones. Epikarst zone is a zone where water is stored, saturated, and drained and consists of soil, bedrock, and regolith [14]. Although the cracks that develop in the epikarst zone increase permeability, the presence of soil with a predominance of clay texture can also cause natural filtration by voids between rock grains to occur properly [15].

Pollutants in karst aquifers can come from various sources, such as agricultural areas and settlements. Pollution from agricultural waste comes from the residuals of organic and inorganic fertilizers carried by rainwater to karst aquifers [16, 17]. Pollution from residential waste originating from anthropogenic waste generally comes from septic waste from leaky sewers (blackwater) [18, 19] and from materials used for washing activities such as detergents (greywater). Several studies of karst aquifers pollution in the world show that most of the pollution from these two sources typically demonstrate the exceeding value to the water quality standards [20-22]. Even so, the main pollutant materials in karst aquifers by anthropogenic activities in the world are still dominated by agricultural activities. In Indonesia, the water quality standard is regulated by the Indonesian Government Regulation (PP) 22 of 2021 [23], which specifies a maximum pollutant limit that is even stricter than the limit set by The World Health Organization (WHO) [24]. The pollutant limit value in waters that is more rigid in this regulation is expected to reduce the impact on the health of Indonesian people.

Research on water quality is not only related to determining whether a water resource can be utilized for certain purposes [25]. Moreover, water quality analysis is also an evaluation of the condition of karst drainage basins as water catchment areas [26], an early warning system related to activities that cause pollution and land damage [10], and an evaluation of a land treatment in a karst drainage basin in the context of comprehensive environmental management [27]. The results of water quality analysis can also be the basis for evaluating environmental management that has been carried out in the karst drainage basin [28].

Research on pollution in epikarst springs worldwide is still remarkably rare since epikarst springs rarely get attention in hydrogeological studies. This is due to the focus of pollution research generally being carried out on large springs with a conduit flow type and comprises developed karst [29, 30]. The same pattern of pollution research in karst aquifers also occurs in Indonesia, namely in large springs and underground rivers. This is reflected in a considerable amount of research conducted in the Gunungsewu Karst Area, Java Island, that is dominated by research on highly developed karst in the Ponjong and Wonosari Hydrogeological Subsystem. The researches were mainly carried out in the Bribin Karst Drainage Basin, dominated by conduit flows in underground rivers.

The main reason why epikarst springs are rarely studied for their potential pollution is due to their relatively smaller discharge compared to underground rivers, and their utilization is relatively more limited, which only covers a narrow service area. Even so, some epikarst springs such as the one in the Ponjong Hydrogeological Subsystem have a very large role in meeting the needs for clean water and agricultural irrigation. The results of studies on pollution by agricultural and anthropogenic activities in the Gunungsewu Karst show that pollution has occurred in areas that are developing into underground rivers [31] and in karst areas that are recharged by allogenic rivers [32-35].

Studies related to pollution in epikarst springs in the Gunungsewu Karst Area have been carried out by several researchers, although many of them only conducted temporary sampling (no multi-temporal sampling). Several studies have stated that agricultural and domestic activities have polluted epikarst springs in the western part of the Gunungsewu Karst Area [36, 37]. Different outcomes are observed in the eastern part of the Gunungsewu karst area where pollution by agricultural and domestic activities has not occurred, but there are indications of the effect of these two activities on water quality at the study site [38]. This study is to characterize pollution in epikarst springs dominated by diffuse flow types and is expected to fill in the gaps related to knowledge of pollution patterns that occur in karst areas, especially in undeveloped hydrological systems. The temporal variation analysis conducted in this study (sampling for one year every two weeks) also allows the results of this study to be a model for the pollution characteristics of epikarst springs in the tropics which have very unique climatic conditions. The study area is part of the island of Java which is the center of the Asia-Australia monsoon with relatively contrasting rainfall characteristics in the rainy and dry seasons [39].

One interesting location for pollution research is Gedong Spring, which is an epikarst spring in the Ponjong Hydrogeological Subsystem, Gunungsewu Karst Area, Java Island, Indonesia. Gedong Spring is the main source of water used by residents to meet their needs for clean water and agricultural irrigation. Gedong Spring belongs to undeveloped karst, so the dominant aquifer system of this spring is characterized by the diffuse aquifer system. Land use around the Gedong Spring is dominated by agricultural areas and settlements, while the rock material found around the Gedong Spring area is generally in the form of reef limestone originating from the Wonosari Formation [40, 41]. Gedong spring do not have connectivity with ponds, caves, and underground rivers, so it can be said that the recharge of this spring comes purely from rainwater (autogenic). This rainwater recharge enters the epikarst layer and then flows through a diffuse system slowly into the groundwater zone which then appears in the Gedong Spring due to the existence of the Sumbergiri Fault, which cuts the groundwater table. This research aims to characterize pollution in tropical epikarst springs on a multitemporal basis with a case study in the Ponjong Hydrogeological Subsystem which is part of the Gunungsewu Karst Area, Java Island, Indonesia. This study presents the pollution characteristics of Gedong Spring in the Gunungsewu Karst Area, with a multitemporal analysis throughout the year highlighting the contrasting effects of the wet and dry seasons.

2. Study Area

This study was conducted in the North-Central part of the Gunungsewu Karst Area, Java Island, Indonesia (**Figure 1**). The Gunungsewu Karst area physiographically occupies the southern mountains of central part of Java Island. This area stretches along 85 km in the west-east direction with a width (north-south direction) of 12-40 km. The Gunungsewu Karst Area is about 1,300 km^2, covering three provinces namely Yogyakarta Special Region, Central Java Province and East Java Province. This area is part of the Gunungsewu Unesco Global Geopark.

The Gunungsewu Karst Area is formed in the Wonosari Formation, which is composed of reef limestone, especially in the southern part, and bedded limestone, especially in the northern part, which borders the Wonosari Basin. This formation was formed in the Middle Miocene to Late Miocene with bedrock in the form of volcanic rocks formed from the volcanic period in the Late Eocene to Early Miocene.

The Gunungsewu Karst area has a rainfall of 909 – 3,885 mm/year with an average rainfall of 1,883 mm/year. This location is part of the Indonesian Maritime Continent which is influenced by many phenomena such as Monsoon, El Nino-Southern Oscillation, and Madden Julian Oscillation. In fact, the Gunungsewu Karst Area is referred to as part of Java Island which is the center of the Asia-Australia Monsoon, so the influence of this Monsoon is very strong and produces very contrasting wet and dry season conditions. This condition certainly affect the condition of water resources in the study location [42, 43].

3. Method

This research was conducted for one year which covered 2 seasons, namely the rainy season and the dry season as part of an effort to understand the temporal characteristics of the research location. The tools used in this study were a water quality test kit, 1-liter sample bottles, and an alkalinity test kit. The water quality test kit is used to measure physical parameters in the field such as pH, temperature, total dissolved solid (TDS), and conductivity (EC). A 1-liter sample bottle is used to take samples of Gedong spring water so that the quality of the water sample is maintained for laboratory checking, while the alkalinity test kit is used to test the Bicarbonate (HCO_3^-) parameter in the field.

The first stage of the research consisted of collecting data from direct measurements in the field and taking water samples. The second stage of this research was laboratory testing of Gedong Spring water samples. Water samples were taken from February 2020 to January 2021 every two weeks, with a total of 25 water samples taken. The water samples were then tested in the laboratory with major ion parameters consisting of Calcium (Ca^{2+}), Sodium (Na^+), Magnesium (Mg^{2+}), Potassium (K^+), Sulfate (SO_4^{2-}), and Chloride (Cl^-). The major element analysis method used in this study is presented in detail in **Table 1**.

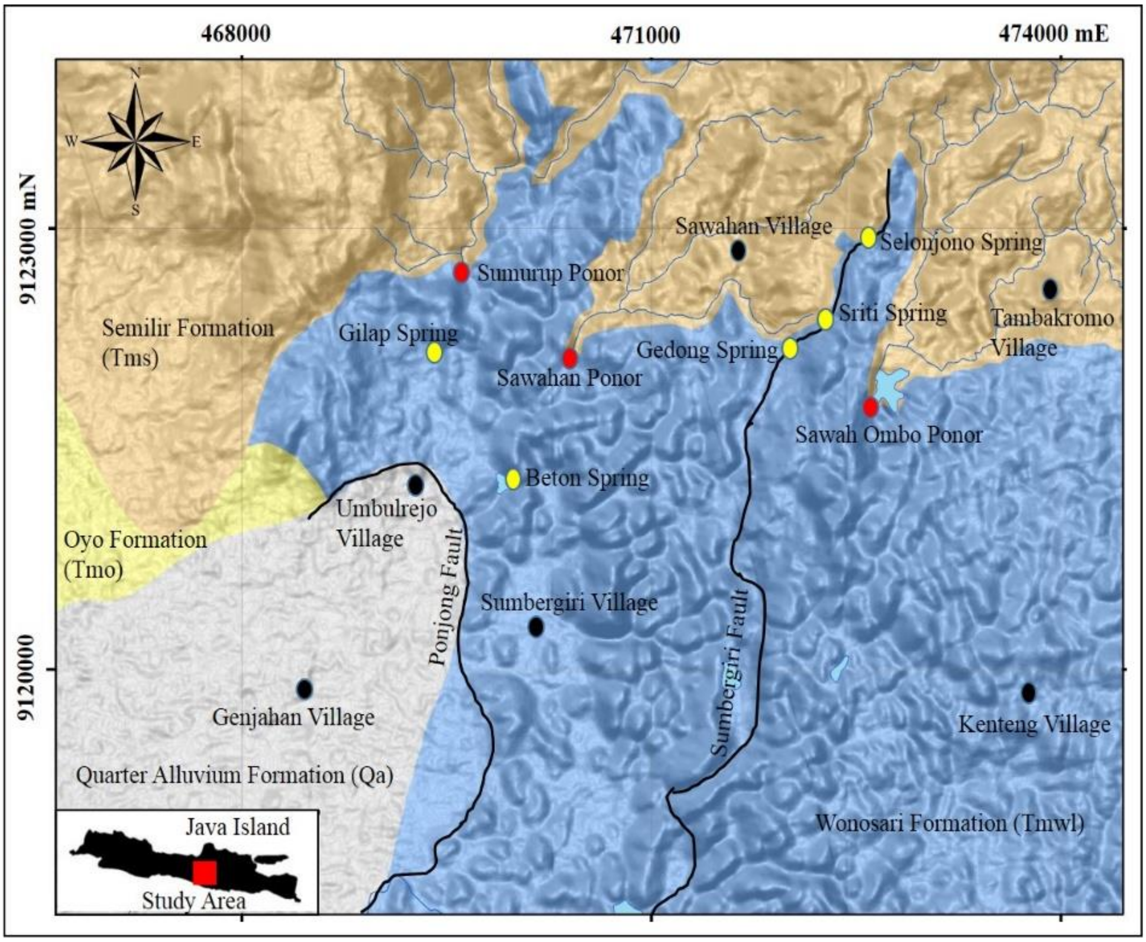

Figure 1. Research Area (Red dots are ponors, blue dots are springs and black dots are villages)

Sources: Topography Map scale 1:25,000, Geological Map Scale 1:100,000, DEMNAS Geospatial Information Agency of Republic Indonesia and Field Survey

Table 1. Major element analysis method used in this research

No	Parameter	Unit	Method Standard	Name of Method
1	Na+	mg/L	APHA 2017, Section 3500-Na	Flame Emission Photometric Method
2	K+	mg/L	APHA 2017, Section 3500-K	Flame Emission Photometric Method
3	Ca^{2+}	mg/L	SNI* 06-69889.12-2004	Titimetry
4	Mg^{2+}	mg/L	SNI 06-69889.12-2004	Titimetry
5	Cl^-	mg/L	SNI 6989.19-2019	Argentometry
6	SO_4^-	mg/L	SNI 6989.20-2019	Turbidimetry

Note: SNI stands for Indonesian National Standard [44, 45] and APHA [46]

The third stage is the outcome of field measurements and analysis of laboratory results. The first analysis was carried out by comparing the results of each parameter with the water quality standards of WHO 2012 [24] and Government Regulation (PP) of the Republic of Indonesia Number 22 of 2021 [23], which is the latest regulation in Indonesia that contains standard water quality standards in Indonesia. The second analysis was carried out to determine the chemical characteristics of Gedong Spring using Piper diagrams and Schoeller diagrams. These two diagrams will be very helpful in explaining the hydrogeochemical facies, the dominant ions present in the water sample and the dominant hydrogeochemical processes, making it easier to identify pollution.The third analysis in the form of an analysis of the effect of anthropogenic pollution is carried out by plotting each ion according to equations (1 – 5) scatter plots [47-50].

$Ca^{2+} + Mg^{2+}$ vs $HCO_3^- + SO_4^{2-}$ ....(1)

Na^+ vs HCO_3^- ....(2)

$Cl^- - SO_4^{2-}$ vs Na^+..............................(3)

Na^+ versus Cl^-..............................(4)

Total Anion – SO_4^{2-} vs SO_4^{2-}..............................(5)

4. Results and Discussion

The results of the analysis of 25 Gedong Spring water samples displayed that several parameters exceeded the quality standards, namely the parameters Ca^{2+}, Mg^{2+}, and HCO_3^- (**Table 2**). Ca^{2+} parameter values exceeding the quality standard (>100 mg/L) were found in 5 samples. High Ca^{2+} values in karst areas are quite common due to the dissolution process in limestone, although the value does not always exceed the quality standard. The average Ca^{2+} value in Gedong Spring is relatively high (103.77 mg/l) when compared to the content in the Seropan underground river (92.14 mg/l) [51] and springs recharged by allogenic rivers, such as Gremeng Spring (47.47 mg/l). This value hints the influence of diffuse flow in Gedong Spring is still significantly dominant, ensuing in the absence of dilution process from rain, conduit systems, and recharge by allogenic rivers in this epikarst spring. In addition, the Ca^{2+} value in Gedong Spring is also relatively higher than that of other epikarst springs such as Selonjono Spring and Guntur Spring. This condition is probably due to the development of the younger tunnel in this spring which has a larger thickness of limestone layers (±125 m) compared to the other two epikarst springs. It is known that even though it is located on the same fault plane, the elevation of Gedong Spring is lower (450 masl) compared to Selonjono Spring (500 masl), so the limestone layer above it is relatively thicker. Meanwhile, the thickness of the Guntur Spring layer is around 72 m.

There is only 1 sample that exceeds the quality standard in the Mg^{2+} parameter with a value of 52 mg/L, with the range of Mg^{2+} parameter values generally being between 4-29 mg/L. The overall value of Mg^{2+} in Gedong Spring is higher value (13.40 mg/l) compared to the Seropan Underground River (9.77 mg/l) and Gremeng Spring which are fed by allogenic rivers (9.24 mg/l). This value is more or less the same as the average value of other epikarst springs, namely Selonjono Spring (14.19 mg/l) and Guntur Spring (12.35 mg/l).

The overall parameters of HCO_3^- exceed the quality standard with a value of more than 350 mg/L. The high HCO_3^- value is caused by the high limestone dissolution process in the karst area at the study site. The HCO_3^- parameter has the same pattern as the Ca^{2+}, which is higher than that of underground rivers or springs fed by allogenic rivers. The average HCO_3^- value for Gedong Spring was 430.04 mg/l, while the average value for the underground river was 183.26 mg/l, and the average value for Gremeng Springs was only 177.89 mg/l. This value is higher compared to other epikarst springs, such as Guntur Spring (347.70 mg/l) and Selonjono Spring (386.60 mg/l). The HCO_3^- parameter at the study site has an increasing trend during the dry season, marked by the number of samples that have a value of more than 400 mg/l from March to November 2020. This condition characterizes flow dominated by diffuse flow, where the water flowing from the spring has spent a long time in the limestone aquifer and has interacted with the rock for an extended period (water-rock interaction), resulting in high HCO_3^- content.

The parameter values of Na^+, K^+, Cl^-, and SO_4^{2-} are known to have values below the water quality standards with average values of 7.32 mg/l, 1.12 mg/l, 4.51 mg/l, and 7.36 mg /l. Overall pH parameter values are within the quality standard with values ranging from 6.26 to 7.91. The EC parameter has a value ranging from 510-650 µs/cm, marking that all samples belong to the freshwater classification. The TDS parameter of the samples also displays values that are below water quality standards, ranging from 260-350 ppm. The temperature parameter shows that the average water sample has a value ranging from 26-27 °C.

Table 2. Major element analysis method used in this research

No	Date	Major Ion (mg/l)							pH	EC (µs/cm)	TDS (ppm)	Temp. (°C)
		Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	Cl^{-}	HCO_3^{-}	SO_4^{2-}				
1	15/02/2020	91,54	20,31	5	1	3	**390,5**	1	7,4	510	260	26,5
2	07/03/2020	**110**	18,47	4	1	3,5	**396,6**	8	7,05	561	279	26,7
3	14/03/2020	**104**	9,09	10	1	3	**402,7**	7	7,48	557	279	26,1
4	28/03/2020	**103**	14,99	3	1	2,5	**402,7**	7	7,06	562	281	26,4
5	10/04/2020	97,6	11,18	6	1	3	**396,6**	7	7,91	546	274	26,9
6	25/04/2020	85,97	**51,74**	9	1	3	**366,1**	6	7,25	544	272	26,5
7	09/05/2020	68,46	14,02	6	1	2,5	**378,3**	6	7,22	538	269	26,3
8	23/05/2020	**103**	16,96	10	1	3	**353,9**	6	7,08	536	268	26,9
9	06/06/2020	91,2	18,46	7	1	2,5	**408,8**	8	7,41	538	325	26,2
10	20/06/2020	98,7	17,41	11	1	2,5	**402,7**	6	7,21	520	280	27,1
11	04/07/2020	88,8	5,35	11	1	3	**366,1**	5	6,96	520	280	26
12	19/07/2020	**102**	9,23	9	1	3,5	**353,9**	15	7,22	520	280	26,8
13	01/08/2020	99,5	11,61	7	1	2,5	**396,6**	2	7,06	530	280	25,7
14	13/08/2020	**102**	13,68	5	1	2,5	**384,4**	9	6,87	520	280	28,1
15	30/08/2020	100,8	14,09	6	1	3	**378,3**	6	7,37	520	280	27,6
16	18/09/2020	99,7	8,79	5	1	2,5	**457,6**	6	6,49	530	280	26,4
17	26/09/2020	68,8	11,66	7	1	2,5	**360,0**	6	6,86	520	280	27,2
18	10/10/2020	98,7	12,57	3	1	3,4	**378,3**	6	6,86	520	280	26,8
19	25/10/2020	98,4	4,37	3	1	20,3	**372,2**	5	7,34	520	280	26,6
20	07/11/2020	28,67	18,4	6	1	3	**384,4**	7	6,45	540	290	28,4
21	24/11/2020	**103**	6,38	8	1	2,5	**408,8**	6	7,18	530	280	26,5
22	07/12/2020	70,75	0,49	3	1	2,5	**396,6**	6	7,74	560	300	26,4
23	21/12/2020	92,93	29,45	9	1	2,5	**372,2**	6	6,28	650	350	26,4
24	02/01/2021	51,74	1,45	3	1	2	**390,5**	6	7,09	550	300	26,6
25	16/01/2021	100	14,58	3	2	4	**366,1**	6	7,08	540	290	26,2
Permenkes 2021		-	-	-	-	300	-	300	(6-9)	-	1000	-
WHO 2012		100	50	200	20	250	350	250	(6-9)	-	500	-

Sources: Field measurements and laboratory analysis in 2020-2021
Note: Bolded numbers exceed the quality standard

The Schoeller diagram analysis shows that the dominant ions at the study site are Ca^{2+} and HCO_3^{-} (**Figure 2**). This is caused by limestone bedrock at the study site from the Wonosari Formation with an abundance of these two dominant ions, enriching the groundwater at that location with these two ion contents. The same Schoeller diagram pattern is also found in all karst springs in Gunungsewu [38, 51], Jonggrangan Karst [52], and Karangbolong Karst [53] which has similar bedrock and is also situated in the southern mountains of Central Java. *Eiche et al.* [54] explained that the hydrogeochemical characteristics of the springs in the Gunungsewu Karst Area are mineralized with the addition of ions, especially Ca^{2+} and HCO_3^{-} which tend to be high in the dry season and decrease in the rainy season due to the dilution process. The dominant diffuse flow process accompanied by a long residence time in the aquifer will cause the dominant ion variations in the flow to be very difficult to replace or vary [31, 55].

The results of the Piper diagram analysis (**Figure 3**) also support the Schoeller diagram analysis by showing that the Gedong Spring has the dominant cation Ca^{2+} and is included in type A (Calcium Type), with the dominant anion in the form of HCO_3^{-} which included in Type E (Bicarbonate Type). The results of the Piper diagram analysis of the overall classification of the dominant ion shows that the samples at the study site fall into Type I (Ca + Mg Type) and J (HCO_3 type). Both the Schoeller and the Piper diagrams confirm the presence of magnesium enrichment at the study site. This can be seen from the samples that are classified as D (No Dominant Type) on the Piper diagram and the high concentration of $Mg^{2+,}$ which is almost the same as Ca^{2+} on the Schoeller diagram. This development is similar to the results of a study conducted by *Maizar and Hastuti* [56], which stated that there was an enrichment of Mg^{2+} in aquifers in the Gunungsewu Karst Area due to the interaction of groundwater with the rocks of the Wonosari Formation.

The results of the Piper diagram analysis presented in **Figure 4** show that Gedong Spring are included in Type C (Calcium Enrichment), Type G (Unpolluted Groundwater), and Type D (Unpolluted Groundwater). The type of calcium enrichment is a pattern that is very common for karst aquifers, which are mostly composed of calcium. This pattern is also found in other sources in Gunungsewu, namely Pindul Cave [33], Gremeng and Beton Spring [32], springs in the Karangbolong Karst Area [53], and karst springs in Jonggrangan Karst Area

[51]. Based on the Piper diagram analysis, the Gedong Spring is included in the Unpolluted Groundwater category, which exhibits that the water quality at the study site is classified as good based on the analysis of major ion elements.

The high content of Ca^{2+}, Mg^{2+}, and HCO_3^- at the study site induces the absence of anthropogenic characteristic major ions. The main disadvantage of Piper diagram analysis, according to Hodgson's modification [57], is the inability to identify processes that arise from minor elements. This was also found in the pollution analysis conducted in the Seropan Underground River [51] and Selonjono Springs [39]. Although based on the analysis, it was observed that the main process occurring at the study site was Ca^{2+} enrichment, contamination by other processes may also occur considering that anthropogenic major ions characteristics cannot be observed in the Piper diagram.

Sample plot analysis for Ca^{2+} + Mg^{2+} versus HCO_3^- + SO_4^{2-} using a scatter diagram displays values that incline to the 1:1 line (**Figure 5**). This result pinpoint that the ions contained in groundwater originate from the carbonate rock dissolution [46], which also reinforces the previous results on the Schoeller diagram and Piper diagram. **Figure 5** also shows that there are three samples above the 1:1 line and one sample far below the 1:1 line. Three samples that are located far above the 1:1 line show a drastic decrease in Ca^{2+} + Mg^{2+} values on November 7th, 2020, December 7th, 2020, and January 2nd, 2021, while the presence of one sample far below the 1:1 line occurs due to an increase in Mg^{2+} drastically on April 25th, 2021. This is also what causes the Mg^{2+} parameter in the sample to exceed the water quality standard.

The results of the scatter plot analysis for the content of HCO_3^- versus Na^+ also strengthen the results of previous analysis. The results of the HCO_3^- versus Na^+ scatter plot analysis display that all Gedong Spring samples are away from the 1:1 line (**Figure 6**). This result indicate that the chemical composition of the water that makes up the Gedong Spring does not originate from the silicate dissolving process [58]. *Yuan et al.* [59] added that the value of HCO_3^- versus Na^+ which is far from 1:1 signal that the aquifer at the study site is located quite far from sources of silicate rocks such as volcanic rocks. This shows that although the limestone of the Wonosari Formation at the study site is underlain by ancient volcanic rocks of the Semilir Formation [60, 61], the groundwater that supplies the Gedong Spring only comes from the top layer of the aquifer.

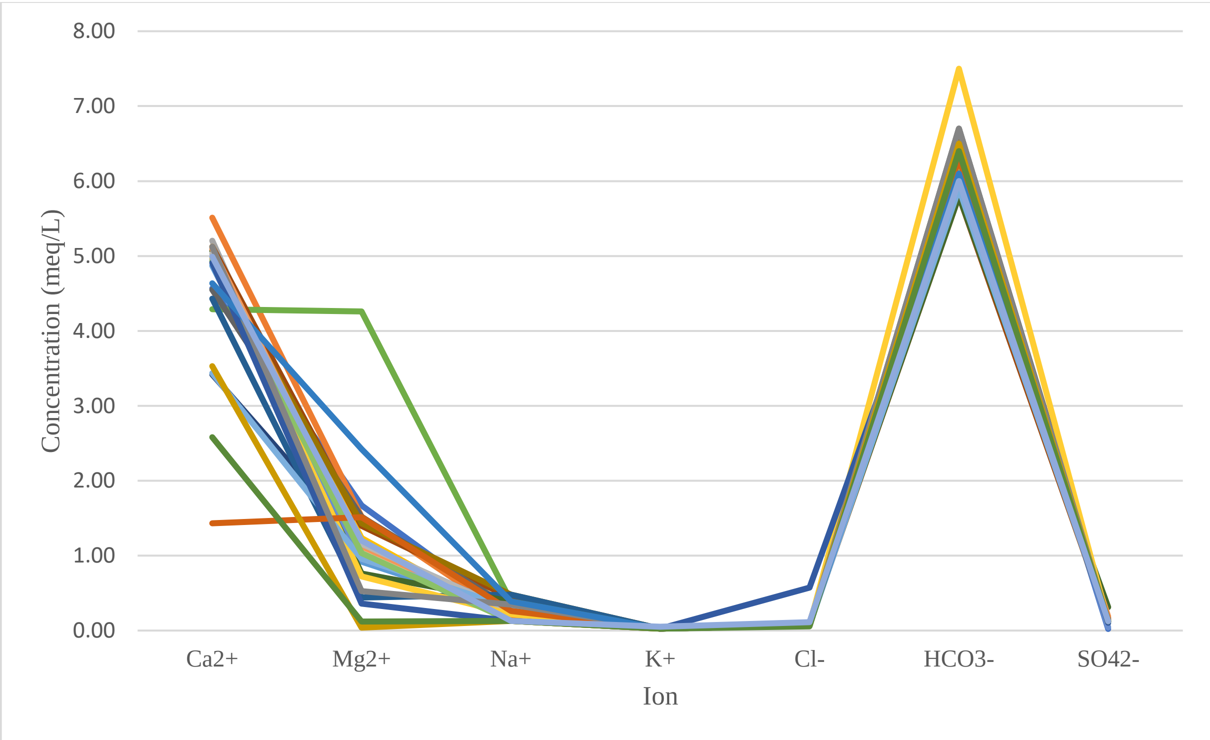

Figure 2. Schoeller diagram of the Gedong Spring

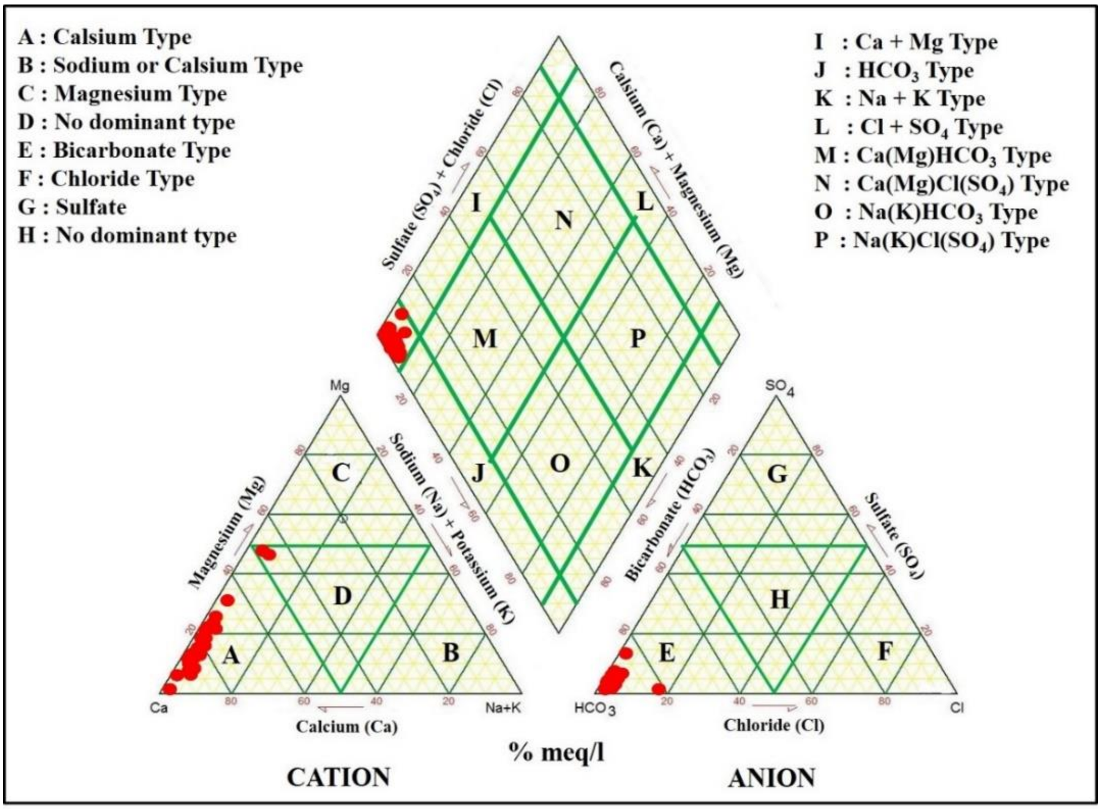

Figure 3. Piper diagram showing the hydrogeochemical facies (type) of Gedong Spring

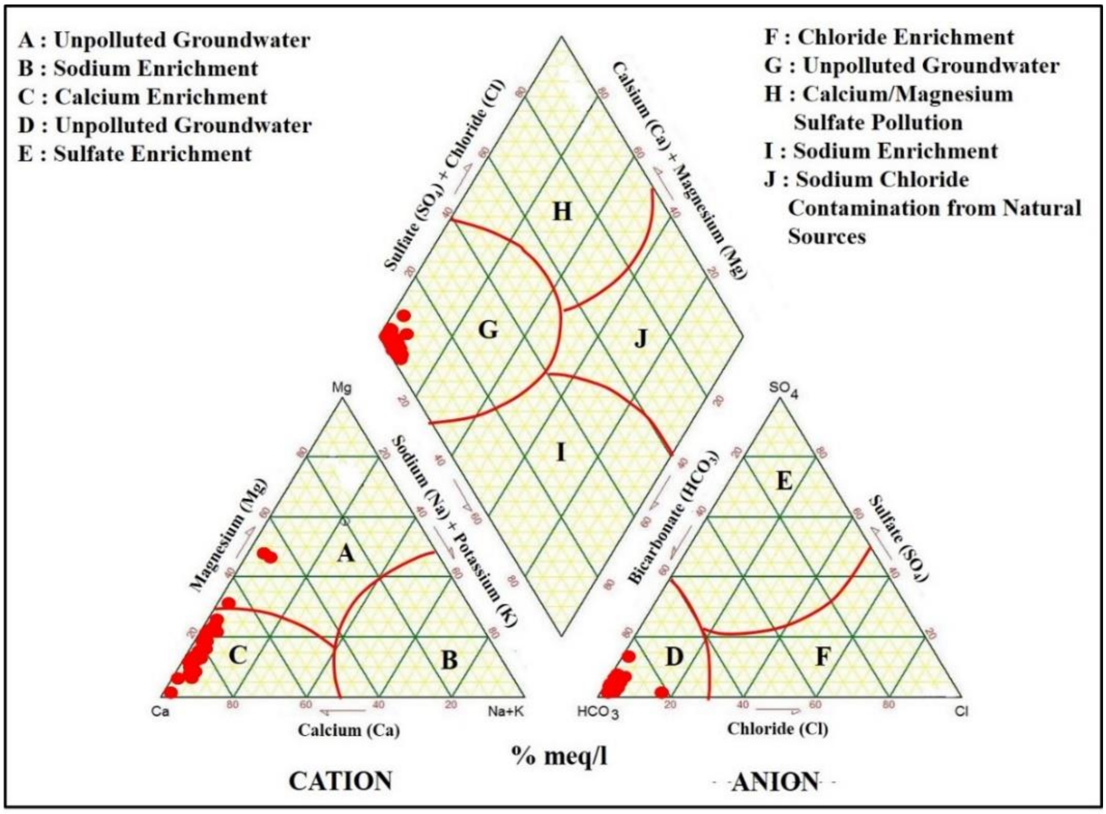

Figure 4. Piper diagram showing the dominant processes occurring in the Gedong Spring

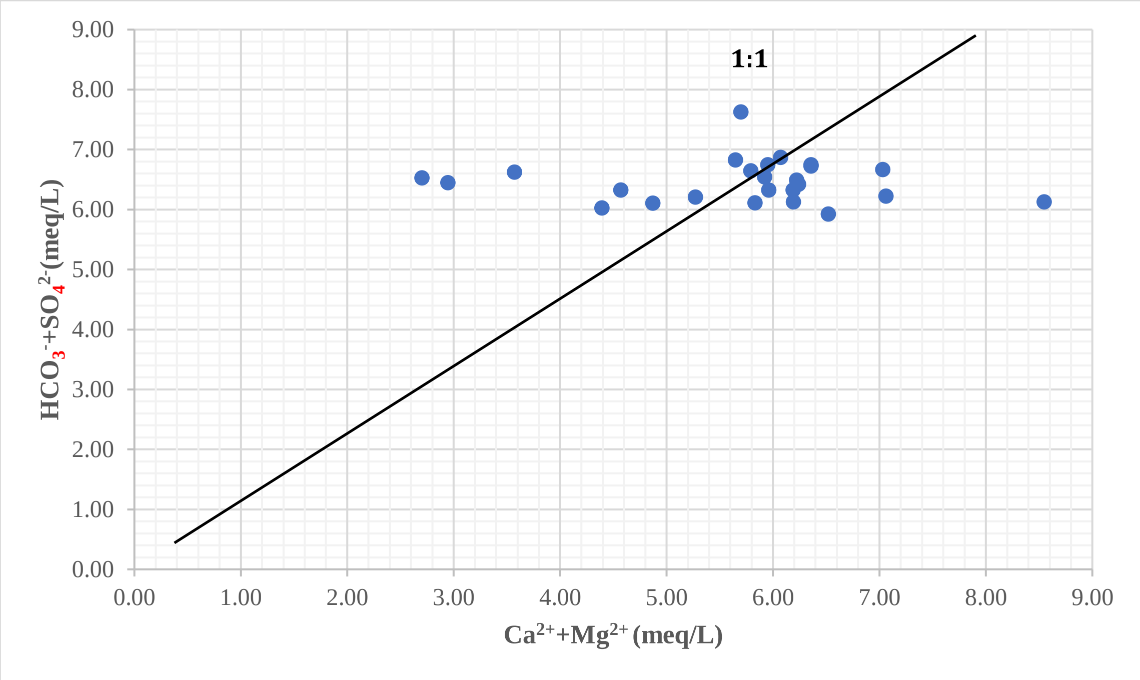

Figure 5. Scatter plot parameter of Ca^{2+} + Mg^{2+} versus HCO_3^- + SO_4^{2-} in the Gedong Spring

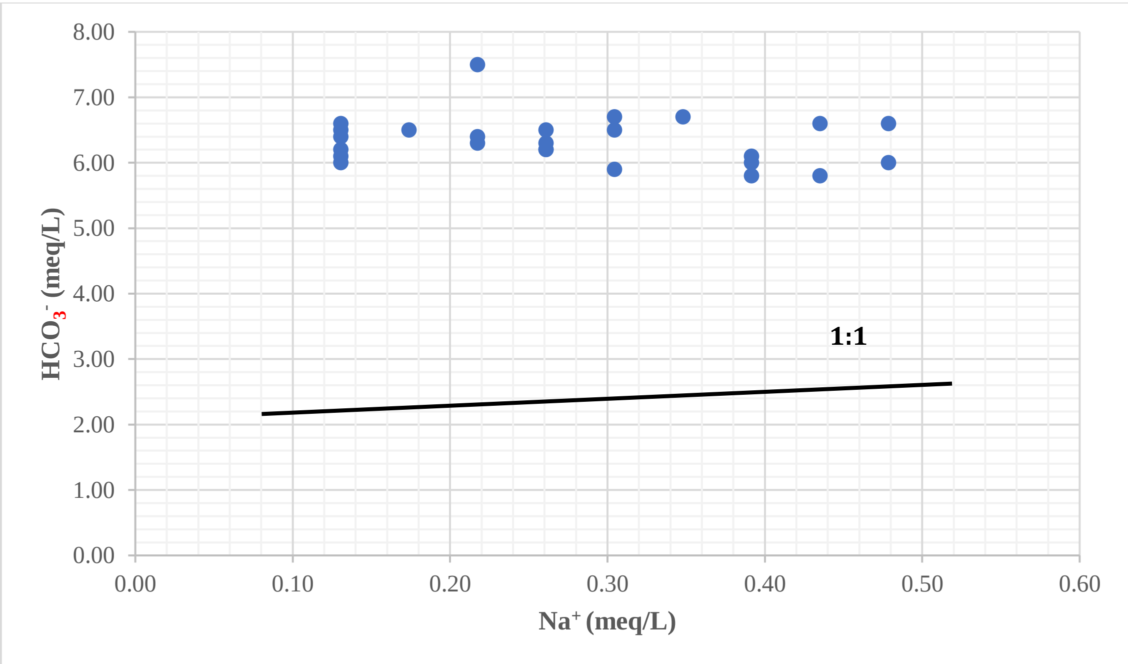

Figure 6. Scatter plot parameter of Na^+ versus HCO_3^- in the Gedong Spring

The scatter plot of Cl^- - SO_4^{2-} data against Na^+ on the scatter diagram shows that the Gedong Spring sample deviates from the 1:1 line. (**Figure 7**). This result imply that the main process affecting the chemistry of groundwater in Gedong Spring does not originate from dissolving sodium/sodium sulfate and halite [46]. This also shows that the faults formed on the Sumbergiri Fault did not lift the rocks in the Semilir Formation up to the Gedong Spring. In addition, this also reinforces the previous analysis which exhibits that the carbonate dissolution process is the most dominant process affecting the chemistry of groundwater in the study area.

The scatter plot of of Cl^- versus Na^+ samples on the scatter diagram displays that almost all of the Gedong Spring samples are below the 1:1 line (**Figure 8**), and implying a cation exchange process in Gedong Spring. There is only one sample that is above the 1:1 line which indicates anthropogenic pollution as mark by the high Cl^- ion in the October 25th, 2020 sample. The presence of this high Cl^- value is known not to originate from precipitation (rain) because it is not close to line 1: 1 which means the source is different from the Na^+ source [62]. This was also influenced by the time of sampling, since the sampling was carried out in October during the dry season. The presence of high Cl^- also does not come from rocks, especially halite, because previous results show that this process does not occur at the study site. Therefore, the high value of Cl observed in **Figure 8** strengthens the indication of anthropogenic pollution at the study site.

Anthropogenic activities that can increase the Cl^- content in groundwater generally include agricultural and domestic activities [63, 64]. More specifically, the source of Cl^- from anthropogenic activities typically comes from human sewage, livestock waste used as manure in agriculture (organic), and synthetic fertilizer mainly in the form of Potassium Chloride (KCl) [65]. Hence, Cl^- is often used to determine the magnitude of the impact of anthropogenic activities [66, 67] in addition to calculating groundwater recharge.

Plotting samples of Total Anion - SO_4^{2-} versus SO_4^{2-} on the scatter diagram shows a very low correlation value (**Figure 9**). This result demonstrate that the source of the addition of SO_4^{2-} is not from natural hydrogeochemical processes, but from anthropogenic processes. The same pattern is also found in the Selonjono Spring [39] and the Seropan Underground River [51]. The geological conditions of the study area which are known to be quite far from volcanic rocks suggest that the addition of SO_4^{2-} to groundwater does not occur naturally from rocks. This strengthens the indication of the addition of contaminants originating from anthropogenic activities at very small levels which are still far below the water quality standard.

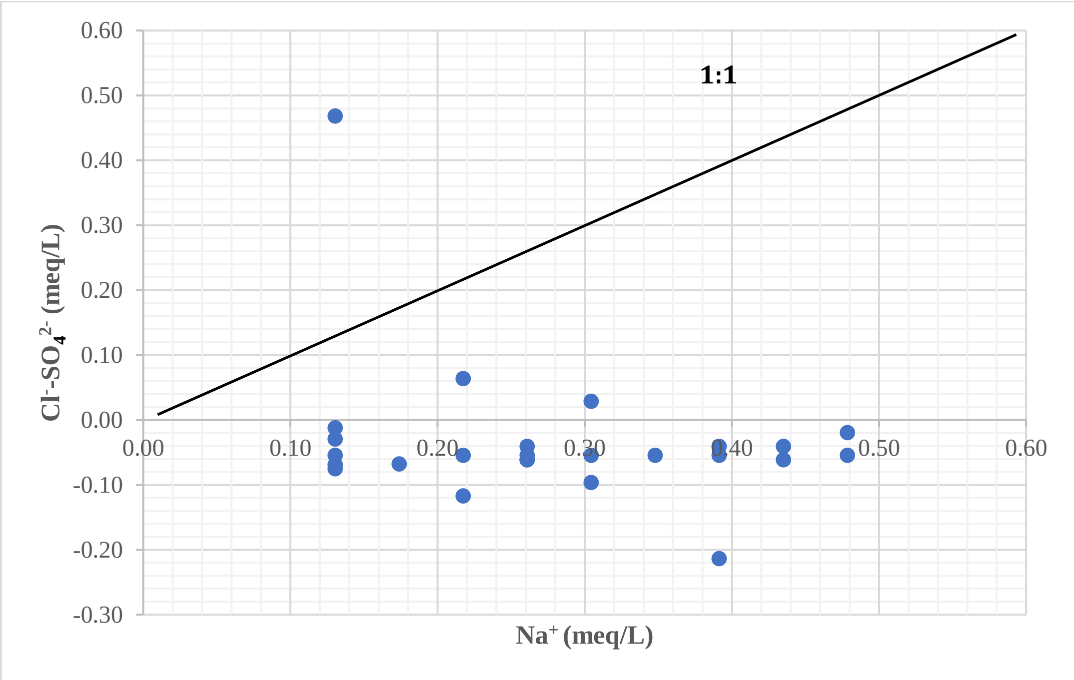

Figure 7. Scatter plot parameter of Cl^- – SO_4^{2-} versus Na^+ in the Gedong Spring

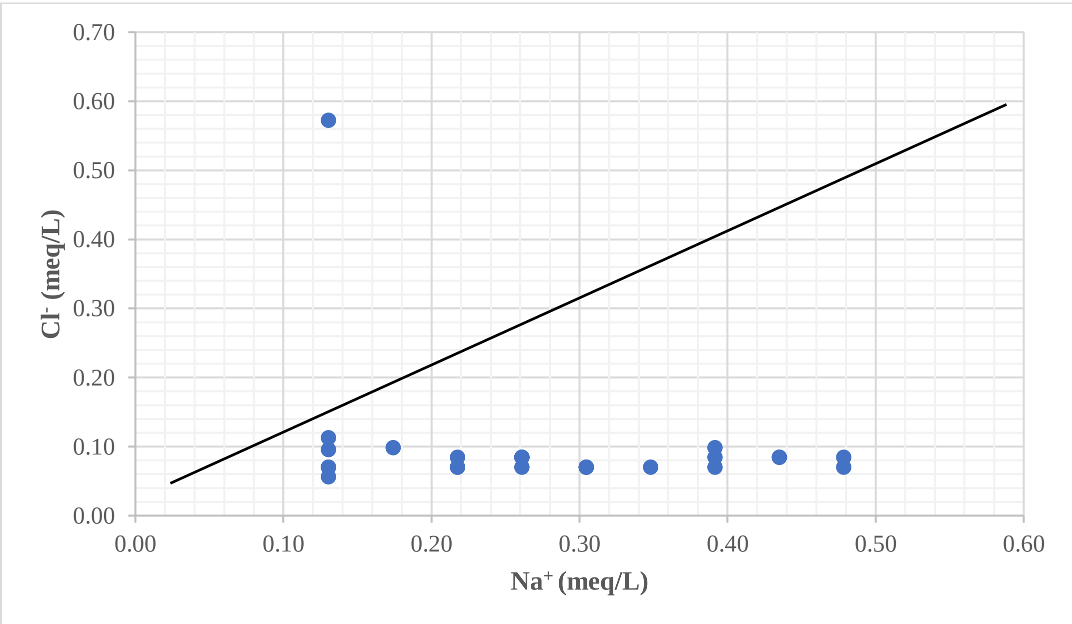

Figure 8. Scatter plot parameter of Na^+ versus Cl^- in the Gedong Spring

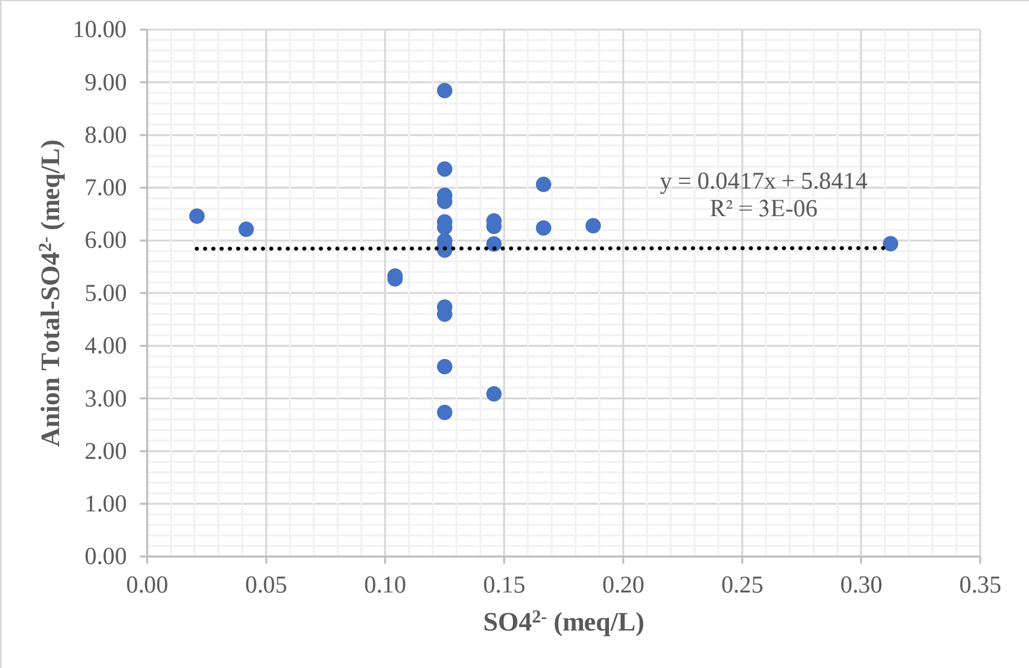

Figure 9. Scatter plot parameter of Total Anion – SO_4^{2-} versus SO_4^{2-} in the Gedong Spring

Based on the findings due to anthropogenic activities pollution is present in Gedong Spring with a very small value. This is proven by the presence of pollution in one sample, yet the quality standards are not exceeded by anthropogenic characterizing elements. Water quality problems in epikarst springs at the study site arise due to the interaction process between water and rock causing the process of dissolving carbonate rock which compels the enrichment of calcium and bicarbonate minerals [68]. This condition can be observed from the high content of these two parameters throughout the year which exceeds the quality standard in most of the year.

Nayono et al. [69] explained that the use of septic tanks that still allow black water waste to seep into the aquifer is a large potential pollutant in the study location. Efforts to manage sanitation properly, either individually in households or communally, are very important [70]. Land degradation, erosion processes and excessive fertilizer usage are such concerns in the management of karst drainage basins in the study area [38, 47, 71]. Therefore, spatial planning and land use are very important in groundwater conservation because they are closely related to sources of pollution and activities that can cause groundwater pollution [72, 73, 74].

Riyanto et al. [38] explained that agricultural land management and spatial planning should be the focus of spring management in the Gunungsewu Karst Area. Furthermore, *Riyanto et al.* [38] recommended several actions such as the creation of structures that prevent pollutants from entering directly through ponors or sinkholes, erosion management and reforestation on karst hills with high slopes, utilization of relatively flat karst cockpits for agriculture supported by multilayer vegetation coverage, regulation and optimization of fertilizer use in agricultural activities and creating protection zones for areas along lineaments, caves and underground rivers. Policies related to zoning or spatial planning are also applied in other regions in the world as stated by *V. Živanović* [75], namely Spring Protection Zone, Inner Protection Zone and Outer Protection Zone. Groundwater protection zone application is also done in India, where the protection zone is divided into four zones that not only consider the buffer zone but also consider geological and geomorphological characteristics such as constituent materials, landform morphoarrangement, lineament density, and land use/cover [76, 77,78].

5. Conclusion

The water quality analysis show that Gedong Spring have HCO_3^-, Mg^{2+} and Ca^{2+} parameters that exceed the quality standards. Schoeller's diagram of Gedong Spring exhibits that the dominant ion pattern in the study area is in the form of bicarbonate (HCO_3^-) while calcium (Ca^{2+}) originats from limestone dissolution. These events are supported by the results of the Piper diagram analysis of the Gedong Spring that fall into Calcium Type, Bicarbonate Type, Ca+Mg type and HCO_3^- type. In addition, the Piper diagram of Gedong Spring displays the events of Calcium Enrichment and Unpolluted Groundwater. Ion comparison analysis with scatter plots shows that the dissolution of carbonate rock is the main process that occurs in groundwater. Indications of pollution by anthropogenic activities have appeared, although it has not exceeded the established water quality standards.

The results also exhibit that even though the study area is located in the karst area, the potential for groundwater contamination in the study area is still fairly small. This happens since epikarst spring have diffuse flow as their dominant flow type which allows natural filtration of water entering the aquifer. However, diffuse flows that have a long residence time in an aquifer may cause excessive mineral enrichment which can exceed the water quality standard.

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Author contributions

Ahmad Cahyadi: Conceptualization, Methodology, data curation, Software, Writing-Original draft preparation **Indra Agus Riyanto:** Data curation, Writing-Original draft preparation, Software, Validation. **Rasyiida Acintya:** Data curation, Writing-Reviewing and Editing, Layouting Manuscript **Ahmad Singgih:** Data curation, Writing-Reviewing and Editing. **Rakhmat Dwi Putra:** Visualization, Mapping, Writing-Reviewing and Editing.

Conflicts of interest

The authors declare no conflicts of interest.

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